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DOE STANDARD

GOOD PRACTICES FOR OCCUPATIONAL RADIOLOGICAL PROTECTION IN PLUTONIUM FACILITIES

U.S. Department of Energy
Washington, D.C. 20585

AREA SAFT

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Foreword

This Technical Standard does not contain any new requirements. Its purpose is to provide information on good practices, update existing reference material, and discuss practical lessons learned relevant to the safe handling of plutonium. U.S. Department of Energy (DOE) health physicists may adapt the recommendations in this Technical Standard to similar situations throughout the DOE complex. The Standard provides information to assist plutonium facilities in complying with Title 10 of the Code of Federal Regulations (CFR), Part 835, Occupational Radiation Protection. The Standard also supplements the DOE 10 CFR 835 Implementation Guide, DOE Orders, and DOE standard, DOE-STD-1098-2008, Radiological Control, (RCS) and has as its sole purpose the protection of workers and the public from the radiological hazards that are inherent in plutonium storage and handling.

This Standard uses the word "shall" to identify a required practice or the minimum acceptable level of performance. The word "should" is used to identify good practices (preferred practices) recommended by this Standard. The word "may" is used to identify permitted practice (neither a requirement nor a recommendation).

This Standard includes provisions in the 2007 amendment to 10 CFR 835. This amendment updated the dosimetric terms and models for assessing radiation doses, both internal and external. Of particular interest for this Standard, the biological transportability of material is now classified in terms of absorption types; F (fast), M (medium) and S (slow). Previously this was classified in terms of material class; D (days), W (weeks) and Y (years). Throughout this Standard, discussions of previous studies describing the biological transportation of material in the body will continue to use D, W and Y, as appropriate. Discussions of other requirements which have not amended their dosimetric terms and models continue to use the older terminology.

This Standard does not include every requirement applicable to every plutonium facility. Individuals responsible for implementing Radiation Protection Programs at plutonium facilities need to be knowledgeable of which requirements (contractual or regulatory) are applicable to their facility.

Copies of electronic files of this Technical Standard may be obtained from either the DOE Radiation Safety Home Page Internet site (http://www.hss.energy.gov/HealthSafety/WSHP/radiation/ts.html) or the DOE Technical Standards Program Internet site (http://www.hss.doe.gov/nuclearsafety/techstds/standard.html).

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U.S. DEPARTMENT OF ENERGY

GOOD PRACTICES FOR OCCUPATIONAL RADIOLOGICAL PROTECTION IN PLUTONIUM FACILITIES

Coordinated and Conducted
for the
Office of Health, Safety and Security
U.S. Department of Energy

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1.0 INTRODUCTION

1.1 PURPOSE AND APPLICABILITY

This Technical Standard (TS) does not contain any new requirements. Its purpose is to provide information on good practices, update existing reference material, and discuss practical lessons learned relevant to the safe handling of plutonium. U.S. Department of Energy (DOE) health physicists may adapt the recommendations from this TS to similar situations throughout the DOE complex. Generally, DOE contractor health physicists will be responsible to implement radiation protection activities at DOE facilities and DOE health physicists will be responsible for oversight of those activities. This guidance is meant to be useful for both efforts. The TS provides information to assist plutonium facilities in complying with Title 10 of the Code of Federal Regulations (CFR), Part 835, Occupational Radiation Protection (DOE, 2007a); hereinafter referred to as 10 CFR 835. The TS also supplements the DOE 10 CFR 835 Implementation Guide, G 441.1-1C, Ch 1 (DOE, 2011a), DOE Orders, and DOE's standard Radiological Control , (DOE, 2008a) and has as its sole purpose the protection of workers and the public from the radiological hazards that are inherent in plutonium storage and handling. This Standard does not include every requirement applicable to every plutonium facility. Individuals responsible for implementing Radiation Protection Programs at plutonium facilities need to be knowledgeable of which requirements (contractual or regulatory) are applicable to their facility.

This TS was originally based upon the data in PNL-6534, Health Physics Manual of Good Practices for Plutonium Facilities (PNL, 1988), which provided information of situations that were typical of DOE's plutonium operations; safe storage, handling and inspection, decontamination, and decommissioning (environmental restoration); and weapons disassembly. This 2013 revision updates the 2008 revision and discusses requirements for DOE accident investigations of particular applicability for plutonium facilities and updates information on chelation therapy.

The technical information presented here represents the best technical information available from within the DOE complex. Except to the extent that the guidance presented here duplicates mandatory regulations or contract requirements, it is not binding or mandatory. Any DOE Orders, manuals or guides, referred to in this TS are not binding unless they have been incorporated into the applicable contract to assist in identifying applicable requirements, “shall” statements are followed by a reference. Should and may statements are provided for consideration. However, judicious use of this TS, along with the regulatory documents discussed above, will help assure a comprehensive and technically defensible radiological protection program.

References are current as of December 2011.

1.2 DEFINITIONS

A glossary is provided (see Appendix) to ensure uniform understanding of words in this document. In all cases, the definitions given here are consistent with those used in the Implementation Guide (DOE, 2011a).

1.3 ACRONYMS

AC	Alternating Current
AMAD	Activity Median Aerodynamic Diameter
ANSI	American National Standards Institute
ALI	Annual Limit on Intake
ALARA	As Low As Reasonably Achievable
BNL	Brookhaven National Laboratory
CDC	Centers for Disease Control and Prevention
CAS	Chemical Abstracts Service
CAM	Continuous Air Monitor
CFR	Code of Federal Regulations
CED	Committed Effective Dose
D	Day
D&D	Decontamination and Decommissioning
DTPA	Diethylenetriamine Pentaacetic Acid
DAC	Derived Air Concentration
DPM	Disintegrations Per Minute
DOE	U.S. Department of Energy
DCF	Dose Conversion Factor
EA	Environmental Assessment
EIS	Environmental Impact Statement
EMG	Emergency Management Guide
EMS	Emergency Management System
EOC	Emergency Operations Center
EPA	U. S. Environmental Protection Agency
EPHA	Emergency Planning Hazards Assessment
ERO	Emergency Response Organization
F	Fast
FDA	U. S. Food and Drug Administration
GI	Gastrointestinal
GM	Geiger-Mueller
HPS	Health Physics Society
HEPA	High Efficiency Particulate Air
HLW	High-Level Waste
HQ	Headquarters
IMBA	Integrated Modules for Bioassay Analysis
IG	Implementation Guide
IRF	Intake Retention Function
ISMS	Integrated Safety Management System
ICRP	International Commission on Radiological Protection
IEC	International Electrotechnical Commission
ISO	International Organization for Standardization
LANL	Los Alamos National Laboratory
LET	Linear Energy Transfer
LLW	Low-Level Waste
M	Medium
MDA	Minimum Detectable Amount/Activity
MDD	Minimum Detectable Dose
MARSAME	Multi-Agency Radiation Survey and Assessment of Materials and Equipment

MARSSIM	Multi-Agency Radiation Survey and Site Investigation Manual
MW	Mixed Waste
NAD	Nuclear Accident Dosimeters
NCRP	National Council on Radiation Protection and Measurements
NEPA	National Environmental Policy Act
NIOSH	National Institute for Occupational Safety and Health
NNSA	National Nuclear Security Administration
NIST	National Institute of Standards and Technology
NRC	U. S. Nuclear Regulatory Commission
NVLAP	National Voluntary Laboratory Accreditation Program
ORNL	Oak Ridge National Laboratory
PAGs	Protective Action Guides
PNAD	Personnel Nuclear Accident Dosimeter
PSO	Program Secretarial Office
QC	Quality Control
RCRA	Resource Conservation and Recovery Act
RCT	Radiological Control Technician
RWP	Radiological Work Permit
RCS	DOE Radiological Control Standard
RESL	Radiological and Environmental Sciences Laboratory
S	Slow
SNAP	Space Nuclear Auxiliary Power
TEPC	Tissue Equivalent Proportional Counter
TS	Technical Standard
TLD	Thermoluminescent Dosimeter
TED	Total Effective Dose
TRU	Transuranic
TSCA	Toxic Substances Control Act
USC	United States Code
WIPP	Waste Isolation Pilot Plant
W	Week
Y	Year

1.4 DISCUSSION

Chapters 2 through 10 provide technical information to assist in safely managing plutonium operations. The topics covered are those considered by representatives of many of DOE's plutonium facilities to be most beneficial: Manufacture, Properties and Hazards, Radiation Protection, Contamination Control, Internal Dosimetry, External Dose Control, Nuclear Criticality Safety, Waste Management, Emergency Management, and Decontamination and Decommissioning.

2.0 MANUFACTURE, PROPERTIES, AND HAZARDS

This chapter briefly describes the manufacture of plutonium and presents the nuclear, physical, chemical, and radiobiological properties of plutonium (and/or sources for these data) that form the basis for radiological and toxic control limits. The data and discussion are intended to provide a basis for understanding the changes in hazards as a function of such parameters as isotopic composition, age since chemical processing, physical form, and chemical form. Data are presented to facilitate the calculation of radiation effects, which occur from a variety of plutonium sources.

Plutonium is the first man-made element produced on an industrial scale. The special nuclear properties of ^{239}Pu and ^{238}Pu have led scientists to focus their efforts on these two isotopes. The fission cross-section of ^{239}Pu makes it a useful energy source for atomic weapons and nuclear power reactors. The 87.7-year half-life of ^{238}Pu makes it an excellent heat source for space applications. Unfortunately, the same nuclear properties of plutonium that make it attractive to science also make this element hazardous to human beings. All 15 plutonium isotopes are radioactive, with half-lives ranging from 26 minutes for ^{235}Pu to 7.6×10^7 years for ^{244}Pu.

2.1 MANUFACTURE OF PLUTONIUM

Because of its high specific alpha activity and high decay heat, ^{238}Pu has been used as an isotopic heat source for devices that generate thermoelectric power, such as the Space Nuclear Auxiliary Power (SNAP) systems used in lunar and deep space missions. Small amounts of ^{238}Pu with low ^{236}Pu content were used as a power source for medical prosthetic devices such as cardiac pacemakers and a prototype artificial heart, but lithium batteries have replaced these plutonium power sources. ^{238}Pu containing a few parts per million of ^{236}Pu is produced by irradiating ^{237}Np with slow neutrons. It can also be produced by irradiating ^{241}Am to form ^{242}Cm, which quickly decays to ^{238}Pu.

In the past, most plutonium in DOE facilities was produced for nuclear weapons and was composed of greater than 90 wt% ^{239}Pu and about 6 to 8 wt% ^{240}Pu. This material has been referred to as “weapons grade” or “low exposure” plutonium. It is produced on a large scale by irradiating ^{238}U in moderated production reactors (see Figure 2.1). Plutonium has also been produced as a byproduct in the operation of research reactors, and commercial nuclear power plants. It is recovered and purified by solvent extraction and ion exchange processes. The resulting highly concentrated $Pu(NO_3)_4$ product solution is converted to a nonhygroscopic PuF_4 intermediate by one of the several processes before being reduced to metal with calcium. Plutonium is also produced from the waste streams of the conversion processes and scrap recovery operations, which include material from research and development efforts. Other processes for reduction to metal include direct reduction of the oxide and electrolytic reduction. Typical isotopic compositions of three common grades of plutonium are given in Table 2.1.

$$^{238}_{92}\mathrm{U} + ^{1}_{0}\mathrm{n} \longrightarrow ^{239}_{92}\mathrm{U} \xrightarrow[23.5\ \mathrm{min}]{\beta^-} ^{239}_{93}\mathrm{Np} \xrightarrow[2.3565\ \mathrm{d}]{\beta^-} ^{239}_{94}\mathrm{Pu}$$

Figure 2.1. Principal Mode of Plutonium Production by Neutron Irradiation of Uranium

Table 2.1. Isotopic Composition of Three Grades of Plutonium: Heat Source, Weapons, and Reactor (PNL, 1988)

Isotope	Heat Source	Weapons Grade	Reactor Grade
^{238}Pu	90.0	<0.05	1.5
^{239}Pu	9.1	93.6	58.1
^{240}Pu	0.6	6.0	24.1
^{241}Pu	0.03	0.4	11.4
^{242}Pu	<0.01	<0.05	4.9

Overviews of plutonium process chemistry at DOE's Hanford, Los Alamos, Rocky Flats, and Savannah River sites are given by Christensen et al. (1983), Baldwin and Navratil (1983), Coops et al. (1983), and Christensen and Mullins (1983). In each case, solutions for recovery, purification, and waste treatment operations are emphasized.

2.1.1 Future Sources of Plutonium

High-exposure plutonium, i.e., plutonium containing significant fractions of ^{240}Pu, ^{241}Pu, and ^{242}Pu and generating external dose rate fields greater than the other isotopes of plutonium, is produced in power reactor fuels. Currently, this form of plutonium is in the irradiated fuel in spent-fuel storage basins and other sources resulting from development work performed to demonstrate plutonium fuel cycles. Because recycling of commercial reactor fuel is not anticipated, future supplies of plutonium will be primarily from DOE production facilities and from reprocessing of current material. In the more distant future, Space Nuclear Auxiliary Power may be a potential source of plutonium.

Special isotopes of reasonably high purity are also available, which can be useful to health physicists for calibration purposes. These isotopes and their sources are listed in Table 2.2.

New sources of plutonium include the return of atomic weapon components and plutonium recovered from decontamination and decommissioning (D&D) operations. Foreign plutonium from states of the former Soviet Union may become an additional source. Their weapons-grade plutonium is believed to contain 5% ^{240}Pu. Americium is not periodically removed from their stockpile material.

Table 2.2 Uses and Availabilities of Plutonium Isotopes

Isotopes	Uses	Availability
^{236}Pu, ^{237}Pu	Popular environmental and biological chemical tracers.	Both available in microcurie quantities.(a)
^{238}Pu	Small thermal and electric-power generators.	Available in various isotopic enrichments, ranging from 78% to 99+%.(a)
^{239}Pu	Nuclear weapons and as a fast reactor fuel. Also, frequently used in chemical research where production-grade material of mixed isotopic content is suitable.	Available enrichments range from 97% to 99.99+%.(a)
^{240}Pu	Principally in flux monitors for fast reactors.	Available enrichments range from 93% to 99+%.(a)
^{241}Pu	The parent from which high-assay ^{241}Am can be isolated for industrial purposes.	Samples available in enrichments of 93%(a)
^{242}Pu	For study of the physical properties of plutonium; also as a mass spectroscopy tracer and standard.	Samples available in enrichments ranging from 95% to 99.9+%; enrichments of production-grade material range from 85% to 95%.(a)
^{244}Pu	Available as a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM)	DOE's New Brunswick Laboratory.

(a) Available in small quantities from the Oak Ridge National Laboratory (ORNL): ORNL Isotopes Sales Office, Oak Ridge National Laboratory, P.O. Box X, Oak Ridge, Tennessee 37830.

2.2 NUCLEAR PROPERTIES

Of the 15 plutonium isotopes, the two that have proven most useful are masses 239 and 238. Plutonium-239 is fissile, i.e., atoms of plutonium split upon exposure to thermal or fast neutrons. Chemical reactions can release a few electron volts of energy per atom; however, when a plutonium nucleus splits, it releases about 200 MeV of energy and two or three neutrons. This release of energy makes ^{239}Pu useful for nuclear weapons and reactor fuel. In fact, in light water reactors much of the power originates from the fission of ^{239}Pu, which is produced by neutron capture in ^{238}U. Because of its higher specific activity, ^{238}Pu is used as long-lived heat sources for powering planetary space missions where adequate solar energy is not available.

As mentioned before, all plutonium isotopes are radioactive. Isotopes with even mass numbers (except mass number 246) are primarily alpha emitters. Isotopes of mass numbers 232, 233, 234, 235, and 237 also decay by electron capture; isotopes of mass numbers 241, 243, 245, and 246 decay by beta emission. Many of the alpha-emitting isotopes, such as ^{238}Pu and ^{240}Pu, also fission spontaneously and emit neutrons. All of the particle emissions are accompanied by X-ray and gamma-ray emissions over a wide range of energies.

A review of the nuclear properties of plutonium (e.g., cross-sections, nuclear levels, half-lives, and fission yields) can be found in Volume 1 of the Plutonium Handbook: A Guide to the Technology (Wick, 1967) and in American National Standards Institute (ANSI) Standard N317, Performance Criteria for Instrumentation Used for In-Plant Plutonium Monitoring (ANSI, 1980a). Plutonium decay schemes, neutron yields, and neutron energy spectra are described in the following sections.

2.2.1 Decay Schemes

The decay modes of some important plutonium and other isotopes and decay products are shown in Table 2.3. For brevity, only the most abundant radiations have been included in the table; more detailed information can be found in papers by Gunnink and Morrow (1967) and Klein (1971), in International Commission on Radiological Protection (ICRP) Publication 107 (ICRP, 2007), and from the National Nuclear Data Center. Most of the isotopes are strong alpha-emitters, making alpha heating a problem for the storage and handling of large amounts of plutonium. The specific activities and decay heats for selected isotopes and decay products are given in Table 2.4. Kilogram quantities of ^{239}Pu or gram quantities of ^{238}Pu can generate enough heat to melt plastic bags. Sources of ^{238}Pu shall be handled with insulated gloves, and special precautions shall be taken to ensure a good thermal heat sink during shipping and storage. (See also Section 2.5.1, “Self-Heating.”)

The plutonium isotopes emit relatively few high-energy gamma rays, so even kilogram quantities can be processed without serious gamma-exposure problems. Because of the high density of plutonium, many gamma rays are self-absorbed. In some instances, the decay products may become significant in radiation protection and metallurgy. For instance, the isotope ^{236}Pu often constitutes less than 1% of plutonium and is often ignored in dose calculations. However, if the plutonium is shielded by greater than 1 cm of lead or steel, the decay products of ^{236}Pu may be the largest contributors to exposure. The decay product ^{208}Tl emits a highly penetrating gamma ray with an energy of 2.615 MeV. Although ^{241}Pu is a beta emitter and not as great an inhalation hazard as other isotopes of plutonium, in plutonium that contains a few weight percent ^{241}Pu the ^{241}Am decay product is important because it emits a large number of 60-keV photons, which can be a significant source of exposure to the hands and forearms when handling plutonium in gloveboxes (See Section 6.3.3 for more information). Also, ^{241}Am can contribute to neutron dose. Americium-241 contributes to increased alpha emission which affects the neutron dose as well as radiolysis and helium retention and release. Because of its importance to radiation exposure, the fractional amount of ^{241}Am produced by beta decay from ^{241}Pu is given as a function of time since chemical separation (see Figure 2.2).

Table 2.3. Radioactive Decay Properties of Selected Isotopes and Decay Products, Excluding Spontaneous Fission (a)

Isotope	Half-Life	Mode of Decay Particle	Energy Mev	Yield %	X-ray (b) Energy MeV	Yield %	Gamma Ray Energy MeV	Yield %
^{236}Pu	2.851 y	α	5.77	69.3	L's 0.011-0.021	13(c)	0.0476	6.6x10^{-2}
		α	5.72	30.6			0.109	1.2x10^{-2}
^{238}Pu	87.7 y	α	5.50	71.0	L's 0.011-0.021	15(c)	0.0425	3.95x10^{-2}
		α	5.46	28.8			0.0999	7.35x10^{-3}
^{239}Pu	2.41 x 10^{4} y	α	5.157	73.1	L's 0.0116-0.0215	5.0(c)	0.099	1.22x10^{-3}
		α	5.144	15.0			0.129	6.41x10^{-3}
		α	5.106	11.8			0.375	1.55x10^{-3}
							0.414	1.46x10^{-3}
^{240}Pu	6564 y	α	5.168	72.8	L's 0.0115-0.0215	10.8(c)	0.0452	4.50x10^{-2}
		α	5.124	27.1			0.104	7.08x10^{-3}
^{241}Pu	14.35 y	β	0.0052(d)	100	--	--	0.077	2.20x10^{-5}
		α	4.896	2.04 x 10^{-3}			0.1037	1.01x10^{-4}
							0.114	6.0x10^{-6}
							0.149	1.9x10^{-4}
							0.160	6.71x10^{-6}
^{242}Pu	3.73 x 10^{5} y	α	4.901	77.5	L's 0.0116-0.0215	9.1(c)	0.0449	3.6x10^{-2}
		α	4.857	22.4			0.104	7.8x10^{-3}
^{241}Am	432.2 y	α	5.486	85.2	L's 0.0119-0.0222	42(c)	0.0263	2.4
		α	5.443	12.8			0.0332	1.2x10^{-1}
		α	5.388	1.4			0.0595	35.7
^{237}U	6.75 d	β	0.039(e)	0.8	L's 0.0119-0.0206	70(c)	0.0263	2.43
		β	0.050(e)	3.4			0.0595	34.5
		β	0.065(e)	51			0.0648	1.28
		β	0.069(e)	42	K's 0.097-0.114	53	0.165	1.85
							0.208	21.1
							0.268	7.1x10^{-1}
							0.332	1.2
							0.335	9.5x10^{-2}
							0.369	4.0x10^{-2}
							0.371	1.1x10^{-1}

(a) Data from Dunford and Burrows (1993).

(b) L's = L X-rays; K's = K X-rays.

(c) Total for all X-rays. The value represents an average obtained from data at Pacific Northwest Laboratory, Lawrence Berkeley Laboratory, and Lawrence Livermore Laboratory.

(d) Average beta energy given. The maximum beta average for ^{241}Pu is 0.0208 MeV.

(e) Average beta energy. The maximum beta energy for ^{237}U is 0.248 MeV.

Table 2.4. Specific Activity Decay Heats of Selected Isotopes(a)

Isotope	Half-Life Y	Specific Activity, Ci/G		Average Particle Energy per Disintegration, MeV(b)		Decay Heat W/g(b)
^{236}Pu	2.851	α	53.4	α	5.75	18.2
^{238}Pu	87.7	α	17.1	α	5.49	0.567
^{239}Pu	2.407 x 10^{4}	α	6.22 x 10^{-2}	α	5.14	1.93 x 10^{-3}
^{240}Pu	6564	α	0.229	α	5.16	7.13 x 10^{-3}
^{241}Pu	14.35	α	2.52 x 10^{-3}	α+β	5.27 x 10^{-3}	3.29 x 10^{-3}
		β	103			
^{242}Pu	3.733 x 10^{5}	α	3.93 x 10^{-3}	α	4.90	1.16 x 10^{-4}
^{232}U	72.0	α	21.5	α	5.31	0.690
^{233}U	1.59 x 10^{5}	α	9.75 x 10^{-3}	α	4.72	2.84 x 10^{-4}
^{234}U	2.45 x 10^{5}	α	6.29 x 10^{-3}	α	4.76	1.81 x 10^{-4}
^{235}U	7.04 x 10^{8}	α	2.17x10^{-6}	α	4.24	6.02 x 10^{-8}
^{236}U	2.34 x 10^{7}	α	6.5 x 10^{-5}	α	4.48	1.77 x 10^{-6}
^{238}U	4.47 x 10^{9}	α	3.38 x 10^{-7}	α	4.18	8.58 x 10^{-9}
^{237}Np	2.14 x 10^{6}	α	7.08 x 10^{-4}	α	4.76	2.08 x 10^{-5}
^{241}Am	432.2	α	3.43	α	5.37	0.115

(a) Data from ICRP 38 (1983).
(b) Includes atomic recoil and low-energy X-ray production.

2.2.2 Neutron Yields and Spectra

Plutonium and plutonium compounds also emit neutrons from spontaneous fission and from alpha-neutron reactions with light elements. The spontaneous fission half-life and the neutron yields from spontaneous fission and alpha-neutron reactions for plutonium metal and plutonium compounds are provided in Section 6.0 of this TS. The approximate neutron yield from a substance with a known isotopic composition can be determined by adding the contributions from each component. This procedure and its limitations are described in detail in Section 6.0, which also discusses neutron equivalent dose rates.

Energy spectra from Pu-Be and Pu-B neutron sources are shown in Figure 2.3 Because of licensing restrictions on plutonium, these sources have been replaced with source fabricated from americium. Metallic plutonium emits neutrons having a Maxwellian energy distribution, with an average energy of about 1.9 MeV. Plutonium compounds and alloys also emit neutrons from alpha-neutron reactions, and these neutrons have significantly different energies:

-- PuF_4, about 1.3 MeV
-- 10% plutonium-aluminum alloys, 1.6 MeV
-- PuO_2, slightly more than 2 MeV
-- PuBe, 4.3 MeV.

Plutonium compounds or alloys containing sodium, magnesium, silicon, chlorine, carbon, or oxygen have significant alpha-neutron yields, but little information is available about their neutron energy spectra.

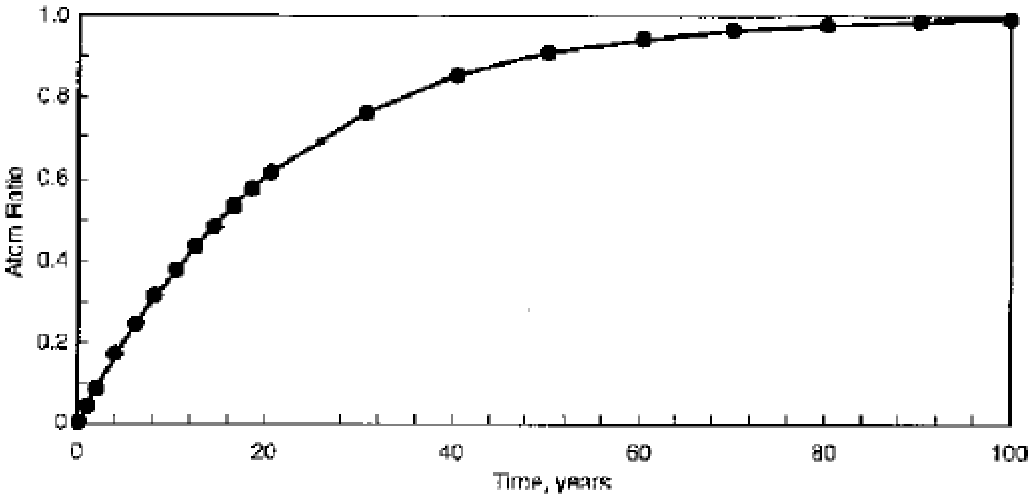

Figure 2.2. Atom Ratio of ^{241}Am to ^{241}Pu (t=0) Produced by the Beta Decay of ^{241}Pu as a Function of Time Since Chemical Separation

2.3 PHYSICAL AND CHEMICAL PROPERTIES

This discussion of plutonium's physical and chemical properties begins with plutonium metal, followed by its alloys and compounds. Knowledge of the physical properties of these classes of materials and how the plutonium was produced is the key to understanding and predicting the hazards of working with this challenging element. According to Healy (1993), “Nature does not decide what happens to any material based on its radioactivity but rather on its form and mass.” Form and mass are determined by the engineering application and the kinds of processes needed to achieve both intermediate and final products. Thus, to prevent nature from taking its course, there can be no shortcuts in good practices for plutonium facilities.

2.3.1 Plutonium Metal

The metallic state of plutonium is undoubtedly the most complicated of all the elements. Plutonium is a silvery-white metal, much like nickel in appearance. It has a low melting point (640°C) and an unusually high boiling point (3327°C). The metal exists in six allotropic forms, as indicated in Table 2.5. Two of the allotropic forms, σ and σ’, contract upon heating; the other forms expand upon heating.

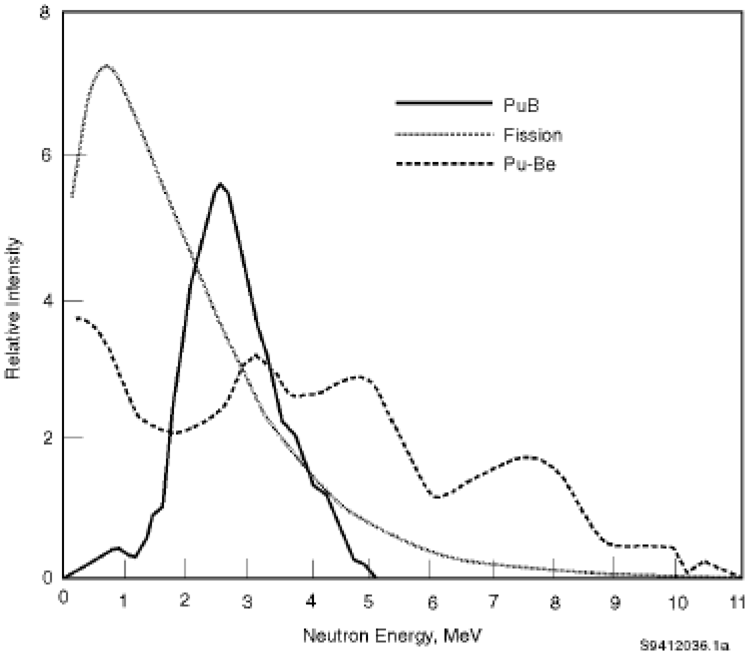

Figure 2.3 Neutron Energy Spectra of Plutonium-Beryllium and Plutonium-Boron Neutron Sources Compared with a Fission Source

At room temperature, pure plutonium exists in the α phase, which has a triclinic structure with a theoretical density of about 19.86 g/cm^3. The dimensional stability of this phase is aggravated by its high linear thermal expansion coefficient and its low α → β transition temperature. This transformation takes place at approximately 115°C, resulting in a 10% volume change. The combination of a high specific activity and low thermal conductivity can result in significant dimensional distortion during metal-forming operations. For this reason, a σ-stabilized dilute gallium alloy, which has a density of about 15.75 g/cm^3, is used when a more dimensionally stable plutonium is desired (Merz, 1971).

Table 2.5. Allotropic Forms of Plutonium Metal(a)

Phase	Stability Range °C	Density g/cm^3(b)
α	Stable below 115	19.86
β	~115 to 200	17.70
λ	~200 to 310	17.14
σ	310 to 452	15.92
σ ’	452 to 480	16
€	480 to 640	16.51

(a) Wick, 1967, p. 34.
(b) Theoretical X-ray density. The actual density is slightly lower due to crystal lattice imperfection.

Plutonium is an active metal. In moist air or moist argon, the metal oxidizes rapidly, producing a mixture of oxides and hydrides (Haschke, 1992). If the metal is exposed long enough, an olive-green powdery surface coating of PuO_2 is formed. With this coating, the metal is pyrophoric, so plutonium metal is usually handled in an inert, dry atmosphere of nitrogen or argon. Oxygen retards the effects of moisture and acts as a passivating agent (Raynor and Sackman, 1963). For a description of the storage hazards that the oxidation of plutonium metal creates, see Section 2.6.3.1, “Oxidation of Plutonium.” A comprehensive treatment of the oxidation of plutonium, the properties of its oxides, oxide growth, and oxidation kinetics was reviewed by Colmenares (1975).

Plutonium metal also reacts with most common gases at elevated temperatures. Plutonium metal is rapidly dissolved by HCl, HBr, 72% $HCl0_4$, 85% H_3PO_4, concentrated CCl_3COOH (trichloroacetic acid), sulfamic acid, and boiling concentrated HNO_3 in the presence of 0.005M HF. The metal reacts slowly with water, dilute sulfuric acid, and dilute acetic acid. There is no reaction with the metal in pure HNO_3 at any concentration, with concentrated acetic acid, nor with dilute sodium hydroxide.

2.3.2 Plutonium Alloys

Alloying plutonium gives rise to a host of materials with a wide range of physical, chemical, and nuclear properties.[1] The search for and development of new alloys has been focused mainly on the manufacture of atomic weapons, reactor fuels, heat sources, and neutron sources. The challenge of alloy development is how to maximize the desired properties without adding undesired ones. Unfortunately, some properties mutually exclude others (e.g., a gain in hardness usually results in a loss of ductility), so users may be forced to rethink their needs.

The radiological hazards of a plutonium alloy taken through its product life cycle differ from those of the pure metal isotope by virtue of the alloy's properties, which affect its form (i.e., its chemical composition, density, and geometric shape). Because form can be radically changed by external conditions (e.g., heat, pressure, and chemical atmosphere), a knowledge of the following properties will aid in evaluating the radioactive hazard:

-- melting point
-- viscosity
-- vapor pressure
-- corrosion resistance
-- diffusivity
-- strength
-- ductility
-- pyrophoricity.

In nuclear fuel applications, the neutron absorption cross-section of the alloying elements and impurities shall also be considered for its effect on radiation exposure.

2.3.3 Plutonium Compounds

Much of what was said in Section 2.3.2 about the properties of plutonium alloys also applies to plutonium compounds because both are mixtures of plutonium and other elements.

Plutonium is the fifth element in the actinide series, which consists of elements with properties that stem from partial vacancies in the 5th electron shell. These elements form the seventh row in the periodic table. In general, there are four oxidation states: III, IV, V, and VI. In aqueous solutions, plutonium (III) is oxidized into plutonium (IV), which is the most stable state. The compounds PuF_4, $Pu(I0_3)_4$, $Pu(OH)_4$, and $Pu(C_2O_4)_2\,6H_2O$

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[1] See Volume 1 (Section 2) and Volume 2 (Section 5) of the Plutonium Handbook: A Guide to the Technology (Wick, 1967); Plutonium (Taube, 1964); and Chapter 11 of the"Reactor Handbook" in Materials, vol. 1 (Tipton, 1960). Beginning in 1957, a series of international conferences were held whose proceedings contain a wealth of information on plutonium alloys. From 1960 through 1975, the conferences were held every five years and produced a proceedings for each conference: Refer to The Metal Plutonium (Coffinberry and Miner, 1961); Plutonium 1960 (Grison et al., 1961); " Plutonium 1965" (Kay and Waldron, 1966); "Plutonium 1970 and Other Actinides," Parts I and II (Miner, 1971); and "Plutonium 1975 and Other Actinides" (Blank and Lindner, 1976).

(plutonium oxalate) are insoluble in water. The chlorides, nitrates, perchlorates, and sulfates are soluble in water. Plutonium (IV) ions complex readily with organic and inorganic compounds. Of particular importance for radiological safety considerations are the solubility, particle size, and surface area of plutonium compounds. These properties play an important part in the transportability of plutonium in the environment and in the body. All plutonium compounds, except the oxides, were assumed in ICRP 30, Part 1 (ICRP, 1979) to behave as class W compounds in the ICRP lung model. Plutonium oxides were assumed to be class Y. The 2007 amendment to 10 CFR 835 adopted models from ICRP 60, 61, 66 and 68 (ICRP, 1991a, 1191b, 1994a, and 1994b). These models assumed plutonium compounds had an absorption type of either M or S (medium or slow). The solubility of plutonium compounds is an important parameter in avoiding “unintentional” homogeneous reactions. Knowledge of this property for both aqueous and organic solvents plays a key role in criticality safety and deserves a high priority.

Unfortunately, little data on particle-size are available, and those that have been generated focus on the reactivity of the materials in the separation and conversion processes. Much of the data are reported as crystallite size, which relates to surface area and solubility but not necessarily to the way the particles would be dispersed in the air. Surface area plays a role in the ability of materials to adsorb gases and vapors that can affect the long-term storage behavior of plutonium compounds. Pressure buildup in storage containers, either from out gassing due to self-heating or radiolytic effects, will depend on the stability of the compound and the amounts of chemisorbed or physisorbed water or other substances.

The following sections discuss the essential compounds of plutonium: plutonium nitrate and associated compounds, plutonium dioxide, plutonium hydride, plutonium sulfate, plutonium chlorides, and plutonium fuel mixtures.

2.3.3.1 Plutonium Nitrate, Oxalate, Peroxide, and Fluorides

Plutonium (IV) nitrate is the most used of all plutonium compounds. Essentially all chemical processing of plutonium has been conducted in nitrate solutions. These solutions of appropriate acidities range from concentrations of 10g to 250g of Pu/L for efficient precipitation processes. Intermediate compounds are also used in the processing of plutonium prepared from the nitrate: plutonium (III) fluoride, plutonium (II or IV) oxalate, and plutonium peroxide. Plutonium (IV) fluoride can be prepared from any of the preceding solids by hydrofluorination. Plutonium fluoride has been the compound of choice for reduction to the metal with calcium, principally because it is nonhygroscopic. The solubilities in various media, bulk densities, and particle sizes of these compounds are given in Table 2.6.

Table 2.6. Solubilities and Properties of Selected Compounds

	Measured Solubility		Bulk Density, g Pu/L		
Compound	Medium	g/PuL	Filter Cake	Dry Compound	Sintered Media Porosity, μm[(a)]
Flouride (III)	1M HF – 1M HCl	0.03	--	1-2.5	15-20
Fluoride (IV)	2M HF – 2M HNO_3	0.70	0.6-0.8	0.5-2.0	15-20
Oxalate (III)	0.5M $C_2O_4^{2-}$ - 3M HNO_3	0.01	0.6-0.8	-	15-20
Oxalate (IV)	0.1M $C_2O_4^{2-}$ - 4M HNO_3	0.003	0.5-0.6	0.6	15-20
Peroxide (IV)	3M H_2O_2 - 1M HNO_3	0.10	0.10-0.6	-	30-80

(a) Sintered media porosity required to remain precipitate.

Plutonium hexafluoride is the only volatile plutonium compound (bp 62°C) and is marginally stable. It can be prepared by oxidizing $PuF_4$with F_2 at an elevated temperature (Weinstock and Malm, 1956). It can also be prepared at low temperatures by a fluorinating agent, fluorine dioxide (Malm et al., 1984). Plutonium waste treatment and decontamination may benefit from processes using photolysis or microwave discharge to produce active fluorine species from FOOF or CF_4/O_2 mixtures, which will react with plutonium or plutonium dioxide to form PuF_6(Martz et al., 1991).

2.3.3.2 Plutonium Dioxide

Plutonium dioxide may now be the most important and most thoroughly studied of all plutonium compounds. Due to its chemical stability and relative inertness, it is the preferred form for shipping and storing plutonium at the present time. Direct oxide reduction (DOR) of PuO_2 is part of the integrated pyrochemical system used at the Los Alamos National Laboratory (LANL) (Christensen and Mullins, 1983; Mullins et al., 1982). Plutonium dioxide is formed when plutonium or its compounds (except the phosphates) are ignited in air, and often results when oxygen-containing compounds are heated in vacuum or in an inert atmosphere to 1000°C (Cleveland, 1970). The properties of PuO_2 are reported by Moseley and Wing (1965).

Loose PuO_2 powder, as formed by calcination, usually has a density of about 2 g/cm^3. If the oxide is pressed and sintered into pellets, it may have a density of about 10.3 to 11.0 g/cm^3. Surface measurements of typical oxides prepared from the calcination of plutonium (IV) oxalate at various temperatures range from 10 to 60 m_2/g. Caldwell (1961) found that the surface area decreased with increasing temperatures. Plutonium oxide fired at temperatures >600°C is difficult to rapidly or completely dissolve in common acids or molten salts. The best solvents are 12–16M HNO_3 with 0.10–0.1M HF, 5–6M HI, and 9M HBr (Cleveland, 1964; Holley et al., 1958). Processes were developed to correct this deficiency using a superacid, HF/SbF_5 (Olaha et al., 1985) and CEPOD, a fluoride-free electrochemical dissolver that used the silver anion as a redox catalyst (Bray et al., 1987).

2.3.3.3 Plutonium Hydride

Plutonium hydride is a compound of interest for separating plutonium scrap from other materials that do not readily unite with hydrogen.[2] The reaction between plutonium and hydrogen apparently proceeds by the initial formation of PuH_2. As more hydrogen is added, the dihydride becomes PuH_{2+x}. The hexagonal PuH_3 begins to form when the H/Pu ratio becomes about 2.75; when the H/Pu ratio reaches 2.9 to 3.0, only the hexagonal form remains. A wide spread is reported in the measured induction period for the first reaction (Haschke, 1991). Because the hydriding reaction is fully reversible, plutonium metal can be recovered by pumping off the hydrogen in a suitable vacuum furnace. This metal typically contains significant amounts of plutonium oxide but is suitable for feed to either molten salt extraction or electrorefining processes. The hydride can also be converted to the oxide. The advantage of the hydride recovery process is its ability to recover a large fraction of the scrap in metallic form. This method, therefore, has a major economic advantage over chemical recycling and subsequent reduction to metal. It is being used as a production aid for metallic scrap recovery.

2.3.3.4 Plutonium Sulfates

Plutonium sulfate tetrahydrate, $Pu(SO_4)_2 \bullet 4H_2O$, has not been of any process importance but has been of interest as a primary standard for plutonium. It is a good example of a stable compound that could be suitable as an interim storage form. Samples stored at relative humidities of up to 75% showed no evidence of alpha radiolysis of the water of crystallization after 28 months. The compound is hygroscopic in air of 95% relative humidity, and stable up to 650°C, at which point it quickly decomposes to PuO_2 (Cleveland, 1970). The potassium salt, $K_4Pu(SO_4)_4$- $1H_2O$, was under study as a possible primary standard for ^{238}Pu. Crystals stored in an air-tight steel container, which also functioned as a heat sink, proved to be stable. The solubility product of this compound was determined to be 10^{-18}.

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[2] The properties of plutonium hydrides may be found in Volume 3 of the Handbook of Physics and Chemistry of the Actinides (Ward, 1985). Kinetics of the plutonium hydrogen reaction are reviewed by Haschke (1991).

2.3.3.5 Plutonium Chlorides

Chloride salts, which are a very important category of residues, are byproducts of pyrochemical operations. Pyrochemical chloride-based operations currently in use include:

-- DOR
-- electrorefining
-- molten salt extraction
-- pyroredox.

Treatment of chloride-based residues is especially challenging for aqueous recovery techniques because of corrosion problems with stainless steel equipment. At the LANL site, Kynar-lined gloveboxes were installed to evaluate their behavior in production-scale operations. The Rocky Flats Plant (RFP) also had extensive experience in aqueous recovery of plutonium from chloride-based residues (Muscatello et al., 1986a, 1986b, 1987). Cesium chloroplutonate, Cs_2PuCl_6, was a primary analytical standard due to its stability to alpha radiolysis and may now have application as a storage form. It was first prepared by Anderson (1949). There is no evidence of water absorption at relative humidities as high as 53% (Miner et al., 1963). After 64 days at 90% relative humidity, Cs_2PuCl_6 forms a paste.

2.3.3.6 Plutonium Fuels

Plutonium and plutonium-uranium fuel mixtures were developed and tested in experimental reactors to prove the feasibility of operating power reactors. These fuels included both liquids and solids consisting of alloys and ceramic mixtures. Wick (1967) and Schneider and Roepenack (1986) provide comprehensive lists of fuels. Because of their pyrophoric nature, some of these alloys and compounds require special care and handling when exposed to reactive liquids or gases.

2.4 RADIOLOGICAL EFFECTS ON HUMANS

The radiobiological properties of plutonium and other transuranic (TRU) elements are known primarily from experiments performed on rats, dogs, baboons, and rabbits. Human data on plutonium are limited. Reviews of the vast literature on plutonium include Hodge et al. (1973); ICRP 19 (1972); ICRP 30, Part 1 (1979); ICRP 48 (1986); ICRP 30, Part 4 (1988b); and Liverman et al. (1974). Factors affecting radiobiological effects include the mode of entry of plutonium into the body, its distribution in the body, and its transfer to a fetus.

2.4.1 Modes of Entry into the Body

Radioactive material can enter the body by four different pathways: by inhalation, through a wound (including an accidental injection), by ingestion, or by absorption through intact skin. These pathways may occur singly or in any combination.

-- Inhalation is probably the most prevalent mode for occupational intake of plutonium. It also provides a generally conservative assumption of intake for designing bioassay programs.

-- Wounds are potentially the most serious mode of intake because of the high dose-per-unit uptake of plutonium. Wounds can result from direct penetration by an object (i.e., a puncture or cut), from abrasion, or from burning by an acid, caustic, or thermal source.

-- Occupational ingestion of plutonium poses a relatively small risk because the uptake factor from the GI (gastrointestinal) tract to the blood is quite small and because most of the alpha energy from transformations within the GI tract is absorbed by the contents of the GI tract, rather than by the target tissues of the tract itself.

-- Absorption of plutonium through intact skin is, for practical purposes, almost nonexistent. However, when removing skin contamination, care shall be taken to ensure that the skin integrity is not damaged by rough or extensive decontamination procedures. If the skin integrity is damaged, the result can be considered a wound, regardless of how it occurred.

2.4.2 Distribution Within the Body

Three commonly encountered biokinetic models were promulgated by the ICRP for the internal distribution and retention of plutonium. These models are identified by the ICRP publications in which they were first reported: ICRP 30, Part 1 (1979), ICRP 48 (1986), and ICRP 30, Part 4 (1988b). These models were later updated with models derived from ICRP 60, 67 and 68 values. The models are all similar with regard to the organs of significance, but differ with regard to the fraction of uptake deposited in the organ and its respective retention (or clearance) half-time in the organ.

In all the ICRP models, once plutonium has reached the bloodstream, it is translocated primarily to the liver and skeleton. In the skeleton, it is deposited primarily on the endosteal surfaces of mineral bone, from which it is gradually redistributed throughout the bone volume by resorption and burial. Because of the extremely slow nature of this redistribution, plutonium is considered to be uniformly distributed over bone surfaces at all times following skeleton deposition. A small fraction of the translocated plutonium reaches the gonads. Although the gonadal fraction is different for males and females, the calculated gonadal doses are the same regardless of gender because the plutonium concentration in the tissues is assumed to be the same. The ICRP assumes that the remainder goes directly to excretion.

Metabolic distribution and retention parameters for the ICRP models are shown in Table 2.7. The table also includes the absorption factors from the GI tract to the bloodstream, as well as the inhalation class of common forms of plutonium.

Table 2.7 includes values from ICRP 60, 67 and 68 models which includes data from the Human Respiratory Tract Model for Radiological Protection Publication 66 (ICRP, 1994) and Age- Dependent Dose to Members of the Public from Intakes of Radionuclides: Part 2 Ingestion Dose Coefficients (ICRP, 1993). The Human Respiratory Tract Model constitutes an updating of the model used in Publication 30 for workers. The new model takes into account extensive data on the behavior of inhaled materials that has become available since the Publication 30 model was developed.

Americium, as an ingrown impurity from the decay of ^{241}Pu, can behave the same way as the plutonium host matrix in which it is contained. This implies that the ^{241}Am associated with an absorption type S inhalation of plutonium might exhibit absorption type S behavior, rather than the absorption type M behavior assigned by the ICRP. This type of observation was made in ICRP 48 (1996) and by Eidson (1980).

Experience has shown that the biokinetic models in Table 2.7 are subject to some significant variations. A Hanford plutonium-oxide-exposure case described by Carbaugh et al. (1991) has demonstrated lung retention far greater than that expected for a class Y material, leading to the suggestion of a tenaciously retained “super class Y” form. This phenomenon had been informally verified by dosimetry personnel at the Rocky Flats, Savannah River, and Los Alamos sites, and is supported in the literature by Foster (1991). At the other extreme, La Bone et al. (1992) have identified a circumstance in which a ^{238}Pu oxide inhalation class appeared to exhibit biokinetic behavior more characteristic of an inhalation class D material. These extremes emphasize the importance of addressing the uniqueness of individual workers and exposure circumstances when dealing with known intakes, rather than relying on the assumed standard models.

2.4.3 Transfer to the Fetus

In its most recent review of the metabolism of plutonium and related actinides, it was noted in ICRP 48 (1986) that there is no strong evidence for preferential deposition of plutonium in the fetus and that the concentration of plutonium in the bone of the embryo or fetus is rapidly diluted by growth. However, experimental animal studies have shown that plutonium crosses the placenta after injection in pregnant animals (Green et al., 1979). For fallout plutonium, it has been qualitatively confirmed in humans that plutonium crosses the placenta (Okabayashi and Watanabe, 1973). However, placental and fetal membranes appear to effectively trap a portion of the plutonium that might otherwise reach the fetus.

The behavior of plutonium in the embryo/fetus changes with the development of the embryo/fetus (Sikov, 1987; Sikov et al., 1992). Liver and bone surfaces are the principal sites of plutonium deposition in the embryo/fetus, accounting for approximately 80% of the deposited plutonium (ICRP 48, 1986). Plutonium that deposits on bone surfaces following prenatal or neonatal exposure gradually moves into the bone matrix during subsequent bone-remodeling processes.

The radiation doses produced in the embryonic stage are assumed to be relatively homogeneous and represent a small fraction of the doses received by the pregnant woman when averaged over all tissues. The dose to the fetus would constitute an even smaller fraction of the maternal dose to any tissue in which there was specific deposition (Sikov et al., 1992). As gestation progresses, there is an increase in the relative plutonium concentration in specific fetal tissues, namely the bone and liver (Sikov et al., 1992). Although limited information is available, experimental animal and human data suggest that the average concentration is higher in the fetus during the second or third trimesters than in soft tissues of the pregnant woman, exclusive of the liver, yet significantly less than in maternal tissues of primary deposition, i.e., the bone and liver.

Because placental structures, including the yolk sac, effectively trap plutonium, progenitor cells of the gametes and hematopoietic lines that appear initially in the blood islands of the yolk sac are irradiated while they are primitive stem cells. However, the dose received by the early embryonic cells and the detriment produced is not currently known.

Table 2.7. Common Biokinetic Models for Plutonium and Americium

Model Parameter	**ICRP 30, Pt 1**		**ICRP 48**		**ICRP 30, Pt 4**		**ICRP 66/67/68**
Metabolic Distribution(a)	**F**	**T**	**F**	**T**	**F**	**T**	
Bone Surfaces	0.45	100 y	0.50	50 y	0.45	50 y	n.a.(c)
Liver	0.45	40 y	0.30	20 y	0.45	20 y	n.a.
Gonads(b)							
Male	3.5 x 10-4		3.5 x 10-4		3.5 x 10-4		3.5 x 10-4
Female	1.1 x 10-4		1.1 x 10-4		1.1 x 10-4		1.1 x 10-4
GI Tract Absorption Factor							
Pu oxides	10^{-5}		10^{-5}		10^{-5}		10^{-5}
Pu nitrates	n.a.		10^{-4}		10^{-4}		10^{-4}
Pu others	10^{-4}		10^{-3}		10^{-3}		5×10^{-4}
Am (any)	5×10^{-4}		10^{-3}		10^{-3}		5×10^{-4}
Inhalation Class/Absorption Type							
Pu oxides	Y		Y		Y		S
Pu others	W		W		W		M
Am (any)	W		W		W		M

(a) F is the fraction of plutonium reaching the bloodstream that is translocated to the organ of concern. T is the retention (or clearance) half-time in the organ of concern.

(b) Plutonium is assumed to be uniformly concentrated in male and female gonadal tissue where it is permanently retained. The deposition fractions are derived based on the relative mass of the reference male and female tissues.

(c) n.a. = not specifically addressed, ICRP 67 lists a relative deposition for adults of 5/3 for skeleton/liver.

2.5 RADIATION EFFECTS ON MATERIALS

The following sections discuss, in order, self-heating and the various effects of radiolysis. Radioactive decay, particularly alpha decay, can and does affect operations in plutonium purification processes. The change in emphasis from plutonium production to waste cleanup, environmental restoration, and the retirement of nuclear weapons will present favorable circumstances for cumulative radiolytic effects, especially in the stabilization processes and the final storage form.

Self-heating and helium retention and release are also included in this section since they too are part of the end result of the alpha decay process. Neutron production from the alpha-neutron reaction is discussed in Section 6.0. The degree of all these effects depends on the plutonium isotopic composition and the americium impurity level. Table 2.8 lists potential hazards or damage to materials from exposure to radiation.

2.5.1 Self-Heating

Heat generated by radioactive decay in plutonium, its alloys, or its compounds can be calculated from data provided in Table 2.4, together with the isotopic composition and plutonium fraction. The power output of reactor-produced ^{239}Pu metal is usually in the range of 2 to 10 W/kg. According to Van Tuyl,[3] the equilibrium surface temperature of a metal can that contains 1.2 kg of plutonium at the higher specific power would be 150°C. This calculation is complex because it depends on the thermal conductivities and configuration of all the materials in the shipping container. Thermal diffusivity measurements reported by Kruger and Robbins (1975) were combined with existing heat capacity values to derive a curve for the thermal conductivity of the Pu-1wt% Ga alloy from room temperature to 600°C. Gram quantities of ^{238}Pu can melt from self-heating under poor heat-transfer conditions. The major effects to be expected from self-heating are phase transformation, dimensional changes, chemical reactions (depending on the gaseous environment or other materials in contact with the plutonium), and desorption of previously sorbed gases or vapors.

2.5.2 Radiolysis

In gases, liquids, and covalently bonded solids, the chemical effects of alpha particles and the associated recoil nucleus can cause ionization, excitation, and dissociation of molecules. From the energy requirement for ion pair formation, only about half the energy causes ionization; the other half goes into molecular excitation. Radiation effects are commonly measured by a quantity called the G-value, i.e, the number of molecules destroyed for each 100 eV of energy absorbed. For free radical production, this quantity is expressed as the G_R-value. For organic liquids, G_R-values typically range from 0.85 for carbon disulfide to 70 for carbon tetrachloride (Prevost-Bérnas et al., 1952).

Although there is a considerable body of data on the radiolysis of aqueous solutions, organic liquids, and solids irradiated by gamma rays, X-rays, and fast electrons, little has been published on the radiolysis of plutonium compounds, solvents containing plutonium, or radiation-induced damage in materials that come in contact with plutonium. Nevertheless, radiation-induced damage can affect all aspects of plutonium handling.

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[3] Van Tuyl, H. H. 1981. “Packaging of Plutonium for Storage or Shipment.” Unpublished report by the Pacific Northwest Laboratory task force chairperson to the U.S. Department of Energy.

Table 2.8. Potential Hazards or Damage to Materials from Exposure to Radiation

Radiation-Induced Reaction	Potential Hazard or Damage Problem
Radiolysis of oxygen-contaminated glovebox atmospheres	Production of ozone-damage to elastomers: gloves, seals, etc.
Gaseous PuF_6	Deposition of solid PuF_4 on equipment
PuO_2 exposed to hydrocarbons or humid environments	Production of hydrogen gas pressure buildup in nonvented containers.
Ion exchange resins	Damaged resin can react violently with HNO_3 or other oxidizers. Also may result in hydrogen gas- pressure buildup.
CCl_4 saturated with H_2O	Production of Cl_2. C_2Cl_6 HCl, and phosgene.
Polyethylene	Disintegrates with production of H_2.
Polyvinylchloride (PVC) plastics	Disintegrates with production of HCl-corrosion.
Tri-n-butylphosphate	Production of hydrogen and oxygen-pressure buildup in nonvented containers.
Aqueous plutonium solutions	Production of polymeric plutonium hydroxide (plutonium polymer), which plates out on vessel surfaces and piping, producing swelling, cracking, loss of ductility.
Low-acidity plutonium solutions	Increase in leachability.

It would be futile and inappropriate to list, let alone discuss, all the possible radiolytic reactions affecting plutonium-handling. However, it is important to recognize the potential for and anticipate the consequences of these reactions. The following sections cover a broad range of the types of radiation-induced damage common to plutonium handling.

2.5.2.1 Hydrogen Production

The G-value for the production of H_2 by the alpha radiolysis of pure water is 1.9±0.1 molecules of hydrogen per 100 eV (Prevost-Bérnas et al., 1952). Cleveland (1970) calculates that the energy released in 0.001M (0.24 g/L) of plutonium solution is on the order of 2×10^{14} eV per minute. Thus, the hydrogen evolution would be approximately 3.8×10^{15} molecules per liter per day for a 1M solution, or about 73 cm^3 of hydrogen per year.

The G-values for H_2 in solids irradiated by gamma rays are lower: 0.1 for ice (Johnson, 1970) and 0.01 for the hydrates of a large number of sulfates (Huang and Johnson, 1964). Because the stability of $PuSO_4 . 4H_20$ was found to be remarkably high (Cleveland, 1970), one of the sulfates may well serve as an alternate interim waste form. Dole (1974) summarized the radiation chemistry of polyethylene, quoting G-values for hydrogen as 5 molecules per 100 eV. Destruction of plutonium hexafluoride as the solid phase amounts to about 1.5% of the material per day (Weinstock and Malm, 1956). Cleveland (1970) calculated the mean change in average oxidation number in 0.5–2M of perchloric acid to be 0.018 moles per day, corresponding to a G-value of 3.2 equivalents per 100 eV. The formation of hydrogen peroxide from the radiolysis of water is believed to be the mechanism for the reduction of plutonium (VI) ions. Lower oxidation states are formed by the disproportionation of the plutonium (V) species.

Pressurization of storage containers holding TRU wastes is a potential hazard for both long and interim storage periods (Kazanjian et al., 1985). Sampling of TRU waste drums shows that hydrogen is usually created (Roggenthem et al., 1989). Waste drums with pinholes can "breathe" when the atmospheric pressure changes, thereby introducing water vapor. Water vapor adsorbed on plutonium compounds is radiolytically decomposed, thereby producing hydrogen. It may be possible to add pressure relief valves and appropriate in-line filters to waste drums. (See Section 2.7 for more information on storage and containment.)

2.5.2.2 Redox Reactions

In most chemical processes for purifying plutonium, it is essential to maintain its valence state. The formation of hydrogen peroxide from the radiolysis of water is believed to be the mechanism for the reduction of plutonium (VI) ions. Lower oxidation states are formed by the disproportionation of the plutonium (V) species. Cleveland (1970) calculated the mean change in average oxidation number in 0.5-2M of perchloric acid to be 0.018 moles per day, corresponding to a G-value of 3.2 equivalents per 100 eV.

In the radiolysis of solutions, the presence of other ionic species can accelerate or inhibit the disproportionation of plutonium valence states. For example, the presence of the chloride ion in plutonium (VI) solutions prevents reduction to plutonium (IV). Reactions may reverse after long irradiation periods, in which case a steady-state condition should ultimately be reached, resulting in a net decomposition rate of zero. An excellent review of the radiation chemistry of plutonium nitrate solutions may be found in Miner and Seed (1966). In dilute solution (0.1M), G_{H2} is about 0.5 and G_{O2} increases to 1.45. Self-reduction of Plutonium hexafluoride as the solid phase amounts to about 1.5% of the material per day (Weinstock and Malm, 1956). See Cleveland (1970) Chapter 2, for more information.

2.5.2.3 Miscellaneous Radiolytic Reactions

A serious limitation to the use of organic ion exchange materials is their radiation stability. Brookhaven National Laboratory (BNL) reviewed the literature and summarized the effect of ionizing radiation on both organic and inorganic ion exchange materials (Gangwer et al., 1977). Extraction of plutonium (IV) from 3M HNO_3 into 30 vol% tributyl phosphate in kerosene at 5°C decreased the extraction coefficient by a factor of two when irradiated to a dose of 3.6×10^7 R (Tsujino and Ishihara, 1966). The mechanical properties of thin plastic films such as polyethylene and polyvinyl chloride degrade with exposure to plutonium. Cellulose vacuum-cleaner bags will disintegrate in less than a month if used for housekeeping purposes in plutonium-contaminated gloveboxes. Leachability of plutonium-containing wastes could be affected by the production of nitric acid for air-equilibrated dilute salt solutions (Rai et al., 1980).

2.5.2.4 Helium Retention and Release

Helium introduced by alpha-bombardment of plutonium and the alloys and compounds of plutonium can cause lattice expansion. This was first observed for plutonium oxides, carbides, and nitrides by Rand et al. (1962) and was later observed for two plutonium carbide phases. Helium is retained in vitrified compounds. The retention and release behavior of helium in plasma-torch-fused $Pu0_2$ microspheres for SNAP is an important parameter in the design of the heat source. Approximately 530 cm^3 at standard temperature and pressure per year-kg are produced by $^{238}Pu0_2$ (Stark, 1970). Microspheres of 80% $^{238}PuO_2$ and 20% $^{239}PuO_2$ that were approximately 50 mm in diameter, prepared by the sol-gel process, released 92.8% of the helium in 8 months at room temperature (Northrup et al., 1970). Metals at temperatures well below the melting point trap the insoluble helium gas in tiny bubbles, which are more or less evenly distributed through the matrix material (Stevens et al., 1988). Helium buildup in weapon-grade material is approximately 4 standard cm^3 per year-kg.

2.6 OCCUPATIONAL HAZARDS

The major industrial hazard in plutonium facilities is the potential for loss of control of a highly toxic substance, resulting in either the inhalation or ingestion of plutonium or one of its compounds by personnel, or the exposure to excessive radiation from a criticality accident. The possibility of a fire or explosion in a plutonium facility is probably the most serious threat because the consequences of a fire could lead to loss of containment and subsequent dispersal of highly mobile plutonium particulates. In addition, fighting the fire with water to maintain containment could create the potential for a criticality accident and/or loss of containment in the immediate vicinity.

The day-to-day hazards for personnel in plutonium facilities involve exposure to gamma rays, X-rays, and neutrons, as well as possible accumulation of plutonium in the body. These hazards are described in more detail in Section 3.0, “Radiation Protection,” and Section 7.0, “Nuclear Criticality Safety.” The amount of plutonium needed to present potential hazards to personnel in plutonium-handling facilities is summarized in Figure 2.4. Hazards related to interim and long term storage of plutonium will be found in Section 2.7, “Storage and Containment.”

2.6.1 Chemical Versus Radiological Hazards

The radiological toxicity of reactor-produced plutonium far exceeds the chemical toxicity of this heavy element. Furthermore, its low solubility in near-neutral or basic solutions reduces the uptake through ingestion by a factor >1000 for any plutonium compounds except certain complexes, such as the citrate or ethylenediamine tetraacetic acid complex. (Refer to Sections 2.3, “Radiobiological Properties,” and 6.0, “External Dose Control”). Tipton (1960) summarizes the differences in chemical hazards between plutonium and uranium: “In contrast to uranium, the chemical toxicity of plutonium is insignificant in comparison to the hazard arising from its natural radioactivity.” Moreover, “the toxicity of plutonium and other transuranic elements,” according to Voelz et al. (1985), “has only been studied in animals since acute toxicity has never been observed in man for these elements and epidemiologic studies have not produced positive results.” However, recent evidence suggests that plutonium can catalyze reactions including oxidative stress in the absence of significant radioactive decay. These data presented by Claycamp and Luo (1994) suggest that plutonium complexes might contribute to long-term oxidative stress related to tumor promotion.

2.6.2 Associated Chemical Hazards

The main chemical hazard of plutonium is its vulnerability to oxidation and the pyrophoricity of some of its alloys and compounds (see Section 2.6.3).

The processing of plutonium, including separation from irradiated uranium, purification, conversion, waste disposal, environmental restoration, and D&D, necessarily requires the use of chemicals and reagents with varying degrees of toxicity and hazardous properties. A partial list of chemicals that have been used at DOE plutonium facilities is provided in Table 2.9. An abbreviated evaluation of the potential hazards of these substances is also provided. Table 2.9 is not meant to replace the Material Safety Data Sheet available from chemical manufacturers; rather, it is intended to help readers recognize the toxicity of these chemicals and identify any possible side effects from their use that could jeopardize radiation safety or plutonium containment.

2.6.3 Hazards Created by Oxidation and Pyrophoricity

This section describes the oxidation and burning characteristics of plutonium, summarizes the storage properties of the metal and oxides, and presents recommendations for their storage conditions. Waste remediation plans for TRU materials and the necessity for dealing with ton quantities of plutonium metal from the retirement of weapons require the identifying of long-term and intermediate-term waste forms with appropriate stability. Economic considerations make clear the importance of generating few, if any, new wastes in accomplishing this task.

2.6.3.1 Oxidation of Plutonium

The problems of oxidation of metallic plutonium were recognized shortly after the discovery of plutonium, and extensive studies of the low-temperature corrosion of plutonium and its alloys have been performed. Oxidation can produce fine loose plutonium oxide, which disperses easily in glovebox systems, complicating housekeeping chores. If not controlled, loss of accountability and increased radiation exposure to personnel is certain. The reactivity of plutonium metal is discussed in Section 2.3.1. The tendency for enhanced oxidation is promoted by the self-heating properties of plutonium isotopes (discussed in Section 2.5.1). A kilogram of ^{239}Pu can easily reach an equilibrium temperature of 80°C in a glove-box environment (Raynor and Sackman, 1967). Thermally isolated ^{238}Pu metal can easily melt from its own decay heat. The heat generated by oxidation may be sufficient to ignite nearby combustible materials. Metal turnings and scrap should be reprocessed or converted to stable alternatives as soon as practicable. Plutonium metal, its alloys, and its reactive compounds need to be excluded from both oxygen and water vapor, but especially the latter since it catalyzes and accelerates oxidation.

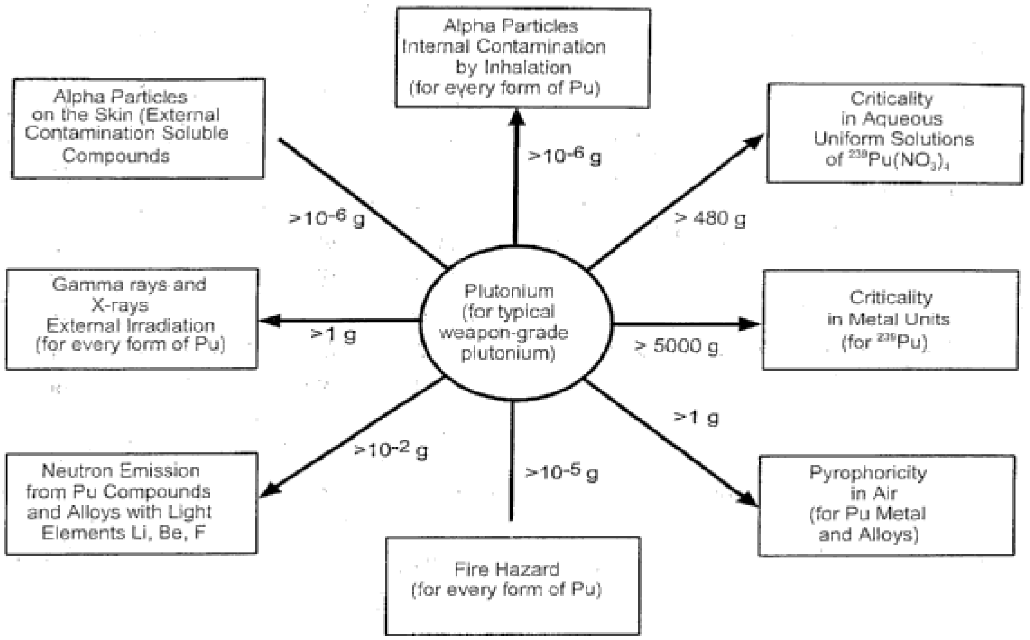

Figure 2.4. Hazards in Low-Exposure Plutonium Handling

Table 2.9. Hazards of Chemicals Used in Processing Plutonium [(a)]

Chemical Name	CAS No.[(b)]	Hazard	Formula
Aluminum nitrate	7784-27-2	--	$AL(NO_3)_3 9H_2O$
Antimony pentafluoride	7783-70-2	--	SbF_5
Beryllium (metal)	7740-41-7	Neutron	Be
Calcium (metal)	7740-70-2	Release H_2 when wet, flammable	Ca
Calcium oxide	1305-78-8	Corrosive	CaO
Calcium chloride	10043-52-4	--	$CaCl_2$
Ferrous ammonium sulfate	7783-85-9	--	$Fe(SO_4)(NH_4)_2 SO_4 6H_2O$
Fluorine	7782-41-4	Oxidizer, poison	F_2
Fluorine dioxide	--	Oxidizer, poison	F_2O_2
Carbon tetrachloride	56-23-5	--	CCl_4
Ferrous sulfamate	--	--	$Fe (SO_3NH_2)_2$
Gallium (metal)	7440-55-3	--	Ga
Hydrogen	1333-74-0	Flammable, explosive	H_2
Hydrogen Fluoride	7664-39-3	Corrosive	HF
Hydrochloric acid	7647-01-0	Corrosive	HCl
Hydrogen peroxide	7722-84-1	Oxidizer	H_2O_2
Iodine	7553-56-2	Poison	I_2
Magnesium (metal)	7439-95-4	Water reactive, flammable explosive, produces neutrons when combined with Pu	Mg
Magnesium chloride	7786-30-3	Neutron	$MgCl_2$
Magnesium oxide	1309-48-4	Neutron	MgO
Mercuric nitrate	10045-94-0	Oxidizer, poison	$Hg(NO_3)_2$
Nitric acid	7697-37-2	Oxidizer, corrosive, poison	HNO_3
Oxalic acid	144-62-7	Poison	$H_2C_2O_4$
Potassium hydroxide	1310-58-3	Corrosive, Poison	KOH
Potassium chloride	7447-40-7	--	KCI
Sodium chloride	7647-40-7	--	NaCl
Sodium Hydroxide	1310-73-2	Corrosive, poison	NaOH
Sodium nitrate	7631-99-4	Oxidizer	$NaNO_3$
Stannous chloride	7772-99-8	--	$SnCl_3$
Sulfamic acid	5329-14-6	Corrosive	NH_2SO_3H
Sulfuric acid	7664-93-9	Corrosive, poison	H_2SO_4
Tri-n-butyl phosphate	126-73-8	Flammable liquid	$(C_4H_7O)_3PO_4$
Urea	57-13-6	--	$CO(NH_2)_2$
Uranium (metal)	--	Flammable	U
Zinc chloride	7646-85-7	--	$ZnCl_2$
Soltrol 170 Phillips 66	68551-19-9	Flammable liquid (isoparafins)	(Mixture C10–C14)
Carbon tetrafluoride	75-73-0	--	CF_4

(a) Refer to Material Safety Data Sheets for complete discussion of hazards.

(b) Chemical Abstracts Service Registry number.

The corrosion or oxidation of plutonium does not always occur in a linear or predictable manner. The oxidation rate is a complex function of the surrounding atmosphere, the moisture content, and the alloys or impurities present in the metallic plutonium.[4]

2.6.3.2 Ignition Temperatures and Pyrophoricity of Plutonium, Its Alloys, and Its Compounds

Plutonium and some of its alloys and compounds are pyrophoric. Pyrophoric material is a liquid or solid that, even in small quantities and without an external ignition source, can ignite within 5 minutes after coming in contact with air (NFPA Fire Protection Handbook). Pyrophoric plutonium metal has been defined as “that metal which will ignite spontaneously in air at a temperature of 150°C (320°F) or below in the absence of external heat, shock, or friction” (Stakebake, 1992).[5] Finely divided plutonium metal would be considered pyrophoric while massive plutonium would be nonpyrophoric. Martz et al. (1994) has proposed a mechanism for plutonium pyrophoricity that predicts the ignition temperature as a function of surface mass ratio and particle size. The most numerous forms of pyrophoric plutonium are chips, lathe turnings, and casting crucible skulls. Plutonium hydride and sesquioxide (Pu_2O_3) are probably the most commonly occurring pyrophoric compounds. Plutonium carbide, oxycarbide, nitride, and oxide phases with compositions between the sesquioxide and dioxide are potentially pyrophoric. Known pyrophoric alloys include Pu-U and Pu-Ce, Waber (1967) summarized much of the early work on plutonium corrosion and oxidation and is a good source for identifying other pryophoric alloys.

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[4] See Wick (1967), Coffinberry and Miner (1961), and Kay and Waldron (1966) for details on the oxidation of unalloyed plutonium and the stabilized alloy of plutonium.

[5] Also in DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

The health physics aspects of an accidental plutonium fire can be serious. A fire can burn through containment structures, resulting in the dispersal of PuO_2 over a wide area, with the potential for inhalation exposure during the fire or during subsequent decontamination efforts. The conditions under which a plutonium fire can occur in a dry glovebox have been studied. With only 5% oxygen in nitrogen, the metal will burn easily. At the 1% level, however, a fire will not continue to burn unless heat is supplied (Rhude, 1962). Turnings shall be generated in a dry atmosphere and should be converted to the oxide as soon as convenient, preferably on the same day they are made. Some solvents and organic compounds form flammable mixtures with plutonium. In one incident, tetrachloroethane was inadvertently substituted for another lathe coolant in a metal-turning operation. Chips of plutonium aluminum alloys were ignited, resulting in the blowout of a glove-box panel. In a separate event, burning plutonium chips dropped into carbon tetrachloride resulted in an explosion (AEC, 1965).

2.6.3.3 Aerolization of Plutonium

The ignition of plutonium metal becomes a major hazard when enough plutonium has burned to produce a significant amount of dispersible material and a serious enough fire to damage the pertinent containment structures. The particle size of PuO_2 fired at a low temperature varies from 3% at <1 µm to 97% at 1-5 µm (Stakebake and Dringman, 1967). Sintered PuO_2 has a particle size <2 µm. Haschke (1992) made an effort to define the maximum value of the source term for plutonium aerosolization during a fuel fire. He found the rate to be constant (0.2-g PuO_2/cm^2 of metal surface per minute) above 500°C. The mass distribution for products of all metal gas distributions are approximately 0.07 mass% of the oxide particles having geometric diameters ≤10 µm.

2.7 STORAGE AND CONTAINMENT

The DOE mission for utilization and storage of nuclear materials has changed as a result of the end of the "Cold War" era. Past and current plutonium storage practices largely reflect a temporary, in-process, or in-use storage condition which shall now be changed to accommodate longer-term storage.

The DOE has sponsored a number of workshops on disposing of plutonium. Two of the objectives of these workshops have been to make recommendations for near-term and long-term storage forms and to identify possible alternatives. At the Hanford Plutonium Disposition Workshop held in Richland, Washington, from June 16 to 18, 1992, the two highest ranking stabilization processes were, first, denitration of plutonium nitrate, and, second, thermal stabilization. The third-ranked process included the precipitation of Cs_2PuC1_6or $K_4Pu(SO_4)_4$ followed by thermal stabilization (Hoyt, 1993). At the workshop on plutonium storage sponsored by DOE Albuquerque, on May 26 and 27, 1993, both metal and oxide were considered suitable storage forms. A report has been issued summarizing information presented here and resulting from this workshop (DOE, 1994a). This important report includes sections on:

-- materials properties relevant to storage;
-- current storage practice;
-- advanced storage concepts;
-- hazard analysis; and
-- recommendations.

Existing storage and handling requirements for plutonium metal and oxides are currently covered in DOE Order 460.1C, Packaging and Transportation Safety (DOE, 2011b). DOE M 441.1-1, Nuclear Material Packaging Manual (DOE, 2008e) and DOE-STD-3013-2012, Stabilization, Packaging, and Storage of Plutonium-Bearing Materials (DOE, 2012a), also provide information on packaging plutonium material.

The following property summaries adapted from Haschke and Martz (1993), are useful for determining potentially unsuitable storage and containment conditions for plutonium metal and oxide. Given that plutonium metal is chemically reactive in air and other environments, it also:

- Exhibits spontaneous self-sustained ignition (becomes pyrophoric) only if the metal dimension

  - is <0.1 mm and T >150°C
  - is >0.2 mm and T >500°C

- reacts slowly in air at room temperature (maximum of about 1 μm/day)

- has limiting (T-independent) oxidation rate in air above 500°C

- is not a dispersible form (<10 μm geometric size) until oxidation occurs:
  - oxide from Pu+Air at ambient T: 100 mass % (ssa = 10-20 m^2/g)
  - oxide from PuH_2+O_2: ~25 mass % (ssa ~ 1 m_2/g)
  - oxide from $Pu+O_2$and Pu+Air at T >500°C: < 0.1 mass % (ssa <0.1 m^2/g)

- radiolytically decomposes organic and covalently bound specific species in the environment

- reacts with most radiolytically produced gases and with nonequilibrium surface:
  - limits pressurization by gases
  - forms low-density (pressure-generating) and pyrophoric products

- retains helium from alpha decay

- is stabilized by certain storage atmospheres (reactivity decreased by 10^{12})

- is stable if isolated from reactive species

- has good storage history when stored properly.

A similar property summary for plutonium dioxide, the most commonly used form of plutonium, shows it to be stable and unreactive in air. Storage and containment recommendations, based on the properties of plutonium metal and dioxide, are shown in Table 2.10.

Table 2.10. Storage Recommendations for Plutonium Metal Dioxide (adapted from Haschke and Martz, 1993)

- Metal and oxide are both suitable storage forms for plutonium (100 years).

- Organics (plastics, elastomers) shall be excluded from the primary container for both forms.

- Converting between metal and oxide is not recommended (negative impact of waste, cost, environmental safety and health risk).

- Both forms shall be properly prepared and certified:
  - Procedures for metal already exist (technology transfer needed).
  - Procedures for oxide need development (stabilization, desorption, loss on ignition.

- Both forms shall be in sealed primary containers for extended storage:
  - Positive seals (e.g., welds and metal seals) are necessary.
  - Seal certification or double sealing is necessary.

- Requirements diverge for short-term/retrievable storage:
  - Containers with metal gaskets are advantageous for metal storage.
  - After stabilization, oxide is best stored in a container fitted with a rupture disk in series with a vented stainless-steel frit container.

- Surveillance of stored materials is required.

Note: DOE M 441.1-1, Nuclear Material Packaging Manual (DOE, 2008e) and DOE-STD-3013-2012, Stabilization, Packaging, and Storage of Plutonium-Bearing Materials (DOE, 2012a), also provide information on packaging plutonium material.

3.0 RADIATION PROTECTION

The radiation protection field is concerned with the protection of individuals, their progeny, and humanity as a whole, while still allowing for necessary activities which might involve radiation exposure. The aim of radiation protection is to prevent deterministic effects and to limit the probability of stochastic effects. Most decisions about human activities are based on an implicit form of balancing risks and benefits leading to the conclusion of whether or not the application of a particular practice produces a positive net benefit. Because the probability of health effects is not zero, the ICRP in Publication 26 (ICRP, 1977) recommended the following criteria for a system of dose limitation:

-- No practice shall be adopted unless its introduction produces a positive net benefit.

-- All exposures shall be kept as low as reasonably achievable, with economic and social factors being taken into account.

-- The dose equivalent to individuals shall not exceed the limits recommended for the appropriate circumstances.

These criteria and related information have been incorporated into DOE regulations, instructions, and manuals for radiation protection.

The successful operation of a plutonium facility requires scrupulous attention to providing adequate radiation protection and maintaining contamination control through the implementation of a quality health physics program. (In this section, “health physics” and “radiation protection” can be used interchangeably when referring to programs or personnel.) Prompt dose assessment is important for demonstrating compliance with standards, providing information to workers, establishing an accurate historical record, and for responding to accident and incident situations. This section defines the basis for the establishment of a sound health physics program at a plutonium facility.

3.1 REGULATION AND STANDARDS

Regulations on radiation protection in DOE and DOE contractor facilities are found in 10 CFR 835, Occupational Radiation Protection: Final Rule (DOE, 2007a). Guidance is found in the supporting document Radiological Control (DOE, 2008a) and the 10 CFR 835 Implementation Guide G 441.1-1C, Ch 1 (DOE, 2011a).

10 CFR 851 specifies health and safety regulations, which also apply to workers in DOE facilities. Other related source documents include publications of the U. S. Environmental Protection Agency (EPA), American National Standards Institute (ANSI), ICRP, National Council on Radiation Protection and Measurements (NCRP), and United Nations Scientific Committee on the Effects of Atomic Radiation. 10 CFR 851, Worker Safety and Health Program (DOE, 2006a) provides requirements for worker safety and health. The worker safety and health program integrates the Rule’s requirements with other site worker protection activities and the integrated safety management system (ISMS) [851.11(a)(3)(ii)].

In addition, each site that handles radioactive materials and/or radiation generating machines is required to establish and maintain its own documented radiation protection program, following the Federal regulations.

3.2 RADIATION PROTECTION PROGRAMS

Radiation protection programs include provisions for quality assurance, administrative controls, protection of visitors, visits by regulatory personnel, and onsite packaging and transportation of hazardous materials.

3.2.1 Quality Assurance

It is highly desirable for laboratories and industrial facilities handling plutonium to have a well-integrated quality assurance program. Such a program should have high visibility and strong management support. Quality assurance should be effectively applied throughout facility activities, including the radiation protection program. The basis for quality assurance programs in DOE facilities is established in 10 CFR 830, Nuclear Safety Management (DOE, 2001). In addition, 10 CFR 830.120, Quality Assurance Requirement, requires the development of a Quality Assurance Program, specifies an implementation schedule, and provides the elements that the program shall address.

An effective quality assurance program for radiation protection will include establishment of appropriate standards of performance for essential activities and equipment, with an effective system of documentation and traceability of those activities and of the use of the equipment. Proper maintenance of those records will be necessary for reference purposes.

3.2.2 Administrative Controls

In any facility that handles radioactive materials, the major controls protecting workers, the public, and the environment are structures and installed equipment, which shield, contain, and confine the radioactive materials. However, to allow useful work to be performed in the facility and to assure that its protective features remain effective, a number of administrative controls are ordinarily required. These administrative controls are usually contained in a series of procedures related to the operations and maintenance activities to be carried out in the facility. All personnel who work in controlled areas should be familiar with the administrative controls that apply to their work. When changes or additions to administrative controls are made, these changes or additions should be effectively communicated to all persons who may be affected.

3.2.2.1 Radiation Protection Procedures

A plutonium facility should have a written policy on radiation protection, including a policy on keeping exposures as low as reasonably achievable (ALARA). All radiation protection procedures and controls should have formal, recognizable technical bases for limits, methods, and personnel protection standards. Procedures should be adequately documented, updated periodically, and maintained in a centralized historical file. A control system should be established to account for all copies and ensure that all new procedures are included in the historical files. A designated period of time for maintaining historical files should be established. DOE Order 200.1A, Information Technology Management (DOE, 2008b) and ANSI/HPS N13.6 (ANSI, 2010) provide guidance on how to maintain historical files. In addition, radiation protection procedures should have a documented approval system and established intervals for review and/or revision. A tracking system should be developed to ensure that the required reviews and revisions occur.

Radiation protection procedures should be provided for but not limited to the following topics:

- Posting and labeling of facilities
- development and maintenance of all radiation protection records
- reporting of unusual radiation occurrences
- use of radiation monitoring instruments
- use of radiation sources (e.g., reference calibration)
- reporting of radiation exposures
- use of protective clothing
- responding to radiological emergency events
- surveying and monitoring
- counting room equipment and use
- instrument maintenance and control
- development and use of Radiological Work Permits (RWPs)
- responsibilities of operations staff for contamination control and personnel surveys.

Two topics, RWPs and facility posting and labeling, are discussed below in more detail.

3.2.2.2 Radiological Work Permits

10 CFR 835.501(d) requires written authorizations to control entry into and perform work within radiological areas. These authorizations shall specify radiation protection measures commensurate with the existing and potential hazards. Radiological Work Permits are a type of written authorization used for entry into high and very high radiation areas, high contamination areas, and airborne radioactivity areas. The RWPs also are used to control entry into radiation and contamination areas and for handling materials with removable contamination. The RWPs should be initiated by the work group responsible for the activity. All RWPs should be reviewed and approved by the radiation protection staff. Radiological

Work Permits are recommended for other radiological work in accordance with the standard, Radiological Control, (DOE, 2008a). Guidance for posting of RWPs and for their contents is contained in the standard, Radiological Control.

Radiological workers should read and understand the applicable RWP before performing work in a radiological area. The RWPs should be located at the access point to the applicable radiological work area. Workers should acknowledge by signature or through electronic means that they have read, understood, and will comply with the RWP before they initially enter the area and after changes. Out-of-date RWPs should be removed.

3.2.2.3 Radiological Surveys and Data Trending

Area monitoring in the workplace shall be routinely performed, as necessary, to identify and control potential sources of personnel exposure (10 CFR 835.401(a) (6)). This monitoring should include surveys in areas that are not ordinarily expected to be contaminated. The program should define minimum requirements, survey types, and frequencies.

Surveys should be performed at frequencies adequate to identify changes in posting required or an activity buildup, and to ensure that current radiological controls are appropriate. The surveys suggested by this section are minimum recommendations; additional surveys should be conducted, recorded, and reviewed as necessary to ensure full protection of personnel.

Contamination surveys should be performed to determine contamination area (CA) boundaries, the appropriate posting of sources or areas, and the location and extent of localized contamination.

Contamination surveys should be performed and documented prior to the start of radiological work, during general work activities at times when changes in contamination level may occur, and following work to assure that final radiological conditions are acceptable and documented. See Munson et al. (1988).

A sufficient number of points should be surveyed to adequately assess the radiological status of the area being surveyed.

Routine radiological surveys should be regularly conducted, recorded, and reviewed for all areas where personnel could be exposed to alpha, beta, gamma, X-ray, or neutron radiation throughout the site. Surveys should be performed at frequencies adequate to ensure protection of personnel. The following surveys should be considered the minimum. Additional surveys should be conducted, recorded, and reviewed as necessary to ensure that personnel exposures are maintained ALARA.

General radiation and contamination surveys should be performed:

-- To identify and verify the boundaries of areas which shall be radiologically controlled.

-- to verify that radiation and contamination Levels outside of radiological areas remain less than specified limits.

-- to determine the appropriate posting of localized higher radiation levels, beams, or hot spots.

-- to ensure that radiological conditions are acceptable and documented prior to, during, and at the completion of work that may cause changes in radiation levels to occur (see Munson et al., 1988, p. 6.1.2).

-- to satisfy required predetermined procedure hold-points in work areas and adjacent areas, whenever operations are performed that may cause significant increases in radiation levels. The survey may be required as part of a radiological inspection step required by the work procedure. This includes areas above and below the work area as appropriate during special processing operations or cell decontamination, movement of permanent or temporary shielding, radioactive waste processing, and relocation of highly radioactive materials.

Routine radiation and contamination level surveys should be performed in the workplace at a frequency commensurate with the radiation hazard, to detect trends related to equipment, systems, environment, and work habits.

Non-routine surveys of radiation and contamination levels in the workplace should be performed:

-- Before initial use of a new installation, system, or equipment, or as soon as possible after a radiation source is brought into the area.

-- whenever changes in procedures, equipment, or sources have occurred that may cause changes in the external radiation levels.

-- after modification to a shield or changes in shield materials.

-- as the basis for trend evaluation of external radiation level conditions.

-- when a radiological accident has occurred or is suspected.

-- when requested by the personnel performing the activity (see Munson et al., 1988, p. 6.1.2).

Radiation surveys should be performed upon initial entry into process cells and tanks that contain radioactive piping or components.

Surveys should be conducted when performing operations that might result in personnel being exposed to small intense beams of radiation (e.g., removing shielding for shielded X-ray devices).

Every reasonable effort should be made to maintain the radiation dose of the surveyor at levels that conform to ALARA guidance.

Surveys should be performed and documented according to established procedures.

Only fully trained and qualified personnel should conduct surveys that are to be the official records of radiation levels or for the protection of personnel; these surveys should be reviewed and approved by the Radiological Protection Manager or his/her designee.

Surveys should be performed with calibrated instrumentation appropriate for the intensity and energy of the radiation anticipated in the area to be surveyed.

Survey instruments should meet the performance check requirements established by the facility in accordance with ANSI N323a (ANSI, 1997b).

Combinations of survey instruments should be used as necessary to provide the capability to measure all types of radiation and dose rates characteristic of that which could be encountered at the facility being surveyed.

Records that establish the conditions under which individuals were exposed to external radiation (such as facility radiological conditions records generated by the monitoring programs) should be retained to provide a chronological and historical record. See ANSI/HPS-N13.6 (ANSI, 2010).

A sufficient number of points should be surveyed in order to adequately assess the radiological status of the area. Regular predetermined points may be used, but additional spot monitoring should be done to ensure that all changes in dose rates are identified, recorded, and reviewed.

All records of surveys should clearly identify, as a minimum:

- The name, signature, and employee number of the surveyor.
- survey instrument(s) model number, serial number, and calibration date.
- the type(s) of radiation being monitored (e.g., neutron, gamma, etc.).
- the dose rates.
- the date and time the survey was performed.

- locations where radioactive material is located temporarily (or is being temporarily stored) or where equipment that generates ionizing radiation is being operated.

Records of the results of radiation surveys should be retained in accordance with facility policy.

Survey data should be reviewed by supervisory personnel. Significant findings should be presented to the facility manager in a timely manner.

Health physics personnel should summarize survey data in each building or area at least once a quarter. Significant changes or trends in area dose rates and/or radiological contamination should be noted and corrective actions assigned. The survey summary should be presented to the facility management quarterly.

Survey results and data summaries should be made available to the ALARA team chair periodically and should be used:

- To provide a basis for evaluating potential worker exposure on a job and in ALARA preplanning.

- to provide a baseline for trend analysis, investigation, and correction of unusual conditions.

- to track the status of jobs (including identification of good practices) and to detect departures from good operating procedures and/or the failure of radiation controls.

- to identify the origin of radiation exposures in the plant by location, system, or component.

Health physics personnel should post the results of radiation surveys or survey maps at the entrance to all permanent radiation areas, high radiation areas, and very high radiation areas. The results should be posted in the form of a survey map so that personnel can be aware of the locations of higher and lower levels of radiation within the area.

A survey data trending program should be conducted; to indicate the continuing effectiveness of existing control; to warn of deterioration of control equipment or effectiveness of operating procedures; to show long-term variations in radiation levels; and to identify and correct improper radiation work practices.

Health physics should perform trend analyses on all permanent radiation, high radiation, and very high radiation areas. At a minimum, one complete survey record should be evaluated and included in the trend analysis program for each survey required to be performed by the facility routine control program.

Health physics should use the facility reporting system to identify discrepancies and abnormal trends and should summarize the data review results in their monthly reports to the Radiological Protection Manager.

Survey data trends should be investigated when either:

- -- an upward trend in general area radiation level occurs, causing a significant increase.
- -- an abrupt change in radiation level occurs that cannot be directly correlated to normal activities.

3.2.2.4 Facility Posting and Labeling

Areas in plutonium facilities shall be posted in accordance with the requirements in 10 CFR 835 (DOE, 2007a). Chapter 12 of Implementation Guide G 441.1-1C, Ch. 1 (DOE, 2011a) provides guidance to ensure compliance. The technical criteria and dose rate and/or levels for defining radiation, high radiation, very high radiation, contamination, high contamination, and airborne radioactivity areas are established in 10 CFR 835. The health physics staff should identify:

- -- Areas to be barricaded and marked to prevent personnel from inadvertently entering them.
- -- Areas to be physically controlled per 10 CFR 835, Subpart F.

Entrance to radiological areas shall be controlled (10 CFR 835.501(a and b)) commensurate with the existing and potential radiological hazard within the area.

The health physics staff should post current radiation surveys of radiation areas at the health physics access control point for use in prejob planning. Airborne Radioactivity Areas shall be posted with the words, “Caution, Airborne Radioactivity Area” or “Danger, Airborne Radioactivity Area” when the airborne radioactivity levels in the occupied area exceed, or are likely to exceed, the derived air concentration (DAC) value listed in Appendix A or Appendix C of 10 CFR 835 or where an individual could receive 12 DAC hours in a week (10 CFR 835.603(d)). These areas are posted to alert personnel of possible respiratory protection requirements.

3.2.2.5 Unposted Areas

Certain areas of facilities that handle radioactive materials should be maintained free of detectable radioactive contamination. These areas should also be maintained at ambient radiation levels equivalent to the environmental background of the facility. Parts of the facility that should meet these requirements include lunchrooms, offices, restrooms, janitor rooms, corridors outside operational areas, foyers, and outside areas surrounding the facility, including the building roofs.

To assure these areas meet the requirements of radiological cleanliness, they should be surveyed with count-rate instruments sensitive to the radioactive isotopes of interest. In a plutonium facility, the instruments should meet the requirements listed in ANSI Standard N317-1991, Performance Criteria for Instrumentation Used for In-Plant Plutonium Monitoring (ANSI, 1980a). These clean areas should be maintained below the surface contamination levels cited in 10 CFR 835 (DOE, 2007a).

3.2.3 Visitors

Regardless of the general radiation safety knowledge of visitors to a plutonium facility, they should be escorted at all times when they go into the posted areas of the plant. In addition, before going into such an area, they should be given a general orientation to the facility radiation protection program and informed about the potential radiation conditions in the areas where they will be going. They also should be provided with the same protective devices worn by facility personnel engaged in similar activities.

Visitors with a demonstrated need to enter the following areas may be allowed access if such access is controlled with a combination of training and the use of escorts trained for the specific area:

-- Radiological Buffer Areas
-- Radiation and High Radiation Areas
-- Contamination Areas
-- Radioactive Material Areas

Guidance for training for visitors is provided in the standard, Radiological Control, (DOE, 2008a), Article 622:

-- Persons under 18 years of age should not be permitted to enter Radiation Areas or Contamination Areas without the approval of the Radiological Protection Manager.

-- Area entry requirements and access restrictions for visitors should be in accordance with established facility procedures.

-- Individuals, visitors included, shall (10 CFR 835.502(b)) be prevented from entering Very High Radiation Areas when dose rates are in excess of the posting requirements of 10 CFR 835.603(c), and visitors should be prohibited from accessing High Contamination and Airborne Radioactivity Areas.

In addition the following is recommended:

All facility personnel serving as a qualified escort should ensure that each visitor under his/her cognizance completes a facility radiological visitor form. The qualified escort should also sign the visitor form and complete it as appropriate.

Facility-sponsored visitors should provide the following before entering radiological areas, unless these records have already been entered into the facility entry control system:

-- Evidence of completing required training, as applicable.

-- visitor radiation exposure disclosure.

The host facility manager should forward the visitor radiation exposure and medical disclosure forms to Dosimetry.

The use of offsite respirator fit test certification may be authorized (if in accordance with the applicable Radiation Protection Program) under the following conditions:

-- A respirator fit test has been completed within the previous year.

-- The individual presenting the respirator fit test certification card has not changed physical appearance in a way that would affect the seal of the respirator facepiece to the face.

-- The facility has the respirator facepieces available that the individual is certified to wear.

3.2.4 Visits by Regulatory Personnel

Periodically, personnel from the DOE and other Federal and state agencies visit radiation facilities for audit purposes or to discuss regulatory changes. In most cases, they will want to look at records of the radiation protection program and, in some cases, will also want to enter posted areas of the facility. They should have ready access to the facility provided that dosimetry and other requirements are met. They should have complete access to facility personnel knowledgeable in the subjects they wish to discuss.

3.2.5 Onsite Packaging and Transportation

DOE Order 460.1C (DOE, 2011b) establishes safety requirements for the proper packaging and transportation of DOE, including NNSA, offsite shipments and onsite transfers of radioactive and other hazardous materials and for modal transportation. DOE M 441.1-1, Nuclear Material Packaging Manual (DOE, 2008e) and DOE-STD-3013-2012, Stabilization, Packaging, and Storage of Plutonium-Bearing Materials (DOE, 2012a), also provide information on packaging plutonium material

3.3 RADIOLOGICAL CONTROL ORGANIZATIONS

The radiological control organization shall be structured so that all of the activities required to provide support to line management and workers can be accomplished.

3.3.1 Management Commitment

Management commitment to safety is the most important characteristic of an effective safety program, including a radiological control program. If the management commitment to safety is strong, the radiological control program will be valued and respected. The radiological control program should be provided adequate authority to permit performance of necessary assignments and program implementation. Management commitment to the ALARA concept is particularly important [see Article 111, Radiological Control (DOE, 2008a)]. Adequate personnel, equipment, and funding should be available as a part of this commitment.

3.3.2 Radiological Control Organization Independence and Reporting Level

The radiological control organization should be independent of the line organization responsible for production, operation, or research activities and should have an equivalent reporting level. Because health physics personnel should have the authority to balance operations with safety, they should not report directly to the administrators of operations. When shift work is involved, the operations shift supervisor may make minor health physics decisions in support of the shift's Radiological Control Technicians (RCTs); however, decisions involving basic policies and procedures should be directed to a separate health physics organization.

If a safety organization includes the health physics program, it should be high enough in the company to assure direct access to the company president or equivalent. If the health physics program is administered by a separate radiological control organization, that organization should also be in a position to assure direct access to the company president. This is to safeguard the program from the pressures of production that exist in the operational environment, by keeping it independent of operating organizations.

A system of guides, policies, and procedures should be established to clearly identify the interrelationships, responsibilities, and authorities of those involved with the development, operation, and maintenance of the facility and the health and safety of the employees. These guides, policies, and procedures should be documented and should be reviewed at least once every year.

3.3.3 Adequacy of Personnel and Equipment

A sufficient number of qualified and, where required, certified radiological control personnel should be available to perform necessary tasks for support of plutonium facility startup and operation (See Section 3.4 for guidance concerning staffing and staff qualifications). Sufficient equipment, including protective clothing, respiratory protective equipment, and radiation detection instrumentation should be available to support RCTs and operating personnel in the performance of work in controlled areas.

3.3.4 Assignment of ALARA Responsibility and Authority

Limiting radiation exposures to the lowest levels commensurate with the benefit of the work to be accomplished has long been a part of health physics and radiological protection programs of DOE and its contractors. 10 CFR 835 (DOE, 2007a) establishes the policy of maintaining ALARA exposures of workers and the public to radiation from DOE operations. Procedures are required to be prepared (10 CFR 835.104) and implemented and records shall be maintained as required by 10 CFR 835.701 to demonstrate the implementation of ALARA. The DOE standard, Radiological Control (DOE, 2008a), provides additional guidance. Munson et al. (1988) and Chapter 4 of Implementation Guide G 441.1-1C, Ch. 1 (DOE, 2011a), may be used in developing an ALARA program.

An ALARA committee should be established at the plutonium facility. The membership should include managers and workers from the line, the technical support organization, and the radiological control organization. A line manager, such as Director of Operations, Research, Training, or Maintenance should serve as the committee chair. The ALARA committee should make recommendations to management to improve progress toward minimizing radiation exposure and radiological releases (DOE, 2008a).

3.4 STAFFING AND STAFF QUALIFICATIONS

A cadre of operating and maintenance personnel that has experience in the operation of a plutonium facility should be established during the construction of a new facility. The remainder of the operating and maintenance staff should be hired as soon as possible and should receive formal and informal training from the experienced personnel. This step is extremely important to enable all personnel to grow with the facility and learn the details of the operations. Once operations start, potential problems already should have been identified and engineering or administrative changes should have been made to resolve them.

Staffing in the radiological control organization requires technicians and professionals in many support areas. A successful health physics program is highly dependent on the availability of adequate staff support in areas such as environmental monitoring, instrument maintenance and calibration, internal and external dosimetry, meteorology, safety analysis, and risk management.

3.4.1 Professional Staffing and Qualifications

The senior staff of the radiological control organization should include health physicists and other professionals with four-year degrees in science or engineering. A continuing training program should be established for facility personnel. Pursuit of certification by the American Board of Health Physics for senior and professional staff members is encouraged (DOE, 2008a).

At least one professional staff member at the plutonium facility should have a minimum of three years of health physics experience in the operation of plutonium facilities.

3.4.2 Technician Staffing and Qualifications

Recommendations for minimum entry-level requirements for RCTs are given in the DOE standard, Radiological Control (DOE, 2008a), and DOE STD-1122-99, Radiological Control Technician Training, (DOE, 2009a). They include a high school education or equivalency and knowledge of certain scientific fundamentals. If a two-year degree in nuclear technology or an equivalent discipline is locally available, completion of such a program should be encouraged.

Where possible, RCTs and other members of the health physics staff should have a minimum of one year's experience working at a plutonium facility. Such experience is an important prerequisite to allowing them to work unsupervised. Personnel hired without such experience should work an internship of six months under the leadership of a qualified RCT or supervisor with experience in that facility.

The RCTs should be encouraged to pursue registration by the National Registry of Radiation Protection Technologists.

3.4.3 Staffing Levels

At least one professional health physicist is recommended to be on the staff of each major plutonium facility as a full-time employee.

There is no rule of thumb for determining the number of RCTs needed for a given plutonium facility. The number of RCTs should be based on an analysis that provides for sufficient coverage on each shift, given the number of samples, surveys, and other work to be performed; the time of training, donning and doffing protective clothing; shift turnover procedures; and other similar considerations. The site collective dose and individual dose limits in the facility may also lead to the need for additional personnel. Consideration should be given to having sufficient personnel to respond to off-normal conditions and emergencies as well as routine work. Major maintenance, modifications, or decommissioning activities may require additional personnel.

3.5 INSTRUMENTATION CONSIDERATIONS

The radiation from the radioactive decay of plutonium includes alpha, beta, gamma, X-ray (photons), and neutron radiation. An effective monitoring program for plutonium requires radiation detection instruments that are responsive to all of these forms of radiation. It is essential that instruments meet the performance criteria outlined in the applicable U.S. and international standards and be properly calibrated for their intended use.

3.5.1 Types of Instruments and Measurements

Alpha-sensitive instruments are necessary for most contamination control surveys. Exposure rate surveys are normally conducted with photon-sensitive instruments with known energy responses. Neutron surveys become important when processing tens of grams of ^{238}Pu or hundreds of grams of mixed isotopes of plutonium, particularly compounds (i.e., PuO_2, PuF_4, etc.). The neutron survey is important in instances where photon shields, such as leaded glass, are used; such shields normally stop all of the charged particles, most of the low-energy photons, and essentially none of the neutrons. Under these circumstances, neutron radiation is likely to be the major contributor to whole body dose. See the 10 CFR 835 Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a) for a discussion of acceptable approaches for evaluating neutron radiation levels using the radiation weighting factors from the 2007 amendment to 10 CFR 835 (DOE, 2007a).

Continuous air monitors (CAMs) are used extensively in plutonium facilities. Continuous air monitors and sample extraction lines that go to CAMs and continuous radiation dose monitors should be placed outside the glove boxes and hoods. In-line processing instrumentation is critical to accurately monitor the work stations and a review should be performed to determine instrument locations. Continuous air monitors may not have adequate detection capabilities for real-time monitoring at the DAC level. For ^{239}Pu, the annual limit on intake (ALI) is 12 nCi for absorption type M compounds based on the DAC of 5×10^{-12} μCi/mL, as given in Appendix A to 10 CFR Part 835 (DOE, 2007a). DOE G 441.1-1C, Ch. 1 recommends that real-time air monitors be capable of measuring 1 DAC when averaged over 8 hours (8 DAC-hours) under laboratory conditions. Alarm set points for real-time air monitors used for routine monitoring should be set at the lowest practical level so as to accurately indicate loss of containment or the need for corrective action without causing a significant number of false alarms. When monitoring for alpha emitters in areas with high radon concentrations an alarm set point greater than 8 DAC-hours may be necessary.

Continuous air monitors (CAMs) typically have had poor large-particle response due to particle loss during transport to the filter inside the system. Newer alpha air monitors are able to handle large particles more efficiently. Background levels of radon-thoron decay products may be present in concentrations up to 50 to 100 times greater than the level of plutonium of interest. If calibrated properly, alpha CAMs will subtract background levels of radon-thoron decay products; however, in practice the detection limit for plutonium may be as high as 40 DAC-h in the presence of high radon levels. A new generation of alpha CAMs is able to compensate for radon more effectively and meet the desired 8 DAC-h alarm level.

Transuranic aerosol measurement units have been developed and adapted to be used in the workplace. These units avoid preferential plate-out of larger particles by using an in-line filter. Higher flow rates than those normally used with CAMs may be used. Increased detection is obtained on a quasi-real-time basis by high-volume air sampling and counting in a separate vacuum chamber. Detection levels of less than 0.5 DAC-h have been quoted for these units. It has been demonstrated that high-volume impact samplers used at some facilities have demonstrated detection capabilities of 0.1 DAC-h in the laboratory and 1 DAC-h in the field. Other monitoring systems that use diffusion, impaction, or electronic discrimination to reduce the effect of background resulting in an increased detection capability have also been used and are being improved upon. However, it is suggested that site-specific testing be performed on any new equipment to ensure compatibility and verify expected performance. See the Health Physics Manual of Good Practices for the Prompt Detection of Airborne Plutonium in the Workplace (Mishima et al., 1988) for additional information on the selection, placement, and operation of plutonium air monitors.

3.5.2 General Performance Criteria for Instruments

Programs for in-plant monitoring of plutonium consist mainly of airborne and surface contamination surveys and dose rate surveys. The general and specific performance criteria for the instrumentation needed to conduct these programs are described in ANSI N317-1991 (ANSI, 1980a). Performance specifications are also given in ANSI N323 (ANSI, 1997b), ANSI N42.17A (ANSI, 2003), and ANSI N42.17C-1989 (ANSI, 1987b) for portable health physics instrumentation and IEC Publication 325 (IEC, 1981) for alpha and beta contamination meters and monitors. Criteria for air monitoring instrumentation are contained in ANSI N13.1 (ANSI, 2011a), IEC Publication 761-2 and draft IEC Publication 761-6 (IEC, 1983), and ANSI N42.17B-1989 (ANSI, 1987a). Criticality alarm systems are discussed in ANSI/ANS 8.3-1986 (ANSI, 1997c). The criteria discussed in the following subsections are specified in these standards as referenced.

3.5.2.1 Portable Survey Instruments

ANSI N317 (ANSI, 1980a) discusses several criteria related to the performance of portable survey instruments; these include the following requirements:

- The overall accuracy shall be within ±20%, and the precision shall be within ±10% at the 95% confidence level.
- The response time (i.e., the time for the instrument reading to go from zero to 90% of full scale) shall be < 10 seconds on the most sensitive scale and < 2 seconds at readings of 100 mrem/h, 100 mR/h, and 500 dpm or greater. (This criterion is unrealistic with current neutron instrument capabilities. Response time is typically 30 to 60 seconds.)
- The instrument shall be able to maintain accuracy and precision for a minimum of 24 hours of continuous operation.
- The instrument shall have a minimum battery lifetime of 200 hours of continuous operation.

ANSI N42.17A (ANSI, 1988a) specifications differ slightly.

- The response of the instrument shall not change by more than ±15% from a reference value taken at 20°C over the anticipated temperature range for operation.

-- The instrument system shall function within specifications over all anticipated combinations of temperature and humidity (e.g., 15° to 65°C, 40% to 95% relative humidity).

Photon survey instruments should meet the accuracy requirements stated in ANSI N317 (ANSI, 1980a) over the energy range of 0.01 to 1.25 MeV. The angular response of this type of instrument should be within ±15% over a 2π steradian frontal direction using at least two photon sources with energies ranging from 0.06 to 1.25 MeV. Experience has shown that this response specification is not met by most instruments at lower energies due to attenuation of the photon. The energy dependence should be within ±15% over the range of very low energy to 1.25 MeV and the operating range should be from 0.5 mR/h to at least 5000 mR/h.

Experience has shown that ±20% over very low energy to 1.25 MeV is more realistic. This specification applies to a specific window selection (e.g., below 0.05 MeV, the electron equilibrium cap or beta shield shall be removed).

ANSI N42.17A (ANSI, 2003) has a broader scope than ANSI N317 (ANSI, 1980a) but the criteria in it apply to portable survey instruments. Additional criteria include geotropism (maximum change of 6% from reference reading for all orientations), temperature shock (a critical parameter for instruments stored inside, but used outdoors during very cold or hot periods), mechanical shock, vibration, and ambient pressure (maximum change of 15% from reference reading for the latter four criteria). Some differences exist between ANSI N42.17A and ANSI N317. In most cases, the criteria for ANSI N42.17A are more applicable because these criteria are based on substantial testing, which was sponsored by DOE. In ANSI N42.17A, precision is tied into a measurement level; for example, it quotes a precision of 15% at <500 cpm and 10% at >500 cpm. Also, with the advent of liquid crystal displays and other digital readouts, “response time” is defined as the time it takes for the reading to move from 10% to 90% of the equilibrium or steady-state reading. Another significant difference in the standard is that the battery lifetime specification is 100 hours instead of the 200 hours mentioned in ANSI N317.

For direct alpha contamination surveys, the use of audible signals (headphones or speaker) greatly facilitates the detection of “hot spots.”

IEC Publication 325 (IEC, 1981) provides additional guidance on the uniformity of probe response for alpha and beta contamination meters. Surface sensitivity measurements are also discussed in this standard.

3.5.2.2 Performance Criteria for Fixed Monitoring Instruments

Airborne contamination monitors, surface contamination monitors, photon and neutron area monitors, and emergency instrumentation are fixed monitoring instruments subject to the following standard performance criteria.

Airborne Contamination Monitors. Airborne contamination monitors, normally CAMS (see Section 3.5.1), should meet the following criteria according to ANSI N317 (ANSI, 1980a). The primary purpose of any CAM is to detect the presence of airborne radioactivity and activate an alarm to warn personnel in the area so that actions can be taken to minimize personnel exposures. The goal for any CAM should be to perform this function as quickly as possible and at the lowest detectable level of radioactive airborne concentration. The quantity of airborne radioactivity that will result in an alarm within a given time interval is defined in units of DAC-h for a particular radionuclide and is a function of the nuclide's airborne concentration in DACs, the sampling rate, the lower limit of detection of the instrument, and the time needed for the alarm to occur. Mishima et al. (1988) provides guidance on each of these functions.

The minimum detection level of ^{239}Pu, in terms of derived air concentration (DAC), should be 8 DAC-h at the point of sampling in the presence of nominal amounts of naturally occurring alpha-emitters such as radon and thoron and their decay products. (No guidance is provided on what a “nominal” amount is, however.) The operating range should be at least 100 minimum detection levels (i.e., up to 800 DAC-h for ^{239}Pu). Instrument error should not exceed ±20% of the reading over the upper 80% of the operating range. The reproducibility of the system for any given measurement should be within ±10% at the 95% confidence level for a mid-scale or mid-decade reading. The instrument should be capable of operating with less than a 5% change in calibration over the ambient temperature range expected. The instrument should be equipped with an adjustable alarm set point (audible and visible alarms) that can be set at any point over the stated range. The air flow rate should be indicated and adjustable. Voltage and frequency variations of ±15% within design values should result in reading variations of no greater than 5% at the minimum detection level.

ANSI N42.17B (ANSI, 1987a) provides additional performance criteria for air monitors used to detect plutonium. This standard provides specifications for general criteria (sampler design, units of readout, alarm threshold, etc.), electronic criteria (alarms, stability, response time, coefficient of variation, and line noise susceptibility), radiation response, interfering responses (radiofrequency, microwave, electrostatic, and magnetic fields), environmental criteria (temperature, humidity, and pressure), and air-circuit criteria. More detailed specifications are provided in ANSI N42.17B than in ANSI N317 (ANSI, 1980a); however, the environmental criteria and the limits of variation are not as restrictive as those in ANSI N317. With respect to accuracy, ANSI N317 requires less than ±20%, and ANSI N42.17B requires 40% at the 95% confidence level. For the environmental criteria, ANSI N317 requires that the readings change less than 5% under ambient conditions, while ANSI N42.17B gives a 15% limit of variation. As discussed previously, criteria from ANSI N42.17B are more applicable because they are supported by instrument testing.

ANSI N13.1 (ANSI, 2011a) provides detailed guidance on sampling methods. One criterion that relates to CAMs is that air sample lines between air inlet and filter media are to be eliminated where possible; where not possible, they are to be designed to meet the sampling criteria contained in the standard (e.g., short lines, proper sampling rate, smooth bends). The use of Tygon tubing as sample lines before the collection filter should be minimized or eliminated. Air in-leakage from surrounding areas can be a problem when using sampling lines. Testing for air in-leakage shall be performed at least annually or when seals or "O" rings are replaced.

Surface Contamination Monitors. Surface contamination monitors include hand and/or shoe counters and instruments (or probes) with sufficient flexibility to survey pieces of equipment, including exterior clothing. ANSI N317 (ANSI, 1980a) states that these instruments shall have an audible alarm, a frequency that is proportional to the count rate, or a preselectable trip setting, and that upon reaching that level shall activate an audible or visible alarm or both. These instruments should be calibrated according to the requirements in ANSI N323 (ANSI, 1997b) and be equipped with a traceable check source. Fixed instruments should be powered by alternating current (AC) and provided with an emergency power source.

Photon and Neutron Area Monitors. Photon and neutron area monitors measure the intensity of photon and neutron radiation in areas where significant quantities of plutonium are stored and/or handled. ANSI N317 (ANSI, 1980a) states that these monitors shall have a preselectable trip setting with audible annunciators, shall provide electronic signals for remote alarms if they are used as alarming devices, and shall be equipped with a visual meter or digital readout. All neutron and photon area monitors should be AC-powered and all critical monitors should be provided with an emergency power source. Many of the requirements that apply to portable survey instruments, as stated in ANSI N317 may also apply to this type of instrumentation. Calibrations should be performed according to the requirements in ANSI N323 (ANSI, 1997b).

3.5.2.3 Performance Criteria for Emergency Instrumentation

Meeting the criteria for criticality accident alarm systems, fixed nuclear accident dosimeters, and other emergency instrumentation is essential.

Criticality Alarm Systems. ANSI/ANS 8.3 (ANSI, 1997c) discusses the performance and design criteria for criticality accident alarm systems. The criteria include the following:

-- Criticality alarm systems shall be designed to detect immediately the minimum accident of concern; the minimum accident may be assumed to deliver the equivalent of an absorbed dose in free air of

20 rad at a distance of 2 meters from the reacting material within 60 seconds.

-- Systems shall be designed so that instrument response and alarm latching shall occur as a result of radiation transients of 1-millisecond duration. The alarm signal shall be for evacuation purposes only and of sufficient volume and coverage to be heard in all areas that are to be evacuated. Very high audio background noise in some areas may require that the alarm be supplemented with visual signals; however, high background noise is a dangerous situation that should be prevented by design. Instrument response to radiation shall be calibrated periodically to confirm the continuing performance of the instrument. The calibration interval may be determined on the basis of experience but shall be no less frequent than annually. Tests should be performed at least monthly and the results of testing should be documented.

The standard does not quantify criteria for reliability or the rejection of false alarms. Consideration should be given to the avoidance of false alarms as accomplished by providing reliable single detector channels or by requiring concurrent response of two or more detectors to initiate the alarm. (ANSI 1986a).

Fixed Nuclear Accident Dosimeters. All DOE facilities that have sufficient quantities and kinds of fissile material to potentially constitute a critical mass, such that the excessive exposure of personnel to radiation from a nuclear accident is possible, shall provide nuclear accident dosimetry for those personnel (10 CFR 835.1304). Requirements for fixed nuclear accident dosimeters are found in DOE Order 420.1C (DOE, 2012b).

Effluent Monitors. Facilities that deal with unencapsulated plutonium should have continuously operating effluent monitors to determine whether or not plutonium is being released to the environment. Effluent monitor criteria is found in IEC Publications 761-1 and 761-6 (IEC, 1983) and ANSI N42.18 (ANSI, 2004) and should be performed. Similar to airborne contamination monitors, effluent monitors should be tested for air in-leakage at least annually or when seals or “O” rings are replaced.

Other Emergency Instrumentation. Other emergency instrumentation should provide ranges for all radiation dose rates and contamination levels potentially encountered at the time of an accident. Normally, dose rate capabilities from a few millirem per hour to a few hundred rem per hour should be required. Performance specifications for emergency radiological monitoring instrumentation can be found in ANSI N320-1979 (ANSI, 1975) and BNWL-1742 (Andersen et al., 1974).

3.5.3 Instrument Calibrations and Testing

Radiation doses and energies in the work areas should be well characterized. Calibration of instruments should be conducted where possible under conditions and with radiation energies similar to those encountered at the work stations. Knowledge of the work area radiation spectra and instrument energy response should permit the application of correction factors when it is not possible to calibrate with a source that has the same energy spectrum. All calibration sources should be traceable to recognized national standards, such as NIST. Neutron energy spectral information is considered particularly important because neutron instruments and dosimetry are highly energy-dependent.

When the work areas have been well characterized, the calibration facility used by the plutonium plant should be set up to represent as closely as possible the work area's radiation fields. Californium-252 or PuBe calibration sources should be used for work areas that process plutonium metal and plutonium oxide because their neutron energy distribution is similar to those compounds. Facilities that process PuF_4 should use a PuF_4 source. Most work areas at processing plants are high-scatter areas and thus have significant quantities of low-energy neutrons. Because it may not be feasible to have sources and scatter geometries representative of all work locations at the facility, it should be important to determine specific spectra and correction factors for work locations to correct for the calibration. Scatter conditions should be taken into account when setting up a calibration facility. The effect of room scatter in a neutron calibration facility can be significant and may account for as much as 20% of the measured dose rate. Modeling, such as a Monte Carlo N-Particle (MCNP) code, should be used to correct for room scatter.

ANSI N323 (ANSI, 1997b) provides requirements on the calibration of portable instruments and periodic performance testing (e.g. source response checking) of instruments. Section 9.4 of G 441.1-1C, Ch. 1(DOE, 2011a) has additional guidance on this topic.

The reproducibility of the instrument readings should be known prior to making calibration adjustments. This is particularly important if the instrument has failed to pass a periodic performance test (i.e., the instrument response varies by more than ±20% from a set of reference readings using a check source) or if the instrument has been repaired. The effect of energy dependence, temperature, humidity, ambient pressure, and source-to-detector geometry should be known when performing the primary calibration. Primary calibration should be performed at least annually.

Standards referenced in Section 3.5.2 discuss specific performance testing of radiation detection instruments. Testing procedures in these standards should be used for periodic requalification of instruments or detailed testing of instruments.

The calibration of photon monitoring instruments over the energy range from a few keV to 300 keV is best accomplished with an x-ray machine and appropriate filters that provide known x-ray spectra from a few kiloelectron volts to approximately 300 keV. Radionuclide sources should be used for higher energies. Most ion chambers used to measure photon radiations have a relatively flat energy response above 80 to 100 keV; ^{137}Cs or ^{60}Co are typically used to calibrate these instruments. These sources also may be used to calibrate Geiger-Mueller (GM) type detectors used for dose rate measurements. It should be noted that some GM detectors (e.g., those with no energy compensation) can show a large energy dependence, especially below approximately 200 keV. GM detectors should not be used if not energy compensated.

The calibration of alpha-detection instruments normally should be performed with ^{239}Pu, ^{241}Am, or ^{230}Th sources. Several sources of different activities should be used to calibrate different ranges.

Whenever possible, beta detectors should be calibrated to the beta energies of interest in the workplace. A natural or depleted uranium slab source can be used for calibration of beta detectors when beta radiations in the workplace have energies similar to the uranium. The energy dependence of beta detectors can be tested using the calibration sources listed in the International Organization for Standardization (ISO) Publication 1980 (1984); such as ^{90}Sr.

The calibration and testing of crucial monitoring systems are extremely important to the overall radiation protection program but have often been neglected. Effluent monitoring and sampling systems and remote area monitoring systems should be given several tests. The radiological, environmental, and mechanical characteristics of the instrumentation portion of the system should be fully evaluated prior to its first use to ensure its compatibility with performance requirements and facility operating conditions. The effluent sampling losses from the sample probe to the collector/detector should be determined. This test should be repeated at least annually and when a significant change in the sampling equipment is made. The sample probe should be examined at least once a year to verify that its design or performance has not been changed by corrosion. The recorder of the sample flow rate should be calibrated when it is installed and annually thereafter. The operability of the overall system should be completely tested once, with repeat tests only after modification, repair, or maintenance. Operability checks should be scheduled at least monthly and calibration performed at least annually.

The operation of criticality or other radiation alarm signal systems should be checked periodically to ensure that the alarms are audible at all potentially occupied locations. To prevent any desensitizing of staff, the staff should be aware that the tests will be performed, and where possible, tests should be scheduled during off-shift hours. Building systems should be tested semiannually and the area-wide system should be tested at least annually. Any portion of the detector/alarm system that is affected by the test should be reconfirmed for operability after the test is completed (e.g., if a detector is disconnected and a signal is injected at that point, the detector should be tested immediately after it has been reconnected).

3.6 RADIATION SAFETY TRAINING

A thorough radiation protection training program should be established at plutonium facilities. Separate training programs should be established for general employees, radiation workers, and RCTs. The training of all staff members should be carefully documented. The DOE standard, Radiological Control (DOE, 2008a), and DOE standardized training programs (DOE, 2007b and DOE, 2007c) provide guidance on information to be presented during the training programs.

The frequency requirements for Radiation Safety Training are specified in 10 CFR 835.901. Refresher training in the alternate year when retraining is not performed is recommended. Individuals who work with plutonium should have special plutonium facilities training, such as DOE HDBK-1145-2008 Radiological Safety Training for Plutonium Facilities (DOE, 2008c) in addition to Radiological Worker Training, (DOE, 2007c).

Training requirements shall ensure that personnel have the training to work safely in and around radiological areas and to maintain their individual radiation exposure and the radiation exposures of others ALARA.

3.6.1 Radiological Worker Training

Before working in plutonium operations, all radiological workers shall be trained and qualified according to 10 CFR 835.901. A thorough radiation protection training program should be established at plutonium facilities. Before beginning plutonium training, each plutonium worker should receive Radiological Worker Training and other radiation safety topics as required by 10 CFR 835.901(a).

The level of radiation worker training should be determined in accordance with the standard, Radiological Control, Table 6.1 (DOE, 2008a). All training should be in accordance with Radiological Worker Training, (DOE, 2007c) and implemented by the guidance of Chapter 14 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). All training dispositions and records shall be documented in accordance with 10 CFR 835.704 (DOE, 2007a).

3.6.2 Radiological Control Technician Training

A thorough RCT training program should be established at plutonium facilities. Before plutonium operations begin, a trained and qualified staff of RCTs should be present. All RCT training should be accomplished in accordance with DOE HDBK-1122-99 (DOE, 2009a)

3.6.3 Training for Other Facility Personnel

Nonradiological workers in a plutonium facility should be given a general orientation on the radiation safety concerns for working with plutonium, the general protective measures used for work with plutonium, and the engineered safety features of the facility.

3.6.4 General Public Education

If there are members of the public who live or work near a plutonium facility, a plan for orientation of members of the public should be developed to inform them of facility activities. Such a plan should include information on the concerns that require protection of people from potential injuries by plutonium, the general protective measures used at the facility to confine it and keep it out of the public domain, and solicitation of information on the concerns of members of the local public about plutonium. To the extent possible, efforts should be made to allay those concerns. The information in the public education plan should also be provided to local news media.

3.6.5 Training Qualifications

All training instructors and materials should meet the requirements in DOE Order 426.2, (DOE, 2010a) and should meet the guidance in the standard, Radiological Control (DOE, 2008a).

Each plutonium facility should develop performance-based training that reflects radiological conditions present at the facility. This training should be monitored to ensure that site specific, worker-performance-based measures, and practical factors are included in the plutonium training.

3.6.6 Health Physicist Training Involvement

Facility health physicists should have comprehensive knowledge of all of the material on plutonium radiation safety that is included in the training programs for radiation workers and RCTs.

3.7 RADIOLOGICAL RECORDS

The systematic generation and retention of records relating to the occupational radiation protection program are essential to describe the occupational radiation exposure received by workers and the conditions under which the exposures occurred. Such records have potential value for medical, epidemiological, and legal purposes.

Regulation 10 C FR 835 (DOE, 2007a) establishes radiation protection program records requirements. The standard, Radiological Control (DOE, 2008a), provides guidance for radiation protection program records.

10 CFR 835 Subpart H requires that records be maintained that document compliance with 10 CFR 835. Subpart H requires specific information on the following types of records:

- Individual monitoring records
- Monitoring and workplace records
- Administrative Records

Most of the required radiological records have established retention periods. The retention periods are discussed in DOE Order 200.1A (DOE, 2008b). Individual records may be covered by the Privacy Act; the DOE has codified the Privacy Act in 10 CFR 1008, Records Maintained on Individuals (Privacy Act) (DOE, 1994b).

Detailed guidance on development and maintenance of a radiological exposure recordkeeping and reporting system can be found in Chapter 13 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a).

3.8 ALARA AND OPTIMIZATION

The policy of maintaining radiation exposures ALARA has existed in principle since the early 1940s. The evolution of ALARA into a formal program began in the early 1960s. It is well to remember that the ALARA approach was applied to radiation protection far earlier and is much more institutionalized than any comparable approach to other hazards.

Although there is, and has been since the 1940s, a series of official established dose limits, they do not represent ALARA. ALARA is a continuous process of controlling and managing radiation exposure to workers, the general public, and the environment. Although ALARA is based upon protection of people and the environment, the philosophy is also grounded on sound economic and operating principles. The responsibility for maintaining radiation exposures ALARA is not a unique responsibility of management or health physics personnel. It is a responsibility of everyone involved in managing, supervising, or performing radiation work. It is imperative to teach administrative personnel to support the principles and practice of ALARA, and to train all radiation workers to consider ALARA as they prepare for and perform their work.

10 CFR 835 Subpart K “Design and Control” contains specific requirements relating to ALARA considerations for facility design and modification. Also, DOE Order 458.1, Ch 2 “Radiation Protection of the Public and Environment” (DOE, 2011c) contains environmental ALARA requirements.

3.8.1 Current Status of ALARA Programs

Currently, it is common practice in a DOE facility to have a well-structured ALARA plan for the entire facility, with more detailed plans in the various buildings or functional subunits of the facility. There is ordinarily a facility coordinator who administers the overall ALARA plan and reports to top-level management of the facility. Coordinators for the various buildings or subunits of the facility receive guidance from the overall facility coordinator and report the results of their ALARA programs to that individual.

3.8.2 Achievement of Goals

The standard, Radiological Control (DOE, 2008a), provides guidance to contractors (facility) to provide documentation of the ALARA process. To ensure improving radiological performance, at the beginning of each fiscal year, each facility prepares and submits Radiological Performance Goals. At least quarterly, the contractor (facility) provides the contractor senior site executive with an interim status report of the goals. At the end of the calendar year, an Annual Goal Status Report is issued.

Identifying specific ALARA goals in plutonium facilities requires close coordination between the facility ALARA team members (operations, maintenance, and health physics personnel) made up from a cross-section of personnel representing the various work elements of the facility. ALARA goals may be formulated as qualitative or quantitative types of goals, but shall be measurable and achievable, with clearly defined endpoints.

3.8.3 Quality Assurance

Important aspects of any ALARA program are the measurement of beneficial effects and the determination that important factors, such as economic impacts, the time involved in accomplishing tasks, and the utilization of personnel, are being optimized. To accomplish these objectives, it is necessary to have a written plan for the ALARA program and high quality records of activities involving exposures to workers, the public, and the environment. These permit comparisons with past experiences and analysis of the recorded activities. In many cases, such studies of the recorded activities not only confirm satisfactory execution of the work, but reveal opportunities for future improvements.

One approach which works very well is the inclusion of an ALARA worksheet along with the RWP. Such a worksheet should be prepared by an individual with responsibilities for the work to be performed, a relatively detailed knowledge of the radiological conditions, and knowledge of what is required to accomplish the task. The worksheet should contain estimates of the time to complete the task and the expected radiation doses that will be received. If any special engineered devices are used to control or reduce personnel exposure, they should be noted on the ALARA worksheet, along with any special instructions that they require. These worksheets provide valuable information for analysis of the effectiveness of the ALARA program for each job.

3.8.4 Technical Aspects

The technical aspects of ALARA programs include not only the standard equipment regularly used in controlling dose to workers, the public, and the environment, such as facility shielding, ventilation filters, installed and portable radiation measuring instruments, but also many special devices that may be used temporarily. Special devices can be used to provide exposure control and/or containment when it may not be practical without them. These include temporary shields, tents or greenhouses, portable fans, ductwork and filters, and special fixtures to hold highly radioactive materials requiring detailed inspections, repairs, modification, or fabrication. Such devices can permit doing difficult work at low radiation doses, which might not be possible otherwise.

Some of these special devices may have general application and can be kept on hand for use as needed. In some cases, devices would have to be especially fabricated for a specific task. Since this would ordinarily have a significant effect on the cost of doing that job, the economic aspects of doing or not doing the job would have to be carefully evaluated.

3.8.5 Attributes of Effective Review and Audit

Evaluation of the effectiveness of an ALARA program requires both reviews and auditing. The reviews will include detailed examination of the written ALARA program plan and the records of ALARA activities. The objectives in such reviews are to find if the written plan is being followed, and what is working or not working well. Such reviews can be performed adequately by either a knowledgeable member of the facility staff or an equally knowledgeable outsider. The written report of a review should be directed to a member of management who is responsible for implementation of the ALARA program.

Audits are best performed by an outside health physicist who is sufficiently knowledgeable about work with plutonium and its radiological characteristics that he/she knows where to look for problems and can make appropriate evaluations and recommendations. He should not only examine the ALARA program plan and records, but should also visit the working areas and laboratories in the facility, with a knowledgeable escort who can answer questions about activities and conditions in the facility.

There is nothing really unique in ALARA programs at plutonium facilities, compared with facilities handling other kinds of radioactive materials. However, the radioactivity of plutonium, its potential for criticality, and its relatively high radiotoxicity require somewhat more meticulous surveillance and control than many other radionuclides. Therefore, the detail in ALARA programs for plutonium facilities is likely to be somewhat greater than would be found in ALARA programs for many other facilities.

In any plutonium facility, it is highly desirable to have well-structured ALARA teams in each building or subunit of the facility. Facility goals should be developed by the facility ALARA teams. All facility-specific goals should be categorized using the facility-specified format and should include the following:

-- **Exposure Reduction**. Goals listed under exposure reduction may reflect occupational or nonoccupational exposure reduction. Exposure to radiological hazards or nonradiological hazards are relevant. Specific jobs for which exposure reduction plans have been developed should be covered in this section. Exposure may be reduced by reducing other hazards that contribute to the difficulty of performing work in radiological areas. For example, reducing noise, reducing heat stress conditions, or improving lighting may facilitate the completion and accuracy of work performed in radiological areas and, thus, reduce exposure. Such opportunities for exposure reduction should be carefully evaluated and appropriate ALARA goals established to make the most of these opportunities.

-- **Source Reduction**. Source reduction should concentrate on minimizing or eliminating the sources of radiation exposure. Reducing the number of areas with radiological contamination and reducing dose rate are examples of source-reduction goals. Where the presence of nonradiological hazardous materials results in mixed waste, the removal of the hazardous material may have ALARA benefits by reducing the waste classification. Such changes may also reduce exposure at a later time by eliminating the need to store or further treat the waste. In these cases, eliminating the hazardous material may be an appropriate source-reduction ALARA goal.

-- **Administrative**. Administrative goals typically encompass training, program improvements, procedure revision, or other administrative-type activities. Administrative goals are generally qualitative, so it is difficult to develop endpoints for them. Specific efforts shall be made to ensure that adequate closure mechanisms exist for administrative goals.

During all phases of ALARA goal-setting, the facility health physics personnel should be intimately involved in providing advice and expertise on ALARA actions.

When addressing exposure reduction, a cost/benefit analysis should be made to determine the real cost of implementing a dose reduction plan. The Health Physics Manual of Good Practices for Reducing Exposures to Levels that are as Low as Reasonably Achievable (Munson et al., 1988), provides an excellent methodology for conducting a cost/benefit analysis by health physics personnel.

The application of ALARA principles to the performance of work in the field is the main objective of any ALARA program. ALARA design, engineering, planning, and administration come to fruition in maintaining exposures ALARA to workers and the public. The operational application of ALARA requires cooperation and coordination of many functional groups, including radiation protection, operations, maintenance, planning and scheduling, training, engineering, and administration.

The primary responsibility for controlling radiation exposure during operations rests with the individual and his/her immediate supervisor. The support functions provide the training, resources, guidance, and measurements, but it is in the application that the effectiveness of an ALARA program is realized. Operational measures for controlling exposure shall be applied to assure that any work with radioactive materials is carried out in the safest manner reasonable. Both engineered and administrative control measures should be used for limiting exposure.

Engineered controls should be utilized whenever possible. In addition, periodic verification of the continued effectiveness of these controls should be performed by facility health physics personnel. Ventilation and filtration systems should be routinely checked and inspected to assure that operation is maintained within the design criteria. The integrity of shielding, the reliability of equipment, and the calibration of instruments should likewise be routinely verified.

Although administrative controls are not an adequate substitute for engineered features, they are necessary. They are a part of the management systems developed and implemented to provide guidance, direction control, and limitations for activities. Administrative controls include the documents that describe organizational interfaces and prescribe controls for radiation protection. Administrative controls, especially procedures, should be reviewed by those responsible for ALARA to ensure that radiation exposure activities include dose limitation considerations.

Factors that shall always be considered in an ALARA program are the costs and benefits. This is especially important when the identified benefit represents a very small increment of radiation dose reduction. Funds for dose reduction should always be applied to actions which will achieve the greatest dose reduction for the cost.

The final decontamination and decommissioning (D&D) of a plutonium facility should be given consideration in both the original design of the facility and any modifications done to the facility during its operating lifetime. Likewise, D&D should be given consideration in choosing operating processes and practices for the facility, including any changes in processes and practices during its operating lifetime. Both design and operating activities can affect the radiation levels and personnel doses encountered by workers who perform the D&D activities. To the extent practicable, design and operations should provide for radiation levels that are ALARA during D&D activities.

The successful implementation of an ALARA program requires the commitment, support, attention, and efforts of all members of an organization. In facilities in which the radiation exposures are already relatively low, implementation of the ALARA concept is particularly challenging. The reduction of radiation doses to ALARA levels demonstrates to workers and the public a continued emphasis, commitment, and concern for health and safety.

3.9 CONDUCT OF OPERATIONS

The organization and administration of operations should ensure that a high level of performance in DOE facility operations is achieved through effective implementation and control activities. Administration of operations activities should recognize that protection of the environment, maintaining high-quality safety, and productivity are compatible goals. The DOE policies should describe the standards of excellence under which the facility is expected to operate. Clear lines of responsibility for normal and emergency conditions shall be established. Effective implementation and control of operating activities are achieved primarily by having readily accessible written standards for operations, periodical monitoring and assessment of performance, and personnel accountability for performance. For a more detailed discussion, see DOE Order 422.1, Conduct of Operations (DOE, 2010b).

A high level of performance in DOE operations is accomplished by management establishing high operating standards and then by communicating the operating standards to workers:

-- By providing sufficient resources to the operations department

-- by ensuring that personnel are well trained by closely monitoring performance in operations

-- by holding workers and their supervisors accountable for their performance in conducting activities.

Senior management establishes operating standards, considering input from workers when appropriate. Working-level personnel will more strongly support the standards when they have had appropriate input into their development. The standards should define operating objectives, establish expected performance levels, and clearly define responsibilities in plant operations. Standards for operating activities should be integrated into operations department procedures and programs. Operating standards should also be communicated to workers by training them in operating practices and by having supervisors monitor and guide work involving facility operations. Sufficient staff, facilities, equipment, and funding should be allocated to permit the operations department to effectively perform its functions. Performance in operations should be closely monitored by facility management, preferably using operating reports and goals, so that the performance of the operations department can be effectively measured. Operations personnel should be held accountable for their performance through supervisor counseling, performance appraisals, and, when necessary, disciplinary measures. Remedial training should be provided when appropriate.

The health physics organization, as a support element, shall ensure that all aspects of radiation safety are considered in the establishment of operations standards and policy. A well instituted cooperative relationship between operations and health physics is paramount to the health and safety of workers and the public and to protection of the environment.

A plutonium facility should have a written policy on radiation protection, including an ALARA policy. All radiation protection procedures and controls should have recognizable or formal technical bases for limits, methods, and personnel protection standards. Procedures should be adequately documented, updated periodically, and maintained in a centralized historical file. A control system should be established to assure that all copies are accounted for and that all new procedures are included in the historical files. A designated period of time for holding the historical files should be established. DOE Order 200.1A (DOE, 2008b) and ANSI/HPS N13.6 (ANSI, 2010) provide guidance on how long to keep historical files. In addition, radiation protection procedures should have a documented approval system and established intervals for review and/or revision. A tracking system should be developed to ensure that the required reviews and revisions occur.

The radiation protection procedure system should provide for but not be limited to, the following topics: radiation work procedures, posting and labeling, instrument calibration, and provision for audits.

3.9.1 Radiation Work Procedures

Radiation work procedures, including RWPs, survey procedures, ALARA reviews, sample counting, and other task procedures, fall within the requirements for conduct of operations. All sections of DOE Order 422.1 (DOE, 2010b) apply. The guidance and requirements of Section XVI, “Operations Procedures,” is especially pertinent to radiation work procedures. Procedures are a key factor affecting radiation protection performance. Appropriate attention should be given to writing, reviewing, approving, and monitoring implementation of radiation protection procedures. There should be documented qualification and training requirements for those who prepare and approve procedures. A formal approval process should be established. Procedure changes and revisions should be subject to the same review and approval process as the initial procedure.

Personnel should be trained in the use of the procedures they will be expected to perform. For RWPs, workers should read the RWP and verify by signature that they have read it, understand its contents, and will comply with its requirements in the conduct of the work. Procedures should be available for personnel use. The

RWPs should be posted at the entrance to the work location. There should be a system in place to assure that posted copies of all work procedures, including RWPs, are current.

3.9.2 Posting and Labeling

The requirements for posting and labeling of working areas because of the presence, or potential presence, of radiation and/or radioactive material are specified in 10 CFR 835, Subpart G (DOE, 2007a). Guidance in implementing the regulatory requirements can be found in Chapter 12 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a), and the standard, Radiological Control (DOE, 2008a). Conformance of conduct of operations requirements should assure a reasonable degree of uniformity in the posting and the signs used, as well as verifying that operator aids and other posted information do not interfere with necessary radiological posting. It is necessary to formally review posting of radiological areas in the same manner that the posting of operating aids is reviewed, in conformance with DOE Order 422.1 (DOE, 2010b).

3.9.3 Calibration of Instruments

The status of installed and portable radiological instruments should be well known and appropriate to the use. (Calibration of radiological instruments is discussed in Section 3.5.2.)

“Ownership” of installed radiological dose rate and airborne contamination monitoring instrumentation should be well known and the responsibility and authority for calibration, repair, and notification clearly established. Because such information is often used by more than one group, formal notification procedures should be established to cover those times when the instrument is out of service or beyond the required calibration schedule. Configuration control and quality assurance requirements for installed systems should be established commensurate with their safety significance.

For portable instrumentation, conduct of operations requirements are normally built into the routine calibration and survey program. Functional checks are routinely made to verify calibration, instruments are checked to assure that they are within the calibration period, and survey procedures require identification of the instruments used so that if a problem is later found, measurements can be repeated.

3.9.4 Audits

Conduct of operations does not, in itself, contain requirements on auditing. Inspections, audits, reviews, investigations, and self-assessments are part of the checks and balances needed in an operating program. Auditing is one of the many tools that line management has at its disposal to identify problems. Regulation 10 CFR 835.102 requires internal audits of all functional elements of the radiation protection program no less frequently than every 3 years. These audits are to include program content and implementation. Each one of the 18 topics addressed in DOE Order 422.1 (DOE, 2010b) should be subject to both internal self-assessment and external auditing to assure effective implementation of their requirements. Any deficiencies identified should be documented and corrective actions aggressively pursued and tracked to completion. The self-assessment and audit process should include conducting trend analyses and root cause evaluations of deficiencies and communication of results throughout the organization.

3.9.5 Decommissioning of Weapons and Weapon Facilities

Decommissioning of nuclear weapons and nuclear facilities is subject to the same conduct of operations requirements as operating facilities. In general, some components, once they are separated, can be downgraded in safety significance. Also, facilities undergoing decommissioning will have fewer safety systems. During decommissioning, status control and shift turnover are extremely important considerations and shall be done in accordance with DOE Order 422.1 (DOE, 2010b). Posting and labeling of radiological areas are also an increasing challenge because of the rapidly changing radiological status. In extreme cases, it may be desirable to have workers review or sign the RWP each day to ensure they are aware of the status.

4.0 CONTAMINATION CONTROL

The primary control for contamination in a plutonium plant is the facility design. Contamination is confined primarily by enclosing the process areas and using controlled ventilation systems. The design objective for the confinement system is to essentially prevent or minimize exposure of plant personnel and the public to airborne contamination. To ensure that this objective is met, additional attention should be given to airborne contamination control, surface contamination control, and personnel contamination control. Radiological controls for the workplace should ensure that radionuclides are contained and handled properly and that intakes, if they occur at all, are negligible to the extent achievable with state-of-the-art technology. However, much of the current effort involves decommissioning of no-longer-needed production facilities. The lack of engineered controls or the systematic removal of existing controls during the decommissioning process introduces a completely different set of circumstances that requires special attention for adequate contamination control and worker and public protection.

4.1 AIRBORNE CONTAMINATION CONTROL

To achieve the design objective of preventing (or at least minimizing) internal exposure of plant personnel, airborne contamination shall be confined to process enclosures which have adequate air cleaning systems. Because both equipment and personnel errors can compromise designed protection and because older facilities may already have unconfined plutonium, air monitoring and other contamination control measures are needed. Experience has shown that the most common route for inadvertent plutonium deposition in man is by inhalation even though intakes may also occur by accidental ingestion or by wound contamination. In facilities being decommissioned, the use of temporary containment structures, interim ventilation systems, and administrative controls such as protective clothing and respirators may be required to replace engineered systems.

10 CFR 835.1002 requires that for the control of airborne radioactive material, the design objective shall be, under normal conditions, to avoid releases to the workplace atmosphere and in any situation, to control the inhalation of such material by workers to levels that are ALARA; confinement and ventilation shall normally be used.

Note: The use of ventilation systems may require the approval of Facility Criticality Safety personnel because these systems may concentrate fissionable material.

4.1.1 Internal Versus External Dose Philosophy

The overall goal of radiological protection is to minimize the total dose to the individual. However, because of the difficulties and cost of evaluating internal exposures to plutonium, it is best to avoid all internal exposures during routine operations and anticipated abnormal events by engineered controls and personnel protective equipment. As stated above, this is an extremely challenging goal for those facilities undergoing decontamination/decommissioning activities or facilities/sites in environmental remediation. The conditions encountered in decommissioning and environmental restoration will typically place a heavy reliance on administrative controls.

4.1.2 Purpose of Air Monitoring

Airborne contamination surveys are performed for the following reasons:

- Prompt detection of airborne contaminants for worker protection.
- Personnel exposure assessment.
- Monitoring of trends within the workplace.
- Special studies.

Of primary importance is the prompt detection of airborne contaminants. The rapid, early detection of airborne releases requires knowledge of the potential sources and characteristics of the airborne material, the locations of the personnel who are at risk, and the capabilities of the detection devices. Optimally, the samples should be taken between the source and the person to measure the potential airborne radioactivity exposure to the individual. With the numerous sources and mobility of the workers, accurate measurement of potential exposure to the individual using area air monitoring devices under all conditions is difficult, if not impossible, to achieve. To aid in early detection of unanticipated airborne radioactivity (e.g., as a result of an undetected pinhole leak in a containment glove), samples of airborne materials should be taken as close to their points of potential origin as practicable to maximize the probability of detection (airborne concentrations are at a maximum at their points of origin). To aid in monitoring of an individual's exposure to airborne radioactivity where area sampling is not representative of the individual's exposure, the use of lapel air samplers on the individual is recommended. Detailed guidance for the placement of air samplers and monitors, selection of system characteristics and requirements, and maintenance and calibration of the equipment is available in the Health Physics Manual of Good Practices for the Prompt Detection of Airborne Plutonium in the Workplace (Mishima et al., 1988) and Air Sampling in the Workplace (NRC, 1993).

4.1.3 Regulations and Limits

The regulations for control of radiation work are covered in 10 CFR 835 (DOE, 2007a). Additional requirements and guidance for implementation is provided in the DOE standard, Radiological Control (DOE, 2008a), and the Implementation Guide. While many of the topics included in the Implementation Guide relate to plutonium contamination control, specific guidance on contamination control has not been provided. The limits established for plutonium and other transuranic elements for contamination areas, high contamination areas, and airborne radioactivity areas are given in 10 CFR 835.603 and Appendix D of 10 CFR 835. The Appendix D values are summarized in Table 4.1.

Table 4.1 Surface Contamination Values

Radionuclide	Removable [2,4]	Total (Fixed + Removable) [2,3]
U-nat, U-235, U-238, and associated decay products	[7] 1,000	[7] 5,000
Transuranics, Ra-226, Ra-228, Th-230, Th-228, Pa-231, Ac-227, I-125, I-129	20	500
Th-nat, Th-232, Sr-90, Ra-223, Ra-224, U-232, I-126, I-131, I-133	200	1,000
Beta-gamma emitters (nuclides with decay modes other than alpha emission or spontaneous fission) except Sr-90 and others noted above[5]	1,000	5,000
Tritium and special tritium compounds[6]	10,000	See Footnote 6

[1] The values in this appendix, with the exception noted in footnote 5 below, apply to radioactive contamination deposited on, but not incorporated into the interior or matrix of, the contaminated item. Where surface contamination by both alpha- and beta gamma-emitting nuclides exist, the limits established for alpha- and beta-gamma-emitting nuclides apply independently.

[2] As used in this table, dpm (disintegrations per minute) means the rate of emission by radioactive material as determined by correcting the counts per minute observed by an appropriate detector for background, efficiency, and geometric factors associated with the instrumentation.

[3] The levels may be averaged over one square meter provided the maximum surface activity in any area of 100 cm^2 is less than three times the value specified. For purposes of averaging, any square meter of surface shall be considered to be above the surface contamination value if: (1) from measurements of a representative number of sections it is determined that the average contamination level exceeds the applicable value; or (2) it is determined that the sum of the activity of all isolated spots or particles in any 100 cm^2 area exceeds three times the applicable value.

[4] The amount of removable radioactive material per 100 cm^2 of surface area should be determined by swiping the area with dry filter or soft absorbent paper, applying moderate pressure, and then assessing the amount of radioactive material on the swipe with an appropriate instrument of known efficiency. (Note - The use of dry material may not be appropriate for tritium.) When removable contamination on objects of surface area less than 100 cm^2 is determined, the activity per unit area shall be based on the actual area and the entire surface shall be wiped. It is not necessary to use swiping techniques to measure removable contamination levels if direct scan surveys indicate that the total residual surface contamination levels are within the limits for removable contamination.

[5] This category of radionuclides includes mixed fission products, including the Sr-90 which is present in them. It does not apply to Sr-90 which has been separated from the other fission products or mixtures where the Sr-90 has been enriched.

[6] Tritium contamination may diffuse into the volume or matrix of materials. Evaluation of surface contamination shall consider the extent to which such contamination may migrate to the surface in order to ensure the surface contamination value provided in this appendix is not exceeded. Once this contamination migrates to the surface, it may be removable, not fixed; therefore, a "Total" value does not apply. In certain cases, a “Total” value of 10,000 dpm/100 cm^2 may be applicable either to metals of the types from which insoluble special tritium compounds are formed, that have been exposed to tritium, or to bulk materials to which insoluble special tritium compound particles are fixed to a surface.

7 These limits apply only to the alpha emitters within the respective decay series.

Note: This document concerns release to controlled areas only. Requirements for unrestricted release of materials and equipment are found in DOE Order 458.1, Ch 2 (DOE, 2011c). Refer to that document for guidance regarding unrestricted releases.

4.1.4 Uncertainties and Limitations

Because of the large exposure per unit intake associated with plutonium and the difficulties in evaluating certain exposures, it is important to consider the uncertainty in the measurements when designing a plutonium monitoring program. Although the design objective of the facility will likely be no airborne plutonium contamination, the reality will be a measurement that ensures airborne plutonium is below an acceptable lower limit of detection. The sampling and monitoring program will need to be designed not only for prompt detection of airborne contamination, but to assure that samples are representative of the air that the workers are breathing and have a low enough limit of detection that only negligible doses could go undetected. This is especially important because of the technology shortfall for routine bioassay and in vivo analysis in detecting small intakes of plutonium. Air samples are typically considered representative if they are taken using a personally worn lapel air sampler or if the sample head is within 1 foot of the workers head. The need for an effective sampling and monitoring program is even more critical in the rapidly changing environment of decommissioning activities.

Numerous factors enter into any determination of plutonium contamination levels and the risk to workers. Some of these factors are detection efficiency of the measuring instrument, collection efficiency of the smear media or air sample filter, the location of the smear or air sample in relation to the source of contamination, the physical and chemical properties of the contamination, the representativeness of the air sample to the air being breathed by the worker, the engineered controls available, and the protective equipment used. All of these factors shall be considered in the development of a plutonium contamination control program and in evaluating the actions required for personnel protection.

4.1.5 Samples and Instrumentation

For plutonium facilities, both air sampling and air monitoring are essential elements of the radiological control program. Real-time air monitoring using alpha-sensitive CAMs should be used to alert workers to rapid degradation of radiological conditions. The air sampling system with a lower limit of detection shall be adequate to provide continuing assurance that personnel exposures are within limits and ALARA.

The characteristics of a good plutonium CAM include:

-- A lower limit of detection equal to or better than 8-DAC-h

-- high reliability with a minimum of spurious alarms

-- a stable and constant flow air mover

-- stable and documented detector efficiency with geometry, filter collection efficiency, self-attenuation, etc., considered

-- methodology for radiation discrimination and natural radioactivity discrimination

-- system for activating an alarm

- -- shielding for extraneous sources of interference such as radiation, radiofrequency, temperature, and vibration
- -- mechanical and electrical ruggedness
- -- ease of maintenance and calibration.

A plutonium air sampling program typically includes a system of fixed head air samplers to quantify air concentrations in the workplace. The basic characteristics of the sampling equipment remain the same except that there is normally less flexibility in locating the sampling heads but more flexibility in selecting and operating the counting instrumentation. In many instances, installed sampling systems may no longer be operational or may be in the wrong locations. In those instances, portable air sampling systems, either impactor-head type or filter type may be used to provide required worker protection.

4.1.6 Sample Analysis

Plutonium air samples are typically analyzed by alpha counting, alpha spectral analysis, or chemical analysis. The technique used will depend upon the filter media used, the physical and chemical state of the contaminate, the urgency for the data, interfering radionuclides, and other factors. Authoritative guidance in establishing plutonium air sampling counting and analysis methods can be found in NCRP Report No. 58, A Handbook of Radioactivity Measurements Procedures (NCRP, 1985) and in Air Sampling in the Workplace (NRC, 1993).

4.1.7 Monitoring Strategies and Protocols

The rapid, early detection of airborne releases requires knowledge of the potential sources and characteristics of the airborne material, the locations of the personnel who are at risk, and the capabilities of the detection devices. Optimally, the samples should be taken between the source and the potentially exposed worker (or member of the public) to intercept the airborne materials before they reach the individual. With the numerous sources and mobility of the workers, interception under all conditions is difficult, if not impossible to achieve. Samples of airborne materials should be taken as close to their points of origin as practicable to maximize the probability of their detection (airborne concentrations are at a maximum at their points of origin).

Fixed probes that are positioned to intercept releases from recognized major potential sources should be used along with portable air samplers for planned activities with known potentials for airborne release of contaminants and for temporary storage of contaminated materials in areas of low air flow. If the workplace exhaust system can be shown to provide rapid, essentially quantitative clearance of airborne contamination, fixed probes that sample the exhaust system may be adequate for routine coverage of unplanned activities. If justified by documented studies, other sampling arrangements may be used that provide improved “total” coverage of the workplace environment for the early detection of airborne contamination.

Those responsible for the rapid and reliable detection of airborne plutonium should consider the following workplace characteristics in evaluating monitoring systems and working environments (Mishima et al., 1988):

-- The airflow patterns and airborne transport of plutonium in the workplace

-- The location of personnel within the workplace during various processing procedures

-- The location at which the airborne plutonium sample should be taken to accurately monitor the activity inhaled by workers

-- The ability of the system to transport an undistorted sample to the collection media or measurement device

-- The collection and retention efficiency of the collection medium

-- The efficiency of the measurement device in measuring the plutonium collected and differentiating the plutonium from other materials present

-- The accuracy and reliability of the system.

Guidance for each area listed above is provided in Mishima et al. (1988).

4.2 SURFACE CONTAMINATION CONTROL

Controlling plutonium surface contamination is essential because it may easily be resuspended in air and/or transferred to other surfaces. The following elements are important for controlling surface contamination: keeping plant surfaces clean; monitoring, reporting, and tracking contamination levels; and establishing appropriate control zones with limits and action levels for those zones.

4.2.1 Plant Surfaces

Good housekeeping practices are essential in keeping plant surfaces clean. Periodic housekeeping should be performed within contaminated areas to minimize the buildup of contamination and contaminated waste. Periodic decontamination both within contaminated glove boxes and in the general work area should be conducted to minimize removable contamination.

In some instances, it may be appropriate to apply fixatives to minimize the movement of plutonium contamination. However, it is generally desirable to attempt decontamination first. If decontaminating is not successful or perhaps, not appropriate for the job scope, a fixative may be appropriate. If a fixative is used, typically a paint, two layers of fixative should be used, with the bottom coat yellow and the top coat a different color. When the yellow begins to show through the top coat, additional fixative should be applied. Also, for areas which have had a fixative applied over plutonium contamination, a routine contamination survey should be conducted to assure that no contamination has become movable over time.

In some cases a strippable coating may be used to allow easy decontamination at the completion of a job. These strippable coatings are sometimes used to decontaminate areas. An aerosol fixative is also available that can be pumped into a room, glovebox, or other work space, that coats all exposed surfaces, including the underside of components. This allows work to proceed without disturbing contamination.

Note: The use of fixatives may require the approval of Facility Criticality Safety personnel because fixatives may concentrate or moderate fissionable material.

Outside areas may also require a fixative to minimize the spread of contamination. Historically, some outside contaminated areas have been covered with asphalt to fix contamination. This is not a desirable material to use because it creates a mixed hazardous waste as well as significantly increasing the volume of contaminated material for disposal. Two substances that currently are used as an interim fixative for outside soil/surface contamination areas are (1) a derivative of pine tar (toll oil), which forms a non-toxic surface fixative that is hard and appears to have a relatively durable surface and (2) a mixture of white glue and water (enduro seal), which is easily sprayed on and sets rapidly to a firm surface. A water to glue ratio of about 25 to 1 appears to perform well in preliminary tests. Both of these fixatives are only interim measures because of eventual degradation from the elements. For more localized areas where a permanent fixative/cover is needed, a sprayable concrete (Shotcrete) is available. A disadvantage of this material is cracking, which defeats the sealing surface. Another material that can be used as a carpeting for outside contamination is a spray-on two-part polymer that provides a flexible, semidurable cover.

The characteristics of the cover can be adjusted to vary water transmission and the color can be changed to inhibit growth under the covering. The major problem for outside use of all of these fixatives is the invasion and actions of biota. Mice, rabbits, other wildlife as well as plant growth tend to burrow under any covering and spread the contamination. While these measures do not permanently solve the problem, they may provide a method of preventing the spread of contamination until a permanent, acceptable solution is determined.

4.2.1.1 Housekeeping

The three housekeeping practices listed below should be followed in a plutonium facility as part of the Conduct of Operations [see DOE Order 422.1 (DOE, 2010b)]:

-- The inventory of contaminated and potentially contaminated scrap and equipment should be kept to a minimum because all such materials are subject to special monitoring and accountability.

-- Radioactive contamination should be controlled and the spread of contaminants and the potential for accidents involving contaminants shall be minimized. (In at least one instance, poor housekeeping contributed to a serious criticality accident.) Management at all levels should continuously emphasize the importance of good housekeeping, and operating procedures should be written to ensure good housekeeping practices.

-- Measures shall be taken to maintain radiation exposure in controlled areas as low as is reasonably achievable through engineered and administrative controls (10 CFR 835.1001).

Where possible, materials that are not absolutely necessary to an operation should be kept out of the contaminated or potentially contaminated area. It is very important to minimize the creation of TRU waste. All packaging and unnecessary protective coverings should be removed before materials are introduced into the process area. Likewise, items that are not necessary to the process should be promptly removed, particularly from glove boxes, and not left to accumulate and become safety hazards, potential fire hazards, sources of radioactive (dust) accumulation, or sources of exposure.

Good housekeeping practices inside glove boxes should emphasize fire and explosion control. Only metal or nonflammable plastic containers should be used for the accumulation of scrap and wastes of any kind in the glove boxes and throughout plutonium facilities. Accumulation of combustible materials in glove boxes should be minimized. When explosive, flammable, or volatile liquids are allowed, they should be rigidly controlled and used only in inert gas atmospheres unless a safety analysis review shows it is safe to do otherwise. All residues should be removed immediately at the conclusion of each job or cleaning operation.

Considerable effort has been expended on the development of coated and corrosion-resistant tools. Some efforts have been marginally successful, but in most cases throw-away tools are favored. Electropolishing of contaminated metal tools and equipment has been shown to be a good method of decontamination and allows for their reuse in some cases or disposal as non-contaminated waste. Where possible, all tools with sharp edges or points (e.g., screwdrivers, ice picks, scissors) should be kept out of glove boxes.

Management should constantly demand good housekeeping. Mandatory, routine clean-up periods are becoming more common due to the increasing cost of storing and disposing of contaminated materials. Better housekeeping is required due to real-time, computerized accountability for nuclear materials. It has been demonstrated that kilogram quantities of plutonium oxide dust can accumulate in glove boxes unless they are routinely cleaned. Much of the exposure to workers originates from layers of plutonium oxide dust on the surface of gloves and the internal surfaces of glove boxes. In processes where plutonium oxide powder is handled, the glove boxes should be cleaned weekly to reduce the accumulation of dust layers and to reduce worker exposure. Although difficult to achieve and maintain, good housekeeping is equally essential during decommissioning of plutonium facilities.

4.2.1.2 Vacuuming

The subject of vacuuming within a glove box is somewhat complex. Experience has shown vacuuming to be the most effective and quickest way to clean a controlled-atmosphere (dry) glove box. It is not particularly effective for high-humidity or wet-process glove boxes, particularly those that involve acids. After acids have been used in a glove box, washing and wiping is the preferred method of cleaning the etched surfaces.

Vacuuming is particularly effective in dry-atmosphere and inerted enclosures where the levels of radioactive dust can quickly increase personnel exposure. In many cases, vacuuming reduces the exposure level more than a wipe down with a damp cloth, and it can be done more quickly and with less waste material generated. Two factors weigh against vacuuming: possible safety hazards from electrical sparks, and the occasional difficulty of operating in inert atmospheres (although the last item need not be of importance). However, in dry glove boxes with dusty operations using high-exposure plutonium, personnel exposure control is a problem and vacuuming is a quick and effective method of keeping the dust and exposure rates under control and should be considered.

Note: The use of vacuum cleaners may require review by Facility Criticality Safety personnel because vacuum cleaners are likely to concentrate fissionable material.

The use of vacuum cleaners for contamination control requires careful consideration and strict controls to assure that the process does not spread contamination. As a minimum, all vacuums used for radioactive material should have high efficiency particulate air (HEPA) filtration on the exhaust. In some instances, the additional precaution of having the exhaust vented into a process ventilation system should be considered.

4.2.2 Reporting and Documenting Contamination Levels

Radiological control programs require the performance of contamination surveys to determine existing conditions in a given location. Maps with sufficient detail to permit identification of original survey locations should be maintained. Records should contain sufficient detail to be meaningful even after the originator is no longer available. Contamination surveys should be recorded on appropriate standard forms and include the following common elements:

- Date, time, and purpose of the survey

- General and specific location of the survey

- Name and signature of the surveyor and analyst

- Pertinent information needed to interpret the survey results

- Reference to a specific Radiological Work Permit if the survey is performed to support the permit [see DOE standard, Radiological Control part 751.1 (DOE, 2008a)].

Records should be maintained to document changes in monitoring equipment, techniques and procedures [see DOE standard Radiological Control, part 751.2 (DOE, 2008a)].

In addition, records of contamination surveys should include, at a minimum, the following information:

- Model and serial number of counting equipment
- Contamination levels (using appropriate units) and appropriate supporting parameters, including counting efficiency, counting time, correction factors, type of radiation, and whether the contamination was fixed or removable
- Location of areas found to contain hot particles or high concentrations of localized contamination
- Follow-up survey results for decontamination processes cross-referenced to the original survey (see DOE standard, Radiological Control part 754 (DOE, 2008a)).

Records for the release of material and equipment from radiological areas to controlled areas should describe the property, the date on which the release survey was performed, the identity of the individual who performed the survey, the type and identification number of the survey instrument used, and the results of the survey shall be documented (10 CFR 835.703(c)). Additional details on radiation records can be obtained from Chapter 13 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a).

All skin and personal property contaminations should be documented and evaluated to help improve the contamination control program. Documentation should include the following:

- The person's name and work group
- The location, amount, and type of skin or personal property contamination
- The results of decontamination
- A description of circumstances involved in the occurrence, such as radiation work permit number, protective clothing required, and protective clothing actually used.

4.2.3 Characteristics of Plutonium Contamination

There are few characteristics of plutonium contamination that are unique. Plutonium contamination may be in many physical and chemical forms. (See Section 2.0 for the many potential sources of plutonium contamination from combustion products of a plutonium fire to radiolytic products from long-term storage.) One characteristic of plutonium is its ability to migrate with no apparent motive force. Whether from alpha recoil or some other mechanism, plutonium contamination, if not contained or removed, will spread relatively rapidly throughout an area. This is especially true for plutonium newly released to the environment. For plutonium contamination in soil, after contact with soil, the plutonium tends to have its migration hindered and thus moves slower.

4.2.4 Monitoring

Radiation workers are often assigned tasks that conceivably could expose them to radioactive material. It is not sufficient to rely exclusively on equipment design to minimize contamination and exposure in the workplace. A radiation protection program shall include both monitoring of the workers (discussed in Section 4.3) and monitoring of the conditions in the workplace (10 CFR 835 Subpart E). Both functions are essential to a good radiation monitoring program.

Continuous radiation monitoring should be provided during the periods of high or unusual risk associated with the work in the area. Periods of high or unusual risk include the potential or actual breaching of the integrity of the glove-box or associated systems, including such maintenance as replacement of panels, glove changes, bag-out operations, replacement of filters, or repair of vacuum systems. Work that involves the use of temporary enclosures (greenhouses or glovebags) may also be provided with continuous coverage by an RCT, if the hazard is sufficient to warrant such measures. For decommissioning, most activities will be new, unique, and have no historical precedent. Consequently, high and unusual risks may become the norm and the use of temporary controls and continuous coverage the routine.

Monitoring of the workplace is an essential element of every routine surveillance program. It can be effectively accomplished using any or all of the techniques that are discussed in this section. The rigor with which all of the various elements of a radiation monitoring program are applied should be tailored to meet the needs of the individual work areas and should depend on the kind and quantity of radioactive material present and its potential for dispersion. Each program should be designed to meet existing needs, but also should be flexible to allow for incorporation of the possible advantages to be provided by the various available monitoring practices. Monitoring practices include, but are not limited, to the following:

-- Contamination surveys of the workplace

-- Release surveys

-- External exposure surveys

-- Airborne contamination surveys

-- Routine surveillance by an RCT.

4.2.4.1 Contamination Surveys of the Workplace

The radiation monitoring program should include documented survey procedures, a system for maintaining survey results, and contamination control limits for “fixed” and “removable” contamination. The results of contamination surveys should be reported in activity per area (e.g., dpm/100 cm^2) except for large-area swipes and swipes of very small items. This permits interpretation of the recorded data without requiring knowledge of instrument efficiency or geometry.

All workplaces should be monitored for contamination levels on a regularly scheduled basis. The frequency of such surveys will depend on the potential for dispersion of the radioactive material. As a minimum, all gloves, work surfaces, floors, equipment, etc., within the workplace should be surveyed according to the frequencies listed in the DOE standard, Radiological Control (DOE, 2008a).

The change room and other support facilities within the controlled area should be surveyed for contamination daily. Continuous air monitors, survey instruments at step-off pads, and hand and shoe counters should be functionally tested daily or once per shift in support of the weekly and monthly surveys. These frequent surveys are also part of the routine surveillance program and permit immediate follow-up if low-level contamination is detected to minimize the potential for major incidents. Some fixtures and support areas outside the controlled area, such as door knobs and telephones of adjacent offices and the lunchroom, should also be surveyed daily. Other support areas should be surveyed monthly. If routine survey results detect any contamination in a given area, more detailed surveys shall be performed to determine the extent of the contamination. An investigation should be initiated to determine the source of the contamination and the cause.

To preclude the possibility that contaminated waste would be disposed of as ordinary waste, (1) all process and controlled area waste should be considered contaminated, and (2) mechanisms should be established that prevent the mixing of contaminated and noncontaminated waste.

4.2.4.2 Release Surveys

For transuranic radionuclides, the contamination level (fixed and removable) at which surfaces are considered contaminated are listed in Appendix D of 10 CFR 835 (DOE, 2007a). That document also specifies the criteria for the release of materials and equipment to Controlled Areas.

This document concerns release to controlled areas only. The detailed requirements for unrestricted release of materials and equipment are found in DOE Order 458.1, Ch 2 (DOE, 2011c). Refer to that document for guidance regarding unrestricted releases.

4.2.4.3 External Exposure Surveys

To delineate the levels involved, measurements of external exposure should be made at the time a program is established at all locations where personnel exposure occurs. Additional photon and neutron measurements should be made at the same frequency as the contamination surveys. The buildup of plutonium contamination in glove boxes and on gloves and equipment may contribute substantially to the external dose rates.

4.2.4.4 Measurement and Survey Techniques

This section discusses four types of contamination surveys that are typically used in DOE facilities. Surveys for removable contamination include a large-area wipe survey and a technical swipe or smear survey. Surveys for total/fixed contamination include a scan survey and a statistically based survey. These surveys, or a combination of them, are used to survey material for release from radiological control. The appropriate use of each type of survey is discussed.

Surveys for Removable Contamination

Two types of surveys are used for removable contamination: a large-area wipe survey and a technical swipe or smear survey.

A large-area wipe survey is used to detect gross removable contamination. A large-area wipe survey is typically performed using a large floor cloth and a dust mop type handle to wipe large areas. This technique tends to concentrate any low levels of removable contamination that may be present. The surface to be wiped and the wiping material should be industrially clean (i.e., free of debris, grease, etc.) to reduce self-absorption of alpha contamination. The buildup of material (radioactive or otherwise) that would attenuate alpha radiation needs to be considered in establishing the size of large-area wipe surveys. The survey is performed by wiping the surface of the area being surveyed and conducting frequent checks of the cloth using a portable instrument. For detection of alpha-emitting isotopes, a nonabsorbent material should be used. Removable contamination will be accumulated and concentrated on the wipe, increasing the probability of its detection. Checking for contamination is conducted by placing an alpha measurement instrument approximately 0.25 in. (0.6 cm) from the surface of the wipe for 5 seconds, and the count rate observed. If there is no increase above background, then the wipe may be placed in contact with the detector. If radioactivity above background is measured, the material is contaminated. Depending upon the specific circumstances, a series of technical smears may be required to locate and quantify the contamination within the area covered by the large-area wipe. In most instances, if contamination is detected on the large-area wipe, decontamination should be considered.

For transuranic radionuclides, the guideline values for removable contamination may be lower than the detection limit or MDA (minimum detectable activity or amount) of the portable instrument. See G 441.1-1C, Ch. 1(DOE, 2011a) for a discussion on MDA. If this is the case, the surface area of wipe surveys needs to be large enough that the quantity of radioactivity collected on the swipe will be greater than the detection limit of the instrument. Wipe surveys of areas smaller than this minimum surface area require more sophisticated measuring instruments, such as a scaler measurement, and the entire surface of the material should be wiped. The minimum area (A) for using a large-area wipe survey is given by:

$$A = \frac{MDA}{L} \text{ x } 100 \text{ cm}^2 \qquad (4.1)$$

where L is the removable surface radioactivity value in dpm/100 cm^2 of the potential contaminant, given in Table 4.1. and MDA is in dpm.

The purpose of a technical smear survey is to locate and quantify removable contamination that is known or suspected to exist. For small items, a technical smear may be used at any time to verify the item's contamination status. A technical smear or swipe survey is performed by wiping a cloth, paper, plastic foam, or fiberglass disk over a 100-cm^2 area of the surface. The wipe should be taken with a dry medium using moderate pressure. A common field practice is to use two fingers to press the wipe medium against the surface to be wiped. The wipe is then moved along an “S” shaped path that has a nominal length of 8 in. (20 cm) to 10 in. (25 cm).

When the potential contaminant emits alpha radiation, paper or fiberglass filter papers should be used to assure that alpha activity is not attenuated by becoming imbedded in the wipe. To improve the MDA, smears may be taken over areas larger than 100 cm^2. However, for purposes of demonstrating compliance with contamination limits, 100-cm^2 smears need to also be taken and evaluated. The size of the area smeared should be limited to prevent buildup of material (radioactive or otherwise) that would attenuate alpha radiation. Appropriate corrections should be made for objects smaller than 100 cm^2.

If contamination is detected during a scan survey for fixed contamination, a survey for removable contamination should be performed to determine if the contamination is fixed and to quantify any removable contamination. The survey should be performed using a small piece of absorbent material, such as a standard paper smear. This type of survey for removable contamination is often called a technical smear survey. If no contamination above the values for removable contamination in Table 4.1 is detected during the smear survey, the contamination is fixed, and the area should be appropriately marked.

In addition, a technical smear survey may be used routinely to detect removable contamination, especially for contamination surveys of radiological areas.

Scan Survey for Fixed Contamination

A scan survey for fixed contamination requires passing a portable instrument over the surface of the area being surveyed at a fixed, known scan speed and at a specified distance from the surface. Typically, the scan speed is 1 - 2 in./s (2.5 - 5 cm/s) and the maximum distance is 0.25 in. (0.6 cm) for alpha contamination instruments, but this can vary depending on the instrument, probe configuration and background. A scan survey should be used to survey material that resides in an area controlled for contamination purposes, an area where unsealed radioactive sources are used, or a radiological buffer area surrounding an area controlled for contamination purposes. A scan survey in conjunction with a wipe survey should be used to release from radiological control material with a total surface area less than 5 ft^2 (0.46 m^2). A statistically based survey, which will be discussed later, should be used to release from radiological control material with a surface are greater than 5 ft^2 (0.46 m^2).

During the performance of scan surveys, the audible response of the instrument is faster than the needle deflection (some instruments do not have a needle, e.g., digital read out instruments). Therefore, audible response should be used in conjunction with meter readings. For alpha surveys, the surveyor should pause for 3 to 5 seconds each time an individual pulse is detected in order to allow a longer count time at the location of the detected pulse, until it is determined whether the response indicates random background noise or detected contamination.

Several important factors affecting scan survey detection sensitivity are: instrument detection efficiency, background, size of the effective probe area, and the speed at which scan surveys are performed. For a given instrument, scan speed can be a critical factor as counting time is inversely proportional to scan speed. For instruments with larger detector faces, the scan speed is faster for a given rate of meter movement because a point on the surveyed surface remains beneath the window longer. To ensure that low levels of contamination can be detected, it is necessary that a maximum scan speed be mandated and that this speed be implemented during field measurements. As noted above, a typical scan speed for instruments in current use is 1 - 2 in./s (2.5 - 5 cm/s). However, the scan speed for a specific application should consider the instrument, probe, guideline value, and confidence level desired. The MARSSIM (DOE, 2000) contains guidance for determination of scan rates. It also suggests that an empirical method be used to verify scan rates. The equipment and method used in this determination may be incorporated into training for survey personnel to enhance their survey skills.

Similarly, the Multi-Agency Radiation Survey and Assessment of Materials and Equipment (MARSAME) document for releasing equipment such as earth movers, trucks and other equipment has been published (DOE 2009b).

4.2.5 Release Criteria

The release of material from radiological areas shall be performed according to 10 CFR 835.1101. In these areas, material and equipment should be treated as radioactive material and should not be released from radiological areas to controlled areas if either of the following conditions exist:

-- Measurements of accessible surfaces show that either the total or removable contamination levels exceed the values specified in Table 4.1

-- Prior use suggests that the contamination levels on the inaccessible surfaces are likely to exceed the values specified in Table 4.1.

Wire rope and electronic gear with cooling fans are examples of equipment that are difficult to survey and require special procedures to be released from contaminated and airborne radioactivity areas. Additional release criteria can be found in Section 4 of the DOE standard, Radiological Control (DOE, 2008a).

It may be noted that Appendix D of 10 CFR 835 allows that surface radioactivity values be averaged over 1 m^2 provided that the activity in any 100 cm^2 is not more than three times the specified value.

The material release methodology has four main components: material evaluation, scan survey for fixed contamination, large-area wipe survey for removable contamination (described above), and statistical survey for fixed contamination. The material evaluation process involves consideration of the previous known uses of the material, as well as typical uses and the environment in which the material was used. Material evaluation places the material into one of two categories: not potentially contaminated or potentially contaminated.

Non-radioactive material can be released without an instrument survey if its documented history ensures

- That it has never been used or stored in an area controlled for contamination purposes (i.e., a Contamination Area, High Contamination Area, or Airborne Radioactivity Area)
- That it has never come into contact with unsealed radioactive material
- That it has not been stored or used in a Radiological Buffer Area (RBA) surrounding a Contamination Area, High Contamination Area, or Airborne Radioactivity Area.

This material may be considered to be not contaminated and an instrument survey is not necessary according to the DOE standard, Radiological Control (DOE, 2008a). A material history release form should be used to document the release of material that is known to be free of contamination by its history of use. If the material history release form cannot be completed, or if the history of the material is unknown, an instrument survey shall be made of the material. Material released from RBAs around Contamination Areas, High Contamination Areas, or Airborne Radioactivity Areas should also be evaluated using an instrument survey. Material cleared for unrestricted release from a radiological area shall be done in accordance with DOE O 458.1.

The material evaluation process should also consider the nuclides to which the material was potentially exposed. If the material was exposed to significant quantities of nuclides that are difficult to detect, including tritium, ^{14}C, ^{125}I, or ^{129}I, an appropriate survey methodology should be applied.

4.2.6 Plutonium Contamination Detection

The detection and measurement of plutonium contamination is necessary to ensure control of contamination and compliance with DOE requirements. Typically, detection of plutonium contamination has been performed using survey instruments that detect the alpha activity. Routinely used health physics instruments (i.e., alpha survey instruments) may not be adequate for some D&D operations. Self-absorption of plutonium alpha particles within the source or in an irregular surface area may require the use of special X-ray and low energy photon instruments (e.g., a NaI detector). The NaI detector should also be used to detect plutonium contamination that has been painted over.

Discussions of methods used to detect plutonium contamination for past D&D operations can be found in publications by Umbarger (1982) and West et al. (1991). Umbarger reported on nondestructive assay techniques (including portable field instrumentation and laboratory-based methods) for sorting waste in low-level (class A) and TRU waste. Portable field instruments included the field FIDLER (i.e., thin NaI detector), phoswich detector (i.e., thin NaI detector coupled with a thicker CsI detector), ZnS alpha scintillation detector, a portable multichannel analyzer, and a hand-held gamma-ray spectrometer gun. The advantage of a phoswich detector over a NaI detector is its lower operating background. Laboratory-based systems include active and passive gamma-ray spectroscopy, passive neutron detection, and pulsed portable neutron generator interrogation.

During the decommissioning of a mixed-oxide fuel fabrication facility, West et al. (1991) used a nondestructive assay system to provide criticality safety monitoring, track the plutonium inventory, provide measurement of decontamination effectiveness, and provide quantitative characterization/assay of the waste. The system consisted of an integrated set of two passive neutron networks, two pulsed active neutron units, a high-resolution gamma spectrometer [high-purity germanium (HPGe)], and a neutron-coincidence counting unit. Waste determined to be less than 10 nCi/g was certified as class A low-level waste (LLW).

4.2.7 ALARA Guidelines

Contamination levels should be maintained ALARA to minimize the potential for the spread of contamination and to reduce the protective measures and equipment required. Control of radioactive material at the source and prevention of the generation of contamination are more effective and less costly than remediation.

4.3 PERSONNEL CONTAMINATION CONTROL

As described earlier, the purpose of contamination control is to prevent the ingestion or inhalation of plutonium by workers. This is primarily achieved by the engineered barriers discussed previously, containment, confinement, and ventilation control. Only if the primary controls fail or if there is a potential for personnel contamination during an activity are administrative controls such as protective clothing and respirators advisable.

4.3.1 Monitoring Philosophy

Monitoring the worker is necessary, not only to ensure that a potential intake is detected promptly and that the resulting internal dose is assessed, but to confirm the integrity of the engineered containment system and ensure the effectiveness of the overall radiation protection program.

There are several types of worker monitoring, some during and immediately following work with radioactive material and some scheduled for a later time at a preset frequency. This section addresses only methods of monitoring the worker at the workstation. Other methods are discussed in the section that deals with internal and external exposure controls.

Techniques to monitor the individual worker at the work site include:

-- Frequent/routine surveys of gloves

-- Exit surveys

-- Nasal swipes

-- Personal air sampling.

4.3.2 Monitoring Program

Instrumentation shall be provided and persons entering a plutonium work station shall be required to survey themselves at established frequencies. The requirements for radioactive contamination control and monitoring are found in 10 CFR 835.1102. As a minimum, workers should survey their gloves and coverall sleeves each time they are withdrawn from a glove box (or similar containment system) and after each glove replacement or bag-out operation.

Personnel monitoring for contamination should be mandatory at the egress from controlled areas and should be conducted in a verifiable manner. Assurance should be provided that personnel are monitored prior to breaks, meals, or exits from the plant site. Portal monitors, hand-and-shoe counters, and/or portable survey instruments may be used for this purpose. If employees are instructed to perform self-monitoring, the equipment should be set up in a “go/no-go” mode and employees should be clearly instructed in the required actions to take if predetermined action levels are exceeded. Frequent audits should be performed to verify that controls are adequate. Limiting the number of egress points and controlling personnel movement can minimize the numbers of locations where positive control of personnel monitoring shall be maintained.

4.3.3 Protective Clothing

Various types of protective clothing, including laboratory coats, shoe covers, gloves, coveralls, plastic or rubber suits, and air-purifying or atmosphere-supplying respiratory protective equipment, may be required for operations with transuranic radionuclides. The use of company-issue shoes and clothing for employees with work assignments in process areas can be a major aid in contamination control. Recently, some facilities are using disposable anti-contamination clothing. This may be a cost savings from a handling standpoint. However, disposal costs shall be considered. Additionally, consideration should be given to the potential for heat stress.

4.3.3.1 Requirements for Routine Operations

As a minimum, personnel who handle or work with unsealed sources of plutonium should wear coveralls, gloves, and shoe covers. For inspections or visits, lab coats and shoe covers may be permissible in those same areas. When contaminated wet areas are to be entered, water-repellent (plastic or rubber) clothing should be worn. No personal outer clothing should be permitted under coveralls.

When working with unsealed plutonium sources or in glove boxes, hands should be protected by a minimum of two barriers, for example, at least one pair of surgeon's gloves and one pair of glove-box gloves. Where manual dexterity is not required and the work involves a potential for piercing one or both layers of rubber gloves, leather gloves should be worn over the surgeon's gloves. Automated methods should be considered for replacing routine manual methods that have a high risk of piercing the gloves.

Protective clothing should be removed at the exit to radiologically controlled areas and personnel monitoring for contamination performed. If for some reason this is not practical, the movement of personnel should be strictly controlled from the exit area to a location where protective clothing can be removed.

4.3.3.2 Requirements for Special Maintenance

For special maintenance work that involves significant quantities of plutonium, a double barrier concept should be implemented. An example of minimum requirements for protective clothing is provided below:

- Two pairs of coveralls (and sometimes a plastic suit)
- Canvas boots taped to the inner pair of coveralls, with rubber boots over the canvas boots
- One pair of surgical gloves taped to the inner coveralls, with a leather, cotton, or rubber outer pair of gloves
- Respiratory protective device with hood taped to respirator.

To create a double barrier between the source and all extremities, surgeon gloves should be worn in addition to the glove-box gloves. In general, black Neoprene gloves are the standard glove-box glove and the most economical to use where process conditions do not produce rapid glove deterioration. However, alpha particles from surface dust layers can induce surface cracking in black Neoprene. Hypalon 0 is more resistant to surface cracking, acid deterioration, and ozone effects, and this characteristic will, in many cases, make Hypalon gloves the most economical, despite their higher unit cost.

In recent years many new types of glove-box gloves have been developed. Glove usage should be tailored to the particular needs of the job. For processes that require maximum dexterity, the 0.014-in. (0.038-cm) Neoprene gloves are still superior. Coated Hypalon gloves are superior to Neoprene for glove-box process operations that involve nitric acid or ozone levels that may cause deterioration. Ethylenepropylenediamine monomer (EPDM) gloves are used in some facilities and have good flexibility and are resistant to degradation caused by radiation and ozone. Greenhalgh et al. (1979) reported that Hypalon and EPDM gloves have greater than 30 times the longevity of Neoprene in low-level ozone concentration atmospheres. Viton gloves have proven to have a longer life than Neoprene gloves under many operating conditions, but suffer somewhat from stiffness. Where high gamma radiation levels are encountered, lead-loaded gloves may be necessary. However, their stiffness and workers' loss of manual dexterity should be considered in determining their influence on work efficiency and the total dose received.

Persons who perform operations that involve microspheres of ^{238}Pu, coated or uncoated, should be aware that the heat generation of a single 100-µm- to 200-µm-diameter sphere can melt through glove material. In addition, containment of a quantity of microspheres, especially coated microspheres, is difficult because of electrostatic repulsion. Microspheres have been observed climbing the walls of a glass beaker and spreading throughout a glove box.

Glove storage problems occur occasionally. Experiments and static tests have not provided an adequate explanation of the sporadic problems that have been encountered. Test results in which gloves were stored under different lighting conditions (ultraviolet and fluorescent) and under stressed conditions (creased or bent) have not been consistent. Tests of gloves seem to indicate that glove degradation is caused by the combined effect of ionizing radiation, ozone, and lighting. The glove inventory should be rotated to prevent the inventory from becoming outdated while on the shelf.

All gloves in normal use at plutonium processing installations should be inspected prior to each use. All operating personnel should perform contamination self-surveys after every glove usage. The glove inspections should be made each time by the same team of trained individuals, and the condition of each glove should be recorded so that glove failures can be anticipated and preventative measures can be taken. The development of a statistical basis for establishing the frequency of glove changes should be considered because such a basis may be cost-effective. For example, the change-out frequency could be planned so that gloves are changed at some fraction of the mean time between failures or more preferably some fraction of the minimum time between failures. This type of change-out program could also minimize personnel doses and potential contamination spread incidents associated with too-frequent glove replacement. This procedure may require that each glove use be categorized. A routine replacement program will not replace an inspection program, but it is a supplement to the inspections. The inspector's surgeon gloves should, of course, be surveyed after the inspection of each glove-box glove. Gloves that are in questionable condition should be changed without delay. Gloves that are not in use for the remainder of that shift should be capped off with a glove cover or plastic bag.

Gloves not in use should be stored inside the glove box in such a manner that they do not interfere with operations.

4.3.4 Respiratory Protection

Respiratory protection should be readily available. Respiratory protective equipment should be used for all bag-out operations, bag and glove changes, and any situation involving a potential or actual breach of confinement. Alternatively, the operation could be performed in a glovebag to maintain confinement. In any case, protection, in the form of air-purifying or atmosphere-supplying respirators, should be considered whenever concentrations of radionuclides in the air are likely to exceed 30% of the Derived Air Concentration (DAC) (i.e., where an individual without respiratory protection could receive 12 DAC-hrs in a week). For good performance, the respirator shall fit closely on the facial contours and make an impenetrable seal so that all air enters through the filter or is supplied by the breathing-air supply. ANSI Z88.2-1992 (ANSI, 1980b) describes qualitative and quantitative tests that should be used to ensure that the respirator fits the individual; only the quantitative test should be used for verification of respirator fit at plutonium facilities. Respirator fit tests should be performed annually.

The respiratory protective device selected should provide a protection factor appropriate for the air concentration anticipated. ANSI Z88.2 provides protection factors guidance.

Air-supplied hoods are becoming more popular because a fitting is not required and facial hair does not prohibit their use. Protection factors up to 1000 are allowed for air-supplied hoods. All respirators, including air-supplied hoods, require approval. While NIOSH approves most respirators, some respirator types in use at DOE facilities are not part of the NIOSH testing program.

4.3.5 ALARA Guidelines

The total dose to an individual and the collective dose to the work force should be ALARA. When applied to personnel contamination or internal intakes, this generally means less than detectable dose with the best available commercial technology.

4.3.6 Personnel Release Criteria

The decision to release personnel with detectable plutonium contamination is made on a case-by-case basis. If the individual is injured and needs prompt medical attention, medical treatment will always take precedence, with compensatory measures made for the protection of medical personnel and facilities. If injuries are absent or do not require immediate attention, decontamination is preferable to ensure that the dose to the contaminated individual and the potential for inhalation by the victim and medical staff are minimized and the spread of contamination is prevented.

In a case where decontamination is incomplete due to injury to the skin or other reasons, the individual may be provisionally released with measures to prevent the spread of contamination. Module 2.13, Radiological Considerations for First Aid, of DOE-HDBK-1122-99 (DOE, 2009a) provides guidance on control of contamination for medical events. Guidance should be provided to the individual on control of potentially contaminated wound dressings if they are removed after the individual is released.

4.4 PERSONNEL DECONTAMINATION

Skin decontamination should be performed by RCTs or other members of the health physics staff. The treatment and decontamination of wounds should be performed by medical staff.

Nonabrasive methods should be used for skin decontamination to protect the tissues from deeper contamination. Masking tape should be used to remove dry contamination. Wet decontamination should be used to remove residual contamination. The skin should be gently scrubbed with soap and water. The following procedure is recommended:

1. Survey the worker to determine the contaminated areas of the skin. Have the medical staff treat and decontaminate breaks in the skin.

2. Wipe loose contamination with a gauze sponge or cotton applicators dipped in mild antiseptic detergent. Do not spread contamination to uncontaminated areas.

3. Rub the skin with the applicators to produce good sudsing.

4. Use soft bristle scrub brushes for fingernails and other difficult-to-clean areas as long as the skin barrier is maintained intact. It may be difficult to decontaminate the cuticles and under the nails.

5. Dry the skin area with cleansing tissue.

6. After the skin is thoroughly dry, survey it for any remaining contamination.

7. If no contamination is detected, apply a good-quality hand cream to prevent chapping.

Another effective nonabrasive decontamination method involves placing the contaminated hand in a cotton glove and then a Latex glove (causing the hand to perspire).

The decontamination factor is the ratio of the initial contamination level to the contamination level after decontamination methods are applied, as determined by survey instrument readings. Nonabrasive methods should be repeated until the decontamination factor between washes drops below 2 or 3 with significant contamination still remaining.

If contamination persists on the skin, a more abrasive decontamination method may be necessary. The decision to proceed with a more abrasive method should be based on the effectiveness of the decontamination. An abrasive soap should be applied with a moist gauze sponge or soft handbrush while rubbing the skin to develop a soapy lather. Care should be exercised to prevent damage to the skin surface.

Liberal irrigation with lukewarm water or saline solution is recommended for eye, nose, and mouth contamination. These procedures are performed by the medical staff to remove contamination.

5.0 INTERNAL DOSIMETRY

Internal dosimetry is an essential part of a quality health physics program at every facility where plutonium is handled or processed. The purpose of an internal dosimetry program is to monitor workplace activities, to assess accidental or inadvertent intakes of radioactive material, and to conduct internal dose assessments from bioassay measurement data.

It is DOE policy that facilities are designed, operated, and remediated to prevent intakes of radioactive materials. Radiological controls for the workplace should ensure that radionuclides are contained and handled properly, and that intakes, if they occur at all, are negligible to the extent achievable with state-of-the-art technology. In spite of excellent design and operation policies, inadvertent intakes of radioactive material can occur as a result of equipment malfunction, failure to follow procedures, or the unanticipated presence of radioactive material.

Experience has shown that the most common route for inadvertent plutonium intake is inhalation. Intakes can also occur by accidental ingestion or by wound contamination. Surveillance programs should be designed to rapidly detect a release in the event of a loss of radioactive material containment. Internal dosimetry programs should be tailored to the needs of each plutonium handling facility so that inadvertent intakes are discovered and quantified and workers' doses are determined by appropriate methods.

When workers are inadvertently exposed to radioactive material, appropriate corrective action should be taken to ensure that control and containment have been re-established. Prompt detection by routine workplace monitoring practices is essential to regaining control after any contamination spread or loss of containment. Prompt workplace indications of potential intake are also crucial to start special bioassay monitoring for intake and dose assessment. An early assessment of the probable severity of an intake and its corresponding dose, preferably within the first two hours of the intake, is needed for decisions on dose reduction therapy and event reporting. For plutonium and americium intakes, the bioassay data necessary for final dose assessment may require long periods of time (many months) to obtain. Until such data become available, ongoing preliminary assessments of intake and dose may be necessary to provide guidance for the administrative and medical management of the workers.

5.1 INTERNAL DOSE EVALUATION PROGRAM

Internal doses are not directly measured but are estimated or calculated based on knowledge of the material to which a worker may be exposed and it’s known or assumed biokinetic behavior. The common approach to internal dosimetry is to calculate an occupational intake based on worker bioassay measurements or workplace air-sample data and assumed breathing rates. Once an intake is calculated, appropriate internal doses to organs and tissues of concern can be estimated by using fundamental dosimetry principles, by various intake-to-dose conversion factors, which incorporate assumed biokinetic models, or by an appropriate computer code. Intake-to-dose conversion factors can be found in ICRP Publication 68 (ICRP, 1994b). Further discussion on intake and dose assessment is provided in Section 5.8.

Participation in internal dose evaluation programs is required by DOE for conditions identified in 10 CFR 835.402(c) (DOE, 2007a). The internal dose evaluation program shall address both general workplace conditions and individual intakes.

Workplace conditions are monitored through air sampling programs as well as contamination surveys. For work that can have variable or changing conditions, more intensive surveillance may be required, using supplemental portable air samplers, continuous air monitors, or personal air samplers.

Individual worker monitoring for intakes is commonly performed using bioassay procedures. Bioassay monitoring includes both direct (in vivo) measurements of radioactivity in the body and indirect (in vitro) measurements of material excreted or removed from the body. Refer to Section 5.7.4 for information on assessing internal exposures from air monitoring data.

10 CFR 835.402(c) (DOE, 2007a) specifies the requirements for participation in a radiological bioassay program. Because most plutonium facilities have a high degree of radiological control and containment for plutonium, chronic exposure to levels of occupational concern is unlikely and it is not considered likely that a worker would incur more than one unplanned intake in a year. Thus, participation in a bioassay program is generally based on the possibility that a single intake causing a dose in excess of 100-mrem committed effective dose CED might occur. Bioassay is also required if an intake is suspected for any reason.

Indications of intake include (but are not limited to) detection of facial or nasal contamination, air monitoring or sampling that indicates internal exposure, or any wound in which contamination is detected or suspected (See Section 5-9 for internal dosimetry recommended indicator and action levels.) The most common internal exposure monitoring program for workers is the bioassay monitoring program, which shall be designed for the specific nuclides and forms of material at a particular facility. Likely candidates for internal exposure monitoring include personnel who may be routinely exposed to surface or airborne contamination, or those identified by the foregoing workplace indicators.

Workplace monitoring for potential internal exposures is performed to verify the adequacy of containment and work practices. This monitoring includes air sampling, continuous air monitoring, personal contamination surveys, and workplace contamination surveys. Facilities are to be designed and operated to minimize internal exposure. Details regarding workplace monitoring and control practices are discussed in Section 4.0.

5.1.1 Performance Capabilities for Internal Exposure Monitoring

Bioassay monitoring programs shall be capable of showing compliance with the 5-rem/year stochastic and 50-rem/year deterministic dose limits of 10 CFR 835.202 (DOE, 2007a). 10 CFR 835.402(c) (1) (DOE, 2007a) identifies 100-mrem CED for all likely intakes as a level above which workers shall participate in a bioassay program. Therefore, ideally, such bioassay monitoring programs should be capable of detecting this level. In fact, this is not technically achievable for most routine plutonium bioassay programs. In order to meet this requirement, reliance shall be placed on workplace monitoring to identify potential intakes at the time they occur so that special bioassay monitoring can be initiated. Routine, periodic bioassay measurements have little chance of detecting a CED of 0.1 rem and can even have difficulty showing compliance with dose limits.

Performance capabilities for bioassay and internal dosimetry programs can be expressed as the minimum detectable dose, based on some combination of minimum detectable activity and frequency of measurement or time post-intake at which the measurement is made. The term “minimum detectable dose” is preferred over any variants of the occasionally encountered terms “dose-missed” or “potentially undetected dose,” which were usually defined as the same thing. The connotation of the latter terms is that of an actual intake which was not detected, whereas the intent was to define a measure of program sensitivity to doses that might have gone undetected had an intake occurred. The preferred term” minimum detectable dose” (MDD) ties the concept to the recognized terminology of minimum detectable activity (MDA). See G 441.1-1C, Ch. 1(DOE, 2011a), definition of MDA, for information on evaluating measurement results below these quantities.

The MDD for a bioassay monitoring program shall meet the aforementioned dose limit requirements of 10 CFR 835.202. A design goal of 100-mrem CED from all intakes of similar nuclides in a year is desirable but unrealistic for a routine program. To meet these requirements, bioassay programs should have measurement sensitivities (i.e., MDAs for bioassay measurements) established based on the material to which workers might be exposed. Examples of such sensitivities are given in Tables 5.1 and 5.2 (O'Connell, 2009) for pure ^{239}Pu monitored by urinalysis and fecal analysis, respectively. Table 5.3 (O'Connell, 2009) provides an example of the ^{241}Am sensitivity required for monitoring a mixture of weapons-grade plutonium, aged 30 years for ingrowth at time of intake. These tables illustrate the difficulty in relying on routine bioassay to demonstrate compliance with the limits and design goal.

Table 5.1. Urine Bioassay Goals[(a)] for ^{239}Pu

	Type M	Inhalation		Type S	Inhalation	
Days Post-Intake	Urine Intake Retention Fraction[(b)]	Dose Limit Goal[(c)] dpm	100-mrem Committed Effective Dose Goal[(d)] dpm	Urine Intake Retention Fraction[(b)]	Dose Limit Goal[(c)] dpm	100-mrem Committed Effective Dose Goal[(d)] dpm
1	2.46E-04	7.36E+00	4.63E-01	2.50E-06	8.23E-01	1.79E-02
7	2.40E-05	7.18E-01	4.52E-02	3.08E-07	1.01E-01	2.21E-03
30	9.51E-06	2.85E-01	1.79E-02	1.72E-07	5.67E-02	1.23E-03
60	8.11E-06	2.43E-01	1.53E-02	1.65E-07	5.43E-02	1.18E-03
90	7.12E-06	2.13E-01	1.34E-02	1.61E-07	5.30E-02	1.15E-03
200	5.12E-06	1.53E-01	9.63E-03	1.61E-07	5.30E-02	1.15E-03
400	3.71E-06	1.11E-01	6.98E-03	1.70E-07	5.60E-02	1.22E-03
1000	2.44E-06	7.30E-02	4.59E-03	1.77E-07	5.83E-02	1.27E-03
10000	6.87E-07	2.06E-02	1.29E-03	8.25E-08	2.72E-02	5.91E-04
20000	4.83E-07	1.45E-02	9.09E-04	5.83E-08	1.92E-02	4.18E-04

(a) The goals reflect the activity in a 24 hour urine void corresponding to either a 50 rem committed equivalent dose or a 0.1 rem committed effective dose.

(b) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

(c) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 50 rem committed equivalent dose limit/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Bone Surface	3.70	0.337

(d) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 0.1 rem committed effective dose threshold/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Effective Dose	0.118	0.031

Table 5.2. Fecal Bioassay Goals(a) for ^{239}Pu

	Type M	Inhalation		Type S	Inhalation	
Days Post-Intake	Fecal Intake Retention Fraction(b)	Dose Limit Goal(c) dpm	100-mrem Committed Effective Dose Goal(d) dpm	Fecal Intake Retention Fraction(b)	Dose Limit Goal(c) dpm	100-mrem Committed Effective Dose Goal(d) dpm
1	1.10E-01	3.30E+03	2.07E+02	1.16E-01	3.82E+04	8.31E+02
7	2.29E-03	6.87E+01	4.31E+00	2.42E-03	7.97E+02	1.73E+01
30	2.81E-04	8.43E+00	5.29E-01	3.51E-04	1.16E+02	2.51E+00
60	1.31E-04	3.93E+00	2.46E-01	1.86E-04	6.13E+01	1.33E+00
90	6.65E-05	2.00E+00	1.25E-01	1.07E-04	3.52E+01	7.66E-01
200	1.42E-05	4.26E-01	2.67E-02	3.32E-05	1.09E+01	2.38E-01
400	4.67E-06	1.40E-01	8.79E-03	2.13E-05	7.02E+00	1.53E-01
1000	1.04E-06	3.12E-02	1.96E-03	1.12E-05	3.69E+00	8.02E-02
10000	2.96E-07	8.88E-03	5.57E-04	9.53E-08	3.14E-02	6.82E-04
20000	2.11E-07	6.33E-03	3.97E-04	3.20E-08	1.05E-02	2.29E-04

(a) The goals reflect the activity in a 24 hour fecal sample corresponding to either a 50 rem committed equivalent dose or a 0.1 rem committed effective dose.

(b) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

(c) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 50 rem committed equivalent dose limit/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Bone Surface	3.70	0.337

(d) Calculated as Goal (dpm) = Intake x IRF x 2220 dpm/nCi, where Intake (nCi) is the 0.1 rem committed effective dose threshold/dose conversion factor and IRF is the intake retention fraction.

The dose conversion factor (committed dose per unit intake) derived from the ICRP Publication 68 Database (ICRP, 1994b) is shown below:

	Type M, rem/nCi	Type S, rem/nCi
Effective Dose	0.118	0.031

Table 5.3. Lung Measurement Bioassay Goals for ^{241}Am as an Indicator of Aged Weapons-Grade Plutonium[(a)]

		Type M Plutonium	Inhalation
Time - days	IRF$^{(d)}$	Dose limit Goal$^{(b)}$ nCi ^{241}Am	100-mrem Committed Effective Dose Goal$^{(c)}$ nCi ^{241}Am
1	1.25E-02	3.39E-02	2.21E-03
3	1.69E-02	4.58E-02	2.99E-03
7	3.10E-04	8.40E-04	5.49E-05
10	1.54E-05	4.17E-05	2.73E-06
30	3.79E-14	0	0
		Type S Plutonium	Inhalation
Time - days	IRF$^{(d)}$	Dose limit Goal$^{(b)}$ nCi ^{241}Am	100-mrem Committed Effective Dose Goal$^{(c)}$ nCi ^{241}Am
1	1.89E-01	1.73E+00	8.43E-02
7	6.01E-02	5.49E-01	2.68E-02
30	4.96E-02	4.53E-01	2.21E-02
60	4.21E-02	3.84E-01	1.88E-02
90	3.79E-02	3.46E-01	1.69E-02
200	3.14E-02	2.87E-01	1.40E-02
400	2.59E-02	2.36E-01	1.16E-02

(a) Defined as a Pu mixture consisting of, by weight %, 93% ^{239}Pu, 6.1% ^{240}Pu, 0.8% ^{241}Pu, 0.05% ^{238}Pu, and 0.05% ^{242}Pu, with 30 years allowed for ^{241}Am ingrowth (Table 5.4). Additional ingrowth of ^{241}Am post intake is negligible.

(b) Intake of a 30 year aged 6% mix of Pu, with the specific activities listed in Table 5.5, giving a committed equivalent dose of 50 rem to the bone surfaces. Calculated as:

Dose Limit Goal (nCi ^{241}Am) =

$$\frac{\text{50 Rem x specific activity (Ci/g) of the } {}^{241}\text{Am in the 30 year aged 6\% Pu Mix x IRF}}{\text{Mixture dose coefficient (Rem/g) x 1.0E-9 Ci/nCi}}$$

Where: Mixture Dose Coefficient (Rem/g) =
∑ (Specific Activity (Ci/g) of a 30 year aged 6% Pu Mix (from Table 5.5) x applicable radionuclide dose conversion factor (Rem/Ci) derived from the ICRP Publication 68 Database (ICRP, 1994b). See table below.

(c) Intake of a 30 year aged 6% mix of Pu, with the specific activities listed in Table 5.5, giving a committed effective dose of 0.1 rem.

100-mrem committed effective dose goal (nCi ^{241}Am) =

$$\frac{\text{0.1 Rem x specific activity (Ci/g) of the } {}^{241}\text{Am in the 30 year aged 6\% Pu Mix x IRF}}{\text{Mixture dose coefficient (Rem/g) x 1.0E-9 Ci/nCi}}$$

Where: Mixture Dose Coefficient (Rem/g) =
∑ (Specific Activity (Ci/g) of a 5year aged 6% Pu Mix (from Table 5.5) x applicable radionuclide dose conversion factor (Rem/Ci) derived from the ICRP Publication 68 Database (ICRP, 1994b). See following table.

Dose Coefficients (Rem/Ci)

Absorption Type/		Pu-238	Pu-239	Pu-241	Pu-242	Am-241
M	Bone Surface	3.40E+09	3.70E+09	7.40E+07	3.60E+09	4.10E+09
S	Bone Surface	3.00E+08	3.40E+08	7.40E+06	3.20E+08	4.10E+09*
M	Whole Body	1.10E+08	1.20E+08	2.15E+06	1.10E+08	1.00E+08
S	Whole Body	4.10E+07	3.10E+07	3.10E+05	2.80E+07	1.00E+08*

* Type M dose coefficient for Americium-241 is used.

(d) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

The problem is simply that the measurement technology is not available to provide the sensitivities required for the 100-mrem goal using routine, periodic measurements at reasonable frequencies. For example, routine Hanford analyses of plutonium in urine has a detection limit of 0.02 dpm/sample, plutonium in feces has a detection limit of 0.2 dpm/sample and americium lung counting has a detection limit of 0.16 nCi (Carbaugh, 2003). As shown in Table 5.1, for material type M, monthly routine bioassay measurements would not achieve a sensitivity for the 100-mrem goal. For material type S, even weekly routine bioassay measurements would not have the sensitivity for the 100-mrem goal. Monthly, or even bi-monthly, fecal bioassay, as shown in Table 5.2, could achieve the requisite sensitivity for the100-mrem goal for material type M (worst case). However, as discussed in Section 5.3.3.2, there are associated difficulties in including fecal analysis as part of a routine bioassay program. As shown in Table 5.3, lung counting for material types M or S would also not achieve a sensitivity for the 100-mrem goal.

Therefore, because the goal of 100-mrem CED cannot, typically, be met through routine bioassay, the radiation protection organization should take the following administrative actions:

-- ensure that adequate control measures are applied to prevent intakes

-- document the adequate control measures for auditing purposes

-- upgrade bioassay measurement systems and workplace monitoring practices to provide state-of-the-art measurements

-- ensure that internal dose assessments use commercially viable technology.

-- ensure workplace monitoring programs are designed to identify potential intakes.

All confirmed occupational intakes of plutonium, regardless of magnitude, should be assessed. The results of all bioassay and other measurements needed to support the quality of measurements and dose assessment should be recorded and maintained. The recording and reporting requirements for internal dosimetry data are set forth in Section 3.7 of this report; however, the following is a summary list of internal dosimetry information for which recording is required:

- -- Total CED from all intakes during a year
- -- committed equivalent dose to organs or tissues of concern from all intakes during a year
- -- magnitude of intake for each radionuclide during a year
- -- data necessary to allow subsequent verification, correction, or recalculation of doses
- -- gestation period equivalent dose to the embryo/fetus from intake by the declared pregnant worker during the entire gestation period.

Radiation exposure records programs shall also provide for the summation of internal and external doses, as required by 10 CFR 835.702 (DOE, 2007a). While the summation process is not necessarily performed under a site internal dosimetry program, it behooves the program to recognize what is required. The following summations are identified by 10 CFR 835.702(c) (5) and (6):

- -- Total effective dose (TED) defined as the summation of effective dose (for external exposures) and the CED
- -- summation of the equivalent dose to the whole body from external exposure and the committed equivalent dose to organs or tissues of concern
- -- cumulative TED
- -- for the embryo/fetus of a declared pregnant worker, the summation of the equivalent dose to the whole body to the mother from external exposure during the entire gestation period and the gestation period equivalent dose to the embryo/fetus from intakes by the mother during the entire gestation period.

Doses should be calculated and recorded for any confirmed plutonium intake. What constitutes a confirmed intake is discussed in Section 5.7. Along with the doses, supporting records shall be maintained, including the bioassay data, assumptions, biokinetic models, and calculational methods used to estimate the doses. These may be included in letter-report dose assessments, databases, technical basis documents, and similar records, either singly or in combination.

5.1.2 Protection of the Embryo/Fetus, Minors, and Members of the Public

The equivalent dose limit for the embryo/fetus of a declared pregnant worker is 0.5 rem for the entire gestation period, defined as the summation of external dose received and internal dose received during the gestation period (not the 50-year committed internal dose). Internal exposure monitoring is required if an intake is likely to result in more than 10% of that limit (i.e., 50 mrem for the gestation period). As discussed in more detail in Section 5.6., providing adequate protection to keep the mother's intakes below the occupational limits will also provide adequate protection for the embryo/fetus. Thus, special bioassay for plutonium or americium related to pregnancy is not required. As a matter of caution, some sites try to obtain baseline bioassays as soon as a pregnancy is declared, with another baseline bioassay following the end of pregnancy. Some sites also offer to restrict pregnant workers from jobs with relatively high potential for occupational intakes.

Minors and members of the public are limited, in part, by 10 CFR 835.207 and 10 CFR 835.208 (DOE, 2007a) to a TED of 0.1 rem/year. Internal exposure monitoring is required if an intake is likely to result in 50% of that limit (0.05 rem). As noted in Section 5.1.1, because bioassay monitoring is not likely to be sufficiently sensitive to identify such intakes on a routine basis, enhanced workplace surveillance or restriction of access may be required.

5.2 CHARACTERIZATION OF INTERNAL HAZARDS

Plutonium can be encountered in a wide range of mixtures, e.g., a pure isotope in a standard solution, a highly variable combination of isotopes in so-called “weapons grade” or “fuels grade” Pu, or commercial spent fuel. In addition, the age of a mixture significantly affects its isotopic composition. As a typical weapons or fuels grade mixture ages, the ^{241}Pu decays to ^{241}Am. Although the mass changes may be quite small, the overall result can be a significant build-up of ^{241}Am radioactivity with time. This buildup can make the mixture somewhat easier to detect by in vivo methods. Table 5.4 shows some example plutonium mixtures which might be encountered in DOE facilities. Isotopically pure forms of radionuclides can also be encountered. Table 5.5 demonstrates the impact of aging on the activity composition of two mixtures. The composition of plutonium in the facility can significantly affect the design and capabilities of an internal dosimetry program. As part of the program technical basis, the plutonium mixtures need to be determined. In addition, determinations should be made at the time of identified incidents of potential intake. Methods for such determination may include radiochemical analysis or chemistry followed by mass spectrometry.

The physical-chemical form of plutonium also affects the internal hazard posed. Oxides of plutonium tend to exhibit inhalation absorption type S behavior, whereas other compounds such as nitrates are assigned absorption type M by the ICRP. However, as noted in Section 2.4.1, extremes have been observed with regard to both highly soluble and highly insoluble forms, leading to the good practice of performing dissolution rate (i.e., solubility) tests on standard materials in a facility.

As plutonium ages in a residual, loose contamination form, such as might be found in old duct work, glove boxes, or other such components, it can be expected to undergo slow oxidation to a more insoluble form. Thus, absorption type S forms of plutonium may be reasonable assumptions of what to expect during many decommissioning operations.

Particle size is an important consideration for inhalation exposures. The normal practice for an aerosol is to identify the activity median aerodynamic diameter and its associated particle-size distribution. Particle sizes of 10 µm or less are considered respirable. It is acceptable to assume a 5-µm particle size for dosimetry purposes because actual particle size information is usually lacking. Particle size data are most readily obtainable for chronic exposure situations.

Unless representative air sampling is performed in the immediate proximity of a worker during abnormal working conditions, the practical likelihood of obtaining good particle-size information is slim.

5.3 SCOPE OF BIOASSAY PROGRAM

The relatively low annual limit on intake of plutonium renders its radiation hazard substantially more restrictive than its industrial hygiene or chemical toxicity hazard. Thus, internal radiation dose or intake monitoring is the appropriate focus of bioassay monitoring.

Table 5.4. Example Plutonium Isotope Mixtures Immediately Post-Separation wt%

Isotope	Weapons-Grade Plutonium (6% ^{240}Pu Mixture)	Fuels-Grade Plutonium (12% ^{240}Pu Mixture)	Spent Commercial Fuel (25% ^{240}Pu Mixture)
^{238}Pu	0.05	0.10	1.49
^{239}Pu	93.0	84.4	59.50
^{240}Pu	6.1	12.4	23.98
^{241}Pu	0.8	3.0	10.33
^{242}Pu	0.05	0.1	4.0
^{240}Am	0.0	0.0	0.0

Table 5.5. Activity Composition with Age for Reference 6% and 12% ^{240}Pu Mixtures (Carbaugh, 2003)

	Reference 6 % Pu Mix $^{(a)}$		Reference 12% Pu Mix $^{(a)}$	
Isotopic Component	Fresh	Aged	Fresh	Aged
Specific Activity In Mixture, Ci/g				
^{238}Pu	8.6E-3	6.7E-3	1.7E-2	1.4E-2
$^{239+240}$Pu	7.2E-2	7.2E-2	8.0E-2	8.0E-2
^{241}Pu	8.2E-1	1.9E-1	3.1E+0	7.3E-1
^{242}Pu	2.0E-6	2.0E-6	3.9E-6	3.9E-6
^{241}Am	5.3E-5	2.0E-2	2.0E-4	7.6E-2
Pu-alpha	8.1E-2	7.8E-2	9.7E-2	9.3E-2
Total alpha	8.1E-2	9.8E-2	9.7E-2	1.7E-1
Activity Ratios				
$^{239+240}$Pu:^{241}Am	NA	3.5E+0	NA	1.0E+0
Pu-alpha:^{241}Am	NA	4.8E+0	NA	2.2E+0
^{241}Pu:$^{239+240}$Pu	1.2E+1	2.7E+0	3.8E+1	9.2E+0

(a) % = nominal ^{240}Pu weight percent in mixtures.
Fresh = 2 weeks of ^{241}Am ingrowth following separation.
Aged = 30 years of ^{241}Am ingrowth following separation.

5.3.1 Classification of Bioassay Measurements

Bioassay measurements can be classified according to the primary reason for their performance. This is a useful practice for historically documenting why a worker participated in a bioassay program. Numerous reasons for bioassay measurements may be defined for specific facilities; some suggested common classifications are as follows:

-- **Baseline measurements** are used to establish a pre-exposure condition, either for a new employee or as a result of a new work assignment. The standard, Radiological Control (DOE, 2008a), recommends baseline measurements if workers are considered likely to receive intakes resulting in greater than 100-mrem CED. It is a good practice to perform such measurements for newly hired employees, intra-company transferees, or workers transferred from facilities where bioassay measurements may not have been required. In addition, baseline measurements can verify workers' status for special work assignments. For plutonium bioassay, baseline measurements made before any occupational exposure can be expected to yield no detectable results using current technology.

Exempting workers from baseline bioassay implies accepting any detectable results as likely attributable to current occupational exposure. However, requiring baseline measurements can potentially impact the schedule of short-term jobs; the time required to obtain a chest count and a large-volume urine sample may add a day or two delay to entry procedures. Moreover, missing a baseline for a long-term employee who will be placed on a routine bioassay program is not likely to be as troublesome as not obtaining a baseline for a short-term worker who provides a termination sample that shows detection of plutonium after the worker has left the site and is difficult to reach for follow-up.

-- **Routine, or periodic, measurements** are performed on a predetermined schedule (e.g., an annual or quarterly frequency).

-- **Special bioassay measurements** are performed as follow-up to unusual routine results or suspected intakes (See Section 5.9 for recommended internal dosimetry indicator and action levels).

-- **End of assignment or termination measurements** are performed following completion of specific work or at the time of termination of employment. The DOE Standard, Radiological Control (DOE, 2008a), recommends that workers who participate in bioassay programs have appropriate termination measurements.

Bioassay classification is important because the purpose of a sample may affect the collection and analysis or monitoring method chosen. For example, single-void urine samples are not adequate for routine monitoring of potential plutonium exposure, but can provide important information for dose-reduction therapy following a suspected intake; samples representative of excretion over a 24-hour period should be collected for quantitative intake and dose assessment. The date of sample collection (and possibly the time of collection) can be very important to special monitoring performed to assess intake. However, these are much less important with regard to periodic monitoring, for which measurements are not expected to show detectable activity and when any detection whatsoever is likely to initiate investigation and special bioassay.

5.3.2 Monitoring Requirements and Selection of Employees

Workers who are considered likely to have intakes resulting in excess of 100-mrem CED are required to participate in a bioassay program. However, because of the extensive radiological control practices for plutonium facilities, including a high degree of engineered barrier containment, no typical plutonium worker is likely to have intakes of 100-mrem CED or more. However, this should not be used as an excuse to exclude workers from routine bioassay. Although no one should be considered likely to have intakes resulting in 100-mrem CED, some workers are at significantly higher risk for incurring an intake than others and should be on routine bioassay.

The workers at highest risk of incurring an intake are the ones in closest contact with the material. Typically, these are the operators, maintenance, and health physics personnel handling plutonium or plutonium-contaminated objects in the course of routine glove-box, maintenance, or decommissioning operations. In the event of containment system failure, or failure respiratory protection devices, it is these workers who will most likely incur exposure and subsequent intake. These workers should be on a routine bioassay program designed to meet the requirements of 10 CFR 835 (DOE, 2007a) as a kind of safety net to identify intakes which might have gone undetected by workplace monitoring.

Other workers (e.g., supervisors, inspectors, observers, guards, and tour groups) who work in or visit a plutonium facility but are not directly working with the material or contaminated objects are at a substantially lower risk for incurring an intake. Although these people may not need to be on a routine bioassay program, they should be subject to participation in a special bioassay program if workplace indications suggest loss of control or containment.

5.3.3 Selection of Bioassay Monitoring Techniques

Bioassay monitoring techniques fall into two broad categories, direct measurement of radioactive materials in the body (in vivo counting) and analysis of material removed from the body for laboratory in vitro analysis. In vivo counting includes measurements of the chest, lung, skeleton, liver, and wounds. In vitro measurements include urinalysis, fecal analysis, and occasionally analysis of tissue, sputum, or blood samples. Methods for in vitro analysis include liquid scintillation counting, fluorescence measurements, gamma spectrometry, chemical separation followed by electrode position, and counting with radiation detectors. Selby et al. (1994) provide a brief overview of bioassay techniques and capabilities. Further discussion of the techniques is provided below.

5.3.3.1 In Vivo Counting

Direct bioassay (in vivo counting) is the measurement of radiations emitted from radioactive material taken into and deposited in the body. Direct bioassay is appropriate for detection and measurement of photons emitted by plutonium and its decay products. Lung, wound, liver, and skeleton counting are examples of in vivo monitoring most commonly used for plutonium and its progeny. Whole body counting, commonly used for monitoring high-energy fission and activation products in the body, is ineffective for direct measurement of plutonium due to the very low energy of photons emitted from plutonium and its decay products unless the plutonium is intimately mixed in a high-energy photon-emitting matrix, such as spent fuel.

Some low-energy x-rays emitted by plutonium decay products are energetic enough to escape the body. When direct bioassay is used, the detection system should be calibrated for the radionuclides to be measured in the appropriate organs. All calibration procedures, calibration records, and quality control data should be maintained. Energies most commonly used for plutonium monitoring are the 17-keV L X-rays and the 60-keV gamma of ^{241}Am. Mixtures of spent fuel material can lend themselves to whole body counting if the ratio of a readily detectable high-energy gamma-emitter (i.e., ^{137}Cs) to plutonium is known.

A plutonium facility should have the capability to detect and assess depositions of plutonium in the lungs of radiation workers. The major objective of lung counting is to provide measurements of suspected intakes triggered by workplace monitoring results. Lung measurements should be made to provide an early estimate of the magnitude of the intake and resulting lung deposition.

Two methods have been used to detect plutonium in the lung: the L x-ray method and the americium-tracer method. The L x-ray method is based on the measurement of L X-rays following the decay of plutonium. This method provides a direct measurement of plutonium. The detection capability of the method may be on the order of tens of nanocuries for plutonium and requires an accurate measurement of the chest wall thickness (because of the large attenuation of the low-energy X-rays by the rib cage and overlying tissues). Other problems that complicate the measurement of L X-rays are (1) the difference in attenuation in muscle and fat, (2) the possibility of nonuniform distribution of the plutonium in the lung, and (3) interferences from radionuclides in other organs or from other radionuclides in the lung.

The americium-tracer method has the advantage of better detection capability for some mixtures of plutonium. The typical MDA for ^{241}Am lung counting is 0.1 to 0.2 nCi. The americium-tracer method depends on the plutonium/americium ratio, which shall be independently determined or estimated for each intake. The detection level for this method with a plutonium/americium ratio of 15 is typically 2-nCi plutonium in the lung. The americium-tracer method also has the advantage of being less affected by attenuation in the chest wall or by variations in the muscle/fat ratio. However, it has the disadvantage of requiring an estimate of the plutonium/americium ratio, both initially and at long times post intake. This ratio may change over time because of ingrowth of ^{241}Am as the decay product of ^{241}Pu or because americium may naturally clear from the lungs and translocate among internal organs at a rate different than that for plutonium.

The most widely used systems for lung counting are high-purity germanium detectors, thin sodium-iodide detectors, phoswich detectors, and proportional counters. Multiple high-purity germanium detectors have advantages over the other detector systems because of their good resolution, allowing better identification of the radionuclide, better detectability, and better background prediction capability. The main disadvantages of germanium detector arrays are their higher cost relative to other types of in vivo detectors and their lower reliability.

Measurement equipment to detect and measure plutonium contamination in wounds should be available at all plutonium facilities. Instrumentation used for this purpose may include thin-crystal NaI(Tl), intrinsic germanium, or Si(Li) detectors. The detection level for plutonium wound measurements is typically 0.1 nCi for ^{239}Pu. Correction for depth due to absorption of photons in the overlying tissues should be considered. Collimated detectors are useful for determining the location of the plutonium in wounds.

Estimates of the depth of plutonium contamination in a wound may be made using solid-state germanium or Si(Li) detectors to measure the relative absorption of the low-energy X-rays emitted by plutonium. Information about depth is important for determining whether tissue excision is necessary to remove the contamination.

5.3.3.2 In Vitro Analysis

The two most common forms of in vitro analysis are urinalysis and fecal analysis.

Urinalysis. Urine sampling provides useful information about the amount of plutonium excreted following an intake. After chemical isolation, the plutonium in urine samples may be determined by various methods including: alpha spectrometry (gas-flow proportional or surface-barrier detection), alpha counting (zinc sulfide or liquid scintillation counting), fission track counting, and mass spectrometry. Analytical procedures for in vitro measurement of plutonium and other radionuclides have been published (Volchok and dePlanque, 1983; Gautier, 1983).

Urine samples should be collected away from the plutonium facility to minimize cross-contamination. Samples should be collected in contamination-free containers; measures should be considered for minimizing plateout on walls of container surfaces (such as by addition of trace amounts of gold, oxalate, or nitric acid).

Fecal Analysis. Fecal analysis is a useful procedure for evaluating the excretion of plutonium and many other radioactive materials because more than half of the material deposited in the upper respiratory tract is cleared rapidly to the stomach and GI tract.

The total fecal plus urinary elimination for the first few days after exposure, combined with in vivo counts that might be obtained, may provide the earliest and most accurate assessment of intake. Fecal samples taken during the second and third day after an inhalation incident are likely to provide the most useful data because the GI hold-up time may vary from a few hours to a few days.

Fecal sampling is primarily a monitoring procedure for confirming and evaluating suspected intakes, but is used at some plutonium facilities for routine periodic monitoring as well. Workers may find fecal sampling unpleasant or objectionable, and laboratory technicians may also have aversion to fecal sample analysis. Some of these problems may be minimized if commercial fecal sample collection kits are used for convenient collection and handling of samples (Fisher et al., 1982). Collection kits also provide a means for collecting uncontaminated samples. Fecal samples may require additional sample preparation before analysis.

5.4 ESTABLISHING BIOASSAY FREQUENCY

The bioassay measurement frequency should be based on 1) the potential risks of an intake occurring and 2) the sensitivity of a bioassay program to detecting potential intakes. The bioassay program sensitivity can be selected using specified intervals between measurements based on the MDD associated with an interval.

The rationale for the selected bioassay measurement frequency should also be documented. It is appropriate to evaluate the probability of intake and to modify the sampling frequency based on that probability.

The frequency of bioassay measurements should normally not be decreased because analytical results are below the detection level. The bioassay program should be maintained to confirm the proper functioning of the overall internal exposure control program and to document the absence of significant intakes of radionuclides.

5.4.1 Frequency Based on Program Sensitivity

The minimum detectable dose concept refers to the potential dose associated with an MDA bioassay measurement at a given time interval post-intake. The pattern of retention of activity in the body, the MDA for a bioassay measurement technique, and the frequency with which that technique is applied define a quantity of intake that could go undetected by the bioassay program. An intake of such a magnitude would not be detected if it occurred immediately after a bioassay measurement and if it were eliminated from the body at such a rate that nothing was detected during the next scheduled measurement. The dose resulting from such an intake would be the MDD for that particular measurement technique and frequency.

Estimates of MDD in terms of CED should be documented for each measurement technique, MDA, and frequency. Retention functions specific to the various chemical forms and particle size distributions found in the facility should be used. Examples of MDD tabulations can be found in La Bone et al. (1993) and Carbaugh et al. (1994). In establishing MDD tables, it is important to consider dose contributions from all appropriate radionuclides in any mixture, rather than just the dose contribution from the bioassay indicator nuclide.

5.4.2 Frequency Based on Potential Risk of Intake

As discussed in Section 5.3.2, although plutonium workers are not generally considered to be at high risk of incurring intakes that might result in CEDs of 100 mrem or more, any plutonium worker can be considered to have the potential for such an intake. However, having the potential for intake does not mean that they are likely to incur an intake.

Workers who have the highest potential risk for an intake are those most closely working with plutonium or plutonium-contaminated material. Typically, these workers are glove-box workers, maintenance workers, and operational health physics surveillance staff. These workers should be on a routine plutonium or americium bioassay program, including urinalysis and in vivo measurements. Such programs are relatively insensitive compared to the 100-mrem CED goal and are a safety net intended to catch intakes of significance relative to regulatory limits, rather than substantially lower administrative levels. Selection of bioassay frequency depends on the facility experience with potential intakes, the perceived likelihood of intake, and the MDD of a program. Annual urinalyses and in vivo chest counts are fairly typical. More frequent (e.g., semi-annual or quarterly) measurements may permit more timely review of workplace indicators in the event that an abnormal bioassay result is obtained, but do not necessarily mean a more sensitive program.

Plutonium facility decommissioning projects may present a different set of challenges for worker protection. In particular it is likely that clean up of areas will involve more plutonium that is not contained than is the case during normal operations. In addition, the workers involved may be relatively transient as the project progress through phases requiring different craft labor mixes. This being the case, more frequent bioassay may be necessary to provide good assurance that dose limits are not exceeded. As discussed in Section 5.3.1, it is likely that program administrators will require a baseline measurement prior to the start of work and another at the termination of work. However, if the worker moves between tasks, it may be difficult to determine the source of an uptake without intermittent bioassay. In such cases, the use of breathing zone air samplers may be appropriate.

5.4.3 Special Bioassay as Supplements to Routine Bioassay Programs

Special bioassay programs for workers with known or suspected acute inhalation intakes of plutonium or other alpha-emitting radionuclides should include both urine and fecal sampling. Special bioassay measurements should be initiated for each employee in a contaminated work area when surface contamination is detected by routine surveillance if it is possible that the contamination resulted in a CED of 100 mrem or greater. Excreta samples should not be collected where they may be contaminated by external sources of plutonium. Ideally, total urine and feces should be collected for about a week following intake. This permits a sensitive assessment of potential intake and internal dose. Longer term special samples collected at various times from a month to a year following intake can help to discriminate between ingestion, absorption type M inhalation, and absorption type S inhalation. See Section 5.9 for indicator levels where special bioassay should be considered.

5.4.4 Long-term Follow-up Bioassay Programs

Following an intake a long-term follow-up bioassay program may be required for a worker to compare the actual excreta or in vivo results with those projected by the evaluation. This is important to verify the accuracy of intake and dose assessments. The frequency and duration of a special program is dependent upon the projected values; it is suggested that as long as a worker continues to have detectable bioassay results, he or she should continue to be monitored. It is particularly important to have good baseline data and projections for individuals who return to plutonium work. The ability of a bioassay program to distinguish between an established, elevated baseline and a new potential intake is important in the continued monitoring of workers once an intake has occurred. Because of statistical fluctuations in low-level plutonium and americium measurements, it can be very difficult to identify a new intake by routine bioassay if a worker has an elevated baseline.

5.5 ADMINISTRATION OF A BIOASSAY PROGRAM

Administering a bioassay program requires that the policies, procedures, materials, support facilities, and staff be in place to enable a bioassay program to commence. Among the administrative items to address are the following:

-- Management policy requiring participation in bioassay program by appropriate workers (may be part of an overall radiation protection policy)

-- implementing procedures (e.g., criteria for who should participate, scheduling, sample kit instructions, sample kit issue/receipt, follow-up to unsuccessful sample or measurement attempts, data-handling)

-- arrangements with appropriate analytical laboratories, including specifications of analysis sensitivity, processing times, reporting requirements, and quality assurance provisions

-- onsite support facilities (e.g., sample kit storage locations, sample kit issue/collection stations, measurement laboratory facilities, equipment maintenance)

-- staff selection, qualification, and training.

Recommendations for testing criteria for radiobioassay laboratories are in Performance Criteria for Radiobioassay, ANSI/HPS N13.30 (ANSI, 2011b). These recommendations include calculational methods and performance criteria for bias, precision, and testing levels.

Some sites have established brief flyers or brochures describing their bioassay measurements. These may be distributed to workers during classroom training, upon notification of scheduled measurements, or at the time of the measurement or sample.

5.5.1 In Vivo Monitoring

The scheduling and measurement process for obtaining in vivo measurements is usually straightforward. Workers are scheduled for the measurements and results are available shortly after the measurement is completed. Counting times for in vivo ^{241}Am measurements range from about 15 minutes to an hour or more, depending on the type of measurement and sensitivity required. The long counting times can impose limitations on the throughput of workers through a measurement facility, making scheduling an important issue. Procedures should be in place to assure that workers arrive for scheduled measurements and that follow-up occurs when a measurement is not completed or a worker fails to show.

Occasionally, workers are found who are claustrophobic when placed inside in vivo counter cells. Leaving the cell door partially open may help reduce some of the anxiety, but will also likely compromise the low background for which the system is designed.

Many workers want to know the results of their measurements. While a simple statement by the in vivo measurement technician may be adequate, a form letter stating that results were normal (or showed no detection of any of the nuclides of concern) can provide permanent verification. If results are not normal, a form letter can also be used to explain what happens next.

An important aspect of any in vivo measurement program is the calibration and verification testing of the measurement equipment. In vivo measurement results are highly dependent on the determination of a background result. Likewise, calibration using known activities in appropriate phantoms is also important. Phantoms are available commercially or by loan from the USDOE Phantom Library, operated by the Radiological and Environmental Sciences Laboratory in Idaho Falls. For information on or to request loans from the USDOE Phantom Library go to the DOELAP website: http://www.hss.doe.gov/sesa/corporatesafety/doelap/index.html.

5.5.2 Urine Sampling

Urine sampling programs can be effectively administered using either workplace or home collection protocols. Workplace sampling protocols shall assure that adequate precautions are taken to prevent external contamination of the sample by levels of activity well below the detection capabilities of friskers and workplace monitors. Home collection protocols have the advantage of being sufficiently removed from the workplace to render as essentially nonexistent the potential of very low-level contamination of the sample from external sources of plutonium. Avoidance of very minor external contamination of the samples is extremely important due to the dosimetric implications of plutonium in urine.

Large-volume urine samples are necessary for bioassay monitoring due to the very small urinary excretion rates. Ideally, 24-hour total samples would be preferred; however, such samples often impose substantial inconvenience on workers, resulting in noncompliance with the instructions. As an alternative, total samples can be simulated by either time-collection protocols or volume normalization techniques.

One method of time-collection simulation (NCRP, 1987; Sula et al., 1991) is to collect all urine voided from 1 hour before going to bed at night until 1 hour after rising in the morning for two consecutive nights. This technique has been reviewed with regard to uranium by Medley et al. (1994) and found to underestimate daily urine excretion by about 14%. Such a finding is not unexpected, since the time span defined by the protocol is likely to be about 18 to 22 hours for most people.

The volume normalization technique typically normalizes whatever volume is collected to the ICRP Reference Man daily urine excretion volume of 1400 mL. Reference Woman excretion (1000 mL/d) may be used for gender-specific programs. As a matter of practicality, routine monitoring programs do not usually use gender as a basis of routine data interpretation, particularly since results are anticipated to be nondetectable under normal conditions.

A third method calls for collection of a standard volume (e.g., 1 liter) irrespective of the time over which the sample is obtained. This method uses the standard volume as a screening tool only for routine monitoring. It does not attempt to relate measured routine excretion to intake, relying on well-defined and timely supplemental special bioassay to give true or simulated daily excretion rates.

The most common sample collection containers are 1-liter polyethylene bottles. Although glass bottles are also used, they pose additional risks of breakage. Wide-mouthed bottles are preferred for convenience and sanitation. The number of bottles included in the kit should be appropriate to the protocol; for a total 24-hour protocol as much as 3 liters can be expected. Special provisions, such as a funnel or transfer cup, may improve the esthetics of sample collection and provide for added worker cooperation.

Some concerns can exist with length of sample storage before analysis. Storage may come from delays before batching samples in-house or due to transportation times to an offsite laboratory. The longer a sample stands, the more chemical and biological change it can undergo, typically manifesting itself as sedimentation and plateout on container walls. While samples can be preserved by acidification or freezing, good radiochemistry techniques should assure essentially complete recovery of any plateout or sediment. Samples sent offsite for analysis can be preserved with acid, but this method imposes hazardous material shipping requirements. Freezing samples can preserve them, but plateout and sedimentation upon thawing should still be expected.

Precautions are necessary if a lab uses an aliquot for analysis and extrapolates the aliquot result to the total sample. The aliquoting procedure should be tested using spiked samples to assure that it is representative.

A QC verification program should exist for laboratory analyses, including use of known blank samples and samples spiked with known quantities of radioactivity. Ideally, the samples should not be distinguishable by the analytical laboratory from actual worker samples. The number of QC verification samples may range from 5% to 15% of the total samples processed by a large-volume program; a small program focused on submittal of special samples following suspected intakes may have a much higher percentage of controls. An additional QC provision may be to request the analytical lab to provide results of their in-house QC results for independent review.

There are no standard or regulatory requirements for bioassay sample chain-of-custody provisions, nor has there been consensus on their need. Tampering with samples has not been a widely reported or suspected problem. Site-specific chain-of-custody requirements should be based on balancing the need with the resources required to implement them. Some sites have no chain-of-custody requirements associated with bioassay sample collection. At other sites, a simple seal placed on a sample container following collection by the subject worker is an effective means of providing a small degree of chain-of-custody. At the more complex level would be strict accountability requiring signature of issue, certification of collection, and signature of submittal.

Procedures describing details of the bioassay program should be documented. These procedures should include a description of sample collection, analysis, calibration techniques, QC, biokinetic modeling, and dose calculational methods used.

5.5.3 Fecal Sampling

A fecal sampling program shall be designed to ensure worker cooperation, whether collecting samples at home or in the workplace. Since the frequency of fecal voiding varies greatly from person to person, the sample collection program shall be adaptable. Flexibility in sample dates is important. It is suggested that when a fecal sample is required, the worker be provided with a kit and instructed to collect the sample, noting the date and time of voiding on the sample label. This practice can reduce the likelihood of unsuccessful samples. If multiple samples are required (for example, to collect the total early fecal clearance following an acute inhalation exposure), the worker may be given several kits and told to collect the next several voidings. the worker should be told to note the date and time of each sample.

Since the total fecal voiding should be collected, thought shall be given to the kit provided. Fecal sampling kits can be obtained from medical supply companies or designed by the site. A typical kit might include a large plastic zipper-closure bag to hold the sample, placed inside a 1- to 2-liter collection bucket with a tight-fitting lid. The bucket and bag can be held in place under a toilet seat by a trapezoid-shaped bracket with a hole through it sized to hold the bucket. After sample collection, the zipper bag is sealed, the lid is snapped tight on the bucket, and the bucket placed in a cardboard box.

Following collection, the sample handling, control, analytical, and quality control (QC) provisions are similar to those described above for urine samples. One particular concern for fecal analysis is the potential difficulty of dissolving class Y plutonium in the fecal matrix. While nitric acid dissolution may be adequate, enhanced digestion using hydrofluoric acid may be preferred.

5.6 MODELING THE BEHAVIOR OF PLUTONIUM IN THE BODY

A key issue to plutonium dosimetry is the modeling of how the material behaves in the body. Some of the standard models are described below, with additional discussion on the biological behavior given in Section 2.4. It is important that an internal dosimetry program establish and document the routine models and assumptions used for dosimetry. Computer codes typically incorporate standard models but may allow the flexibility to alter parameters. When altered on an individual-specific basis, the revised models need to be addressed in the pertinent case evaluations or the technical basis.

5.6.1 Respiratory Tract

The respiratory tract model of ICRP Publication 66 (ICRP, 1994a) may be used for evaluating inhalation intakes of radioactivity. The model has been widely published and internal dosimetry computer codes, hence it is not reproduced here.

Like all models, the ICRP respiratory tract model represents anticipated behavior. Once an exposure has occurred and actual data become available, deviations from the model in light of the data are appropriate.

In practice, the model has proved extremely valuable for calculating derived investigation levels and estimating intakes from bioassay data, using standard F, M, and S absorption types of material. Model interpretation becomes more subjective when extensive data become available. Carbaugh et al. (1991) and La Bone et al. (1992) have provided excellent examples of two cases where the standard lung model assumptions did not fit the data.

Most internal dosimetry computer codes allow adjustment of particle size and selection of solubility classes. Some codes also permit detailed adjustment of the model's individual compartment parameters; with these codes, it may be possible to arrive at various subjective interpretations to explain the same data. When adjustments are made to the standard assumptions, it is important to explain what those adjustments are and why they were made.

5.6.2 Gastrointestinal Tract

The model used in ICRP Publication 68 to describe the behavior of radionuclides in the GI tract and for the calculation of doses from radionuclides in the lumen of the gut is that described in ICRP Publication 30 (1979 and 1988b). This model is also widely promulgated and used for evaluating ingestion intakes. The model is particularly subject to individual variations in fecal voiding frequency, so judgment shall be used in its application to human data.

A key parameter of the model for internal dosimetry is the f_1 factor for absorption to blood of material in the small intestine. The f_1 factor varies from 10^{-5} for plutonium oxides to 10^{-4} for plutonium nitrates and to 10^{-3} for other compounds and americium.

5.6.3 Systemic Retention and Excretion of Plutonium

Standard models for the systemic retention of plutonium are commonly used for internal dosimetry because in vivo detection of plutonium within the individual systemic compartments is not usually possible. Models proposed by the ICRP over a 10-year period are described in Section 2.4.2 of this document. Each of them has had a wide application, and ICRP has suggested that results derived using one model do not need to be rederived for compliance purposes using the newest model. Studies by the U.S. Transuranium Registry and summarized by Kathren (1994) have indicated that alternate compartments and clearance half-times may be more appropriate.

For plutonium absorbed to the blood the main sites of deposition are the liver and skeleton . ICRP Publication 78, Individual Monitoring for Internal Exposures of Workers.(ICRP, 1997) provides parameter values for biokinetic models of plutonium.

Excretion models for plutonium include the empirical models of Langham (1956) and Langham et al. (1980), Durbin (1972), Jones (1985), and Tancock and Taylor (1993), as well as study models such as Leggett (1984). This technical document does not take a position on the “best” model. Site choices of dosimetry tools such as reference tabulations (Lessard et al., 1987; ICRP, 1988a) and computer codes (such as IMBA - Integrated Modules for Bioassay Analysis), may dictate one model over another. The choice of model and explanation of its selection are among the technical bases of the site internal dosimetry program.

Note: The DOE website:
http://www.hss.energy.gov/nuclearsafety/qa/sqa/central_registry.htm
lists "toolbox" codes that are compliant with DOE's Safety Software Quality Assurance requirements. The toolbox codes are used by DOE contractors to perform calculations and to develop data used to establish the safety basis for DOE facilities and their operation, and to support the variety of safety analyses and safety evaluations developed for these facilities. IMBA is included in the DOE toolbox.

5.6.4 Natural Plutonium Balance in Man

Although plutonium can be found in members of the general public as a result of worldwide fallout from atomic weapons detonations, the levels are quite small. A summary of the literature can be found in ICRP Publication 48 (ICRP, 1986). Data from McInroy et al. (1979, 1981) suggests that median body burdens of plutonium in the U.S. population peaked at about 12 pCi during the 1960s and declined to about 2 pCi by 1977. Tissue concentration data from Nelson et al. (1993) can be used to calculate a median body burden in the early 1970s of 3 to 4 pCi.

These body burdens imply that urinary or fecal excretion associated with worldwide fallout will not be detectable by routinely available bioassay procedures. Consequently, it is reasonable to assume that any bioassay detection by a worker-monitoring problem is likely to be attributable to occupational exposure.

5.6.5 Mother-to-Fetus Transfer

Methods for evaluating embryo/fetal uptake have been described by Sikov et al. in NUREG/CR-5631 (1992) and its 1993 addendum (Sikov and Hui, 1993). For uptakes occurring during the first 2 months of pregnancy, the activity in the embryo/fetus is assumed to have the same concentration as in the mother's “other soft tissue.” For later uptakes, the embryo/fetal concentration gradually increases relative to the maternal concentration, but is assumed to remain uniformly distributed in the embryo/fetus. At 3 months, the embryo/fetal concentration is 1-1/2 times the mother's “other” soft tissues concentration. At 6 months, it is twice the mother's, and at 8 months it is thrice the maternal “other” concentration. Following transfer to the embryo/fetus, activity is assumed to remain, without clearance, until birth.

The Nuclear Regulatory Commission has developed simplified methods for assessing the gestation period dose to an embryo/fetus in Regulatory Guide 8.36 (NRC, 1992). Although the models have not been updated to reflect the 2007 amendment to 10 CFR 835 (i.e., use of dosimetric models based on ICRP Publication 60 and later publications), the Regulatory Guide is still useful in illustrating that very large maternal intakes of plutonium or americium are required to produce uptakes that would deliver 500 mrem, or even 50 mrem to the embryo/fetus. The NUREG/CR-5631 Addendum (Sikov and Hui, 1993) notes that maternal inhalation intakes of nominally 100 times the annual limit on intake (ALI) are required to give a 50-mrem embryo/fetal dose. For ingestion intakes, a 1,000 ALI maternal intake of plutonium is required to give a 50-mrem dose to the embryo/fetus. Thus, providing adequate radiation protection to limit maternal intake of plutonium and americium to the occupational limits will adequately provide for the protection of the embryo/fetus.

ANSI/HPS N13.54, Fetal Radiation Dose Calculations, (ANSI, 2008a) and ICRP Publication 88, Doses to the Embryo and Fetus from Intakes of Radionuclides by the Mother, (ICRP, 1998) provide additional guidance on assessing fetal dose. ICRP Publication 88 uses models that have been updated to reflect the 2007 amendment to 10 CFR 835.

5.7 INTERPRETATION OF BIOASSAY RESULTS

Bioassay measurements detecting plutonium or americium in workers can be initially interpreted as indicating that occupational intakes may have occurred. Standard bioassay procedures are not sufficiently sensitive to detect the worldwide environmental background levels in an in vivo or in excreta. Since most plutonium and americium bioassay measurement procedures include counting for radioactivity as the final step in the measurement process, they are subject to the statistics associated with the counting process.

Two key questions associated with bioassay data are (1) When does a sample result indicate the presence of something (i.e., when is the analyte detected)? and (2) What is the overall capability of the bioassay method for continual assurance of detection of the analyte?

The decision level, L_c (also called the critical level for detection), is the level for a given measurement that indicates the likely presence of the analyte. The L_c is dependent on the probability of obtaining false positive results (type I, or alpha, error) that is acceptable to the program. A 5% probability of false-positive results is a common design parameter of measurement programs, implying that for a large number of measurements, 5% of the time results will be indicated as positive when in fact there is no activity present. The L_c is calculated from results of analyses of blank samples. Once a measurement is performed, it is appropriate to compare it with the L_c to determine whether or not the result is “positive” (i.e., the analyte is detected).

The MDA is the level at which continued assurance of detection can be provided. The MDA is a function of the probabilities of both false positive and false negative (type II, or beta) errors and is typically based on a 5% probability for each kind of error. The MDA is also determined from analysis of blank samples, but is substantially higher than the L_c. The MDA is appropriate for use in designing bioassay programs and as the basis for estimating minimum detectable intakes and doses as indicators of program sensitivity. The MDA should not be used as a comparison with actual measurements to determine whether or not activity is present (i.e., <MDA is not an appropriate use of the concept).

Methods for calculating both L_c and MDA are given in HPS N13.30. (ANSI, 2011b).

As an alternative to the L_c and MDA of classical statistics, Miller et al. (1993) propose the use of Bayesian statistical methods for evaluating bioassay data.

General follow-up actions to abnormal bioassay measurements should include data checks, timely verification measurements, work history reviews, and performance of special in vivo measurements or excreta sample analyses for intake and dose assessments.

5.7.1 In Vivo Count Results

In vivo plutonium or americium measurements are generally relatively insensitive with regard to levels of occupational exposure concern. This applies particularly to routine chest or lung counting, skeleton counting, and liver counting. For that reason, any detection of plutonium or americium should be investigated. The investigation should address the validity of the measurement by reviewing the spectrum and its associated background subtraction. These reviews are particularly important if the result is near the L_c. Follow-up to a positive result should include a confirming measurement. Ideally, this should be an immediate (same day) recount of equal or higher sensitivity. The farther removed in time a verification measurement is from the original measurement, the more important it becomes to factor in potential lung clearance in comparing the two measurements. A follow-up measurement taken 30 days after an initial high-routine may not be capable of providing verification if the material of concern exhibits absorption type M behavior.

Chest-wall thickness has a significant impact on chest counting. Corrections are commonly made using a height-to-weight ratio or ultrasonic methods (Kruchten and Anderson, 1990).

Corrections may be required to address apparent detection in one tissue resulting from photon crossfire from another tissue. For example, chest counting is performed primarily to estimate activity in the lung. Yet, there is substantial bone over the lungs (rib cage, sternum) and behind the lungs (vertebrae). Plutonium and americium are both bone-seeking radionuclides which will deposit on those bone surfaces and can interfere with chest counting. It is possible for a person having a systemic burden of plutonium from a wound in the finger to manifest a positive chest count from material translocated to the skeleton, axillary lymph nodes, or liver (Carbaugh et al., 1989; Graham and Kirkham, 1983; Jeffries and Gunston, 1986). Interpreting such a chest count as a lung burden can render dose estimates somewhat inaccurate.

When comparing in vivo measurements made over many years, it is important to make sure that the measurements are, in fact, comparable. One consideration is to make sure that corrections have been consistently applied to all similar measurements. It is not unusual for measurement systems to be replaced or to change the algorithms used for calculating results over time. Step changes in data can occur and should be addressed by monitoring long-term detectable trends (Carbaugh et al., 1988).

In vivo wound counting for plutonium or americium is usually one facet of special bioassay following a wound. While a portable alpha survey meter may show if surface contamination is present at the wound site or contamination of the wounding object, alpha detectors are not capable of measuring imbedded activity or activity masked by blood or serum. Thus, plutonium and americium facilities should have available a wound counter utilizing a thin sodium iodide or semi-conductor (e.g., planar germanium) detector. Such detectors are capable of measuring the low-energy photons emitted from plutonium and americium. The ability to accurately quantify wound activity is highly variable, depending on the calibration of the equipment and how deeply imbedded material is in the wound. If the object causing a wound and blood smears taken at the time of a wound show no detectable activity, then a wound count also showing no detectable activity is probably sufficient to rule out an intake. If the wounding object or the blood smears show detectable activity, special urine samples should be obtained regardless of the wound count result. In this latter circumstance, lack of detectable activity on a wound count could be attributable to deeply imbedded material at the wound site or to rapid transportation of material from the wound to the systemic compartment.

In growth of ^{241}Am from ^{241}Pu in plutonium mixtures can also significantly impact in vivo data interpretation. Rather than decreasing with time, ^{241}Am results can increase without additional intake. This circumstance is particularly likely if dealing with residual activity bound up in wound sites, but may also be observed by in vivo chest or skeleton counting. A method to evaluate ^{241}Am in growth is described is Section 5.8.4.

5.7.2 Urine Sample Results

Detection of plutonium or americium activity in a routine or special urine sample using commonly available radiochemical measurement techniques should be investigated as a potential intake. A data review should be made to assure that the sample result was correctly determined, and batch quality control sample data should be verified.

If the result is near the L_c, it is possible that statistical fluctuation of the measurement process could account for the apparent detection. Recounting the final sample preparation once or twice can be a helpful technique to verify a result or classify it as a false-positive. If the first recount also detects the analyte, it can be concluded that the sample does contain the analyte (the likelihood of two consecutive false positives at a 5% type I error per measurement is 0.0025, or 0.25 %.) If the first recount does not detect the analyte, a second recount can be performed as a tie-breaker.

An investigation should be initiated for any abnormal plutonium or americium urinalysis result. “Abnormal” for a person with no prior history of intake should be interpreted as any detectable activity.

Once an intake is confirmed, sufficient samples shall be obtained to establish a reasonably anticipated baseline against which future measurements can be compared. This is important both to provide future verification of the accuracy of the assessment and to identify potential additional intakes.

The statistical fluctuation of low-level measurements can be particularly troublesome for long-term excretion patterns. Factors of 2 can be easily expected due to day-to-day variability and imprecise adherence by the worker to urine collection protocols.

5.7.3 Fecal Sample Results

Fecal samples are much more sensitive to detection of intakes than are urine samples and, consequently, are an important part of follow-up bioassay monitoring for potential intakes initially identified by workplace indications. Pitfalls to the data interpretation include highly variable individual fecal voiding patterns, ranging from more than one per day to one every few days. This makes it extremely important to know what time interval is represented by a collected fecal sample. While normalizing a single set of fecal data to reference man daily excretion rate can be done, it is not likely to improve the quality of assessment.

The preferred fecal sampling protocol following an intake is to collect all the early fecal clearance (meaning total feces for the first five-to-seven days). This method will allow a good estimation of inhalation or ingestion intake, but does not readily permit discrimination of inhalation from ingestion, or identify whether inhaled material exhibits absorption type F, M, or S clearance patterns. For optimum interpretation, total fecal collection should be interpreted in light of early urine and in vivo data for preliminary estimates. The urine data is likely to be particularly valuable in conjunction with fecal data to classify an intake as absorption type M or S. Longer-term follow-up fecal samples at nominally 30, 60, and 90 days post intake should substantially improve the classification of material as absorption type M or S.

Fecal sampling can also be applied to monitor excretion at long times post-intake. One caveat in such sampling is that a worker still active in a plutonium facility may be incurring very minor chronic exposure, which can significantly interfere with long-term interpretation of acute exposure data. Bihl et al. (1993) have discussed experience with a routine fecal sampling program.

5.7.4 Use of Air Sample Data in Internal Dosimetry

Results of air sampling and continuous air monitoring implying more than 40 DAC-hours exposure should be used to initiate special bioassay to assess intakes of plutonium. Although bioassay data are the preferred method for assessing intakes and internal doses, air sample data can be used for assessing internal doses if bioassay data are unavailable or determined to be inadequate or nonrepresentative. Air sample data can be used to calculate an exposure to airborne material either in terms of DAC-hours or potential radioactivity intake as follows:

$$\text{DAC - hours} = \frac{\text{Air Concentration}}{\text{DAC}} * \text{Duration (hours)} \tag{5.2}$$

DAC = The airborne concentration for radionuclides listed in Appendix A of 10 CFR 835, taking into consideration the absorption type (F/ M/ S) expressed in μCi/mL or Bq/m^3

$$\text{Intake} = \text{Air Concentration} * \text{Breathing Rate} * \text{Time} \tag{5.3}$$

Air concentration = airborne radioactivity in units of μCi/mL or Bq/m^3

If air sample results are representative of air breathed by individuals, then doses can be calculated using the 5-rem stochastic limit for CED (E_{50}) or the 50-rem deterministic limit for committed equivalent dose ($H_{T,50}$) and the respective stochastic or deterministic DAC or ALI conversion factor, as shown below:

$$E_{50} \text{ or } H_{T,50} = \frac{\text{(\# of DAC - hours)}}{\text{2000 DAC-hours}} * \text{Dose Limit} \tag{5.4}$$

$$E_{50} \text{ or } H_{T,50} = \frac{\text{Intake}}{\text{ALI}} * \text{Dose Limit} \tag{5.5}$$

If respiratory protection is worn by workers, the appropriate respirator protection factor may be applied to the above calculations (i.e., dividing the calculated result by the protection factor.)

General air sampling programs should be augmented by breathing zone sampling when air concentrations to which individuals are exposed might be highly variable. Breathing zone sampling may include both fixed-location and personal (lapel) air samplers. Personal air samples are more likely to be representative of actual exposure conditions than are samples collected at fixed locations, and can be particularly useful for assessing potential intakes involving short-term exposure to well-monitored air concentrations.

5.8 DOSE ASSESSMENT

Dose assessment involves collecting and analyzing information concerning a potential intake and developing a conclusion regarding the magnitude of intake and its associated committed doses. Dose assessments are conducted by investigating the nature of a potential intake and by analyzing bioassay measurement results or other pertinent data.

Biokinetic models are used in conjunction with bioassay data to evaluate the intake, uptake, and retention of plutonium in the organs and tissues of the body. Intake estimates can then be used to calculate committed effective and organ equivalent doses. It is essential that good professional judgment be used in evaluating potential intakes and assessing internal doses. Carbaugh (1994) has identified a number of considerations for dose assessments.

Computer codes are commonly used for assessment of intakes, dose calculation, and bioassay or body content projections. La Bone (1994a) has provided an overview of what should be considered in selecting a computer code, as well as descriptions of a number of internal dosimetry codes available in 1994. Internal dosimetry code users should understand how the code works and be aware of its limitations. Computer codes merely provide the logical result of the input they are given. Use of a particular computer code does not necessarily mean a dose estimate is correct.

As used in this section, the definition of “intake” is the total quantity of radioactive material taken into the body. Not all material taken into the body is retained. For example, in an inhalation intake, the ICRP Publication 66 respiratory tract model predicts that, for 5-µm AMAD particles, 82% of the intake will be deposited in the respiratory tract; the other 18% is immediately exhaled (ICRP, 1994a). For a wound intake, material may be initially deposited at the wound site. Once the material has been deposited, it can be taken up into systemic circulation either as an instantaneous process (e.g., direct intravenous injection of a dissolved compound) or gradually (e.g., slow absorption from a wound site or the pulmonary region of the lung). Both the instantaneous and slow absorption processes are often referred to as uptake to the systemic transfer compartment (i.e., blood). Once material has been absorbed by the blood, it can be translocated to the various systemic organs and tissues.

An understanding of this terminology is important to review of historical cases. In the past sites reported internal doses as an uptake (or projected uptake) expressed as a percentage of a maximum permissible body burden. The standard tabulated values for maximum permissible body burdens were those in ICRP Publication 2 (ICRP, 1959). Many archived historical records may have used this approach. DOE Order 5480.11 (superseded), required calculation of dose equivalent. Now, 10 CFR 835 (DOE, 2007a), has codified the calculation of intakes and committed doses.

5.8.1 Methods of Estimating Intake

There are several published methods for estimating intake from bioassay data (Skrable et al., 1994a; Strenge et al., 1992; ICRP, 1988b; King, 1987; Johnson and Carver, 1981). These methods each employ an idealized mathematical model of the human body showing how materials are retained in and excreted from the body over time following the intake. IRFs are used to predict the fraction of an intake that will be present in any compartment of the body, including excreta, at any time post-intake. Intake retention functions incorporate an uptake retention model that relates uptake to bioassay data and a feed model that relates intake to uptake and bioassay data. ICRP Publication 54 (ICRP, 1988a) and Lessard et al. (1987) have published compilations of IRFs. More recently, in Potter published compilations of IRFs, consistent with the 2007 amendment to 10 CFR 835 (Potter, 2002). Selected IRFs calculated consistent with the 2007 amendment to 10 CFR 835 for the urine and fecal excretion and remaining in the whole body are shown in Tables 5.6 for absorption type M and S forms of ^{239}Pu. These functions would be similar in value to those for other long-lived forms isotopes of Pu.

$$Q_t = \text{Intake} * \text{IRF}(Q_t) \tag{5.6}$$

In its simplest form, a compartment content at any time post-intake (Q_t) can be expressed as the product of intake multiplied by the intake retention function value for compartment Q at time t post-intake, or:

Results predicted by the model can then be compared with the observed bioassay data. Such results are often referred to as expectation values.

Simple algebraic manipulation of the model allows calculation of intake from the compartment content at time t, as shown below:

$$\text{Intake} = \frac{Q_t}{\text{IRF}(Q_t)} \tag{5.7}$$

When multiple data points are available for a compartment, the intake can be estimated using an unweighted or weighted least-squares fitting procedure, as described by Skrable et al. (1994b) and Strenge et al. (1992) or as can be found in most statistics textbooks. As an alternative, data can be fit by eye to a graphical plot; however, the apparent fit can be misleading if data has been logarithmically transformed.

Intake can also be estimated from air sample data, as described in Section 5.7.4. This method is appropriate if bioassay data are not available or insufficiently sensitive. Intake estimates based on air samples and bioassay data are also appropriate as a check on each other. Valid bioassay data showing detectable results should be given preference over intake estimates based on air sample results.

Table 5.6. Intake Retention Fractions[(a)] for ^{239}Pu

	Type M Inhalation			Type S Inhalation		
Days-Post Intake	Urine	Feces	Whole Body	Urine	Feces	Whole Body
1	2.46E-04	1.10E-01	4.95E-01	2.50E-06	1.16E-01	4.90E-01
7	2.40E-05	2.29E-03	8.34E-02	3.08E-07	2.42E-03	6.25E-02
30	9.51E-06	2.81E-04	7.29E-02	1.72E-07	3.51E-04	5.06E-02
60	8.11E-06	1.31E-04	6.68E-02	1.65E-07	1.86E-04	4.29E-02
90	7.12E-06	6.65E-05	5.98E-02	1.61E-07	1.07E-04	3.87E-02
200	5.12E-06	4.67E-06	5.74E-02	1.61E-07	3.32E-05	3.25E-02
400	3.71E-06	3.71E-06	5.74E-02	1.70E-07	2.13E-05	2.74E-02
1000	2.44E-06	1.04E-06	5.44E-02	1.77E-07	1.12E-05	1.79E-02
10000	4.16E-07	2.96E-07	4.11E-02	8.25E-08	9.53E-08	5.12E-03
20000	4.83E-07	2.11E-07	3.29E-02	5.83E-08	3.20E-08	3.13E-03

(a) Incremental (i.e., sample collected in a 24-hour period ending at the time indicated) values for excreta obtained from "Intake Retention Functions Developed from Models Used in the Determination of Dose Coefficients Developed for ICRP Publication 68 – Particulate Inhalation" (Potter, 2002). See Section 5.8.1.

5.8.2 Alternate Methods of Intake Assessment

Historically, intake as described in the foregoing section was not always calculated when assessing plutonium exposures. Estimates of uptake using methods similar to Langham (1956), Healy (1957), or Lawrence (1987) focused on assessing the magnitude of radioactivity retained in the body, rather than intake (which includes material not retained and of no dosimetric significance). These methods were (and are) dosimetrically sound in so far as estimates of deposition and uptake are concerned, but do not meet the current regulatory requirement of 10 CFR 835 (DOE, 2007a) to calculate intake.

5.8.3 Estimating Dose from Intakes of Plutonium

The committed equivalent dose ($H_{T,50}$) and the CED (E_{50}) resulting from an intake of plutonium may be calculated by multiplying the estimated intake (I) by either the dose conversion factor for effective dose (DCF_{eff}) or the dose conversion factor for equivalent dose (DCF_{equ}) :

$$E_{50} = I * DCF_{eff} \qquad H_{T,50} = I * DCF_{equ} \qquad (5.8)$$

Dose conversion factors consistent with the 2007 amendment to 10 CFR 835 can be obtained from the ICRP Publication 68 Database (ICRP, 1994b) or calculated directly using computer programs.

Values for simplified dose conversion factors can be obtained by dividing a dose limit by the corresponding value for the ALI. A caution shall be observed with this approach: not all tabulated valued of ALIs are the same. The ALIs and DACs are commonly rounded in most tabulations to one significant figure (e.g., as in Appendix A of 10 CFR 835). Substantial variation can occur as a result of unit conversion. For example, Appendix A of 10 CFR 835 lists the DACs for ^{246}Pu absorption types M and S as both 8E-08 uCi/ml but lists the DACs in the SI units as 3E+03 Bq/m^3 for absorption type M and 2E+03 Bq/m^3 for absorption type S. Such rounding errors can introduce significant discrepancies in dosimetry calculations. This method also raises a question about which ALI or DAC should be used if compliance monitoring is being based on comparison with secondary limits, such as the ALI or DAC rather than the primary dose limits.

Where individual-specific data are available, the models should be adjusted. However, the general lack of capability to monitor organ-specific retention for plutonium (i.e., content and clearance half-times) makes the use of default models most practical.

Ideally, one should obtain as much bioassay information as possible to determine the intake and track the retention of plutonium in the body to reduce the uncertainty associated with the daily variation in the measurements. A regression analysis should be used to fit the measurement values for estimating the initial intake and clearance half-times.

5.8.4 Evaluating ^{241}Am Ingrowth in an In Vivo Count

Ingrowth of ^{241}Am from ^{241}Pu can significantly impact bioassay monitoring projections. Unless accounted for, it can lead to suspicion of new intakes, or underestimation of clearance rates. The amount of ^{241}Pu present in a plutonium mixture depends on the irradiation history and time since irradiation. Freshly processed mixtures containing 6% by weight of ^{240}Pu may contain about 0.5% by weight of ^{241}Pu and a 12% ^{240}Pu mixture may contain 3% ^{241}Pu. Commercial spent fuel can be much higher. The ingrowth of ^{241}Am occurs following a plutonium intake over a period of years. Less transportable (Material Type S) forms of plutonium may have ^{241}Am ingrowth which gradually becomes detectable. An extreme case of this was demonstrated in a well-documented Hanford plutonium-oxide exposure which exhibited a factor-of-2 increase in ^{241}Am lung content in the 3000 days following intake (Carbaugh et al., 1991). Such an increase could not be explained using the standard 500-day class Y lung clearance half-time; finally, a 17-year biological clearance half-time was estimated. The subsequent CED equivalent was estimated to be a factor of 3 higher than if the standard 500-day half-time had been used. Similar difficulties have occurred with initial detection of ^{241}Am by routine in vivo chest counting or in long-term monitoring of residual wound content.

While many available internal dosimetry computer codes will calculate the projected ^{241}Am lung content following an intake (accounting for ingrowth in the process), none of the current codes will do curve-fitting from long-term data and at the same time adjust the data for ingrowth. Therefore, the following simplistic method was developed to assess that data.

An estimate of the ^{241}Am ingrowth can be made by assuming that, at the time of intake (t = 0), all the material that will compose the long-term component is deposited in a single compartment and that the rate of transfer of material from the compartment at any subsequent time t is proportional to the quantity of material remaining in the compartment (i.e., simple exponential transport kinetics). The following equation will then describe the buildup of ^{241}Am in that compartment following an initial deposition of ^{241}Pu and ^{241}Am and a given or assumed effective clearance rate:

$$A_{t,\,Am} = \lambda_{r,\,Am} \frac{A_{O\,Pu}}{k_{e,\,Am} - k_{e,\,Pu}} \left(e^{-k_{e,\,Pu}t} - e^{-k_{e,\,Am}t}\right) + A_{O,\,Am}\, e^{-k_{e,\,Am}t} \qquad (5.9)$$

where $A_{t,Am}$ = activity of ^{241}Am at time t

$\lambda_{r,Am}$ = radiological decay constant for ^{241}Am

$A_{0,Pu}$ = activity of ^{241}Pu at time 0

$k_{e,Am}$ = effective clearance rate of ^{241}Am

$k_{e,Pu}$ = effective clearance rate of ^{241}Pu

$A_{0,Am}$ = activity of ^{241}Am at time 0

t = elapsed time

The effective clearance rate (k_e) of any nuclide is the sum of the radiological decay constant (λ_r) and the biological clearance rate (λ_{bio}). By assuming that the biological clearance rate is constant for both parent and progeny nuclides, the equation reduces to three unknowns: the initial amount of parent, the initial amount of progeny, and the biological clearance rate. These unknowns can be dealt with by assuming a standard isotopic composition at the time of intake and then solving the equation for a biological clearance rate using an iterative process until the calculated result matches the observed result at a given time t. A computer or calculator algorithm can eliminate the need for lengthy hand calculations.

Once an optimum combination of isotopic compositions and biological clearance rate is found, internal dosimetry codes or hand calculations can be used to estimate organ and effective doses. As a check on the results, standard computer codes can be used in a bioassay projection mode to project the ^{241}Am content based on the estimated intake and biological clearance rate.

5.9 INDICATOR AND ACTION LEVELS

Indicator and action levels are essential to operation of a routine internal dosimetry program. Because a wide range of levels can be defined by various facilities and organizations, this document does not attempt to prescribe particular level titles. As used in this document, indicator and action levels are simply workplace or bioassay measurements, or associated calculated doses, at which specific actions occur.

Indicator levels based on workplace indicators for reacting to a potential intake are suggested in Table 5.7. The intent of these indicator levels is to provide guidance for field response to any potential intake of radioactive material with a potential for a dose commitment that is >100-mrem CED. It is suggested that when these levels are reached, appropriate management members of the health physics and operations organizations be informed. See Section 5.4.3 for guidance on special bioassay. Table 5.8 suggests notification levels to the occupational medicine physician for possible early medical intervention in an internal contamination event. These tables, derived from Carbaugh et al. (1994), are based on general considerations and significant experience with past intakes of radioactive material and, because they are based on field measurements, do not correspond with any exact dose commitment to the worker.

The decision to administer treatment and the treatment protocol are the joint responsibilities of the physician in charge in full coordination with the patient who has been informed of the risks and benefits of any treatment being considered. The basic principle is that the proposed intervention should do more good than harm (Gerber and Thomas, 1992).

Guidelines for the medical intervention of a radionuclide intake can be found in several publications. NCRP Report No. 65 (NCRP, 1980) and the joint publication of the Commission on European Communities (CEC) and the DOE Guidebook for the Treatment of Accidental Internal Radionuclide Contamination of Workers (Gerber and Thomas, 1992) both contain detailed guidance in intervention and medical procedures useful in mitigating radiation overexposures. The ICRP recommends in Publication 60 (ICRP, 1991a) a limit of 2-rem/y (20-mSv/y) on effective dose.

Thus, the ALIs found in ICRP Publication 61 (ICRP, 1991b) and used in the CEC/DOE Guidebook noted above are those which would provide a CED of 2-rem/y instead of current U.S. regulations of 5-rem/y.

Guidance in the CEC/DOE Guidebook can be summarized as follows:

-- When the estimated intake is below one ALI, treatment should not be considered.

-- When the estimated intake is between 1 and 10 times the ALI, treatment should be considered.

Under these situations, short-term administration will usually be appropriate, except for intake of materials poorly transported from the lung (Material Type S).

Table 5.7. Suggested Plutonium or ^{241}Am Indicator Levels for Internal Dosimetry Evaluation

Indicator	Notification Level
Nasal or mouth smears	Detectable Activity
Facial Contamination (direct measurement)	200 dpm
Skin Breaks or Blood Smears	Any skin break while handling material other than sealed sources
Head, neck contamination	2,000 dpm
Contamination in a respirator	Detectable activity inside respirator after use
Hands forearms, clothing contamination (a)	10,000 dpm
Airborne Radioactivity	Acute intake equivalent to 40 DAC-hours after accounting for respiratory protection factor

(a) Clothing contamination levels apply to exposure without respiratory protection, such as on inner coveralls or personal clothing.

Table 5.8. Suggested Plutonium or ^{241}Am Contamination Levels for Notification of Occupational Medicine Physician

Indicator	Medical Notification Level, dpm
Nasal or mouth smears	1,000
Facial Contamination	25,000
Skin breaks or wounds	100

-- When the estimated intake exceeds 10 times the ALI, then extended or protracted treatment should be implemented, except for materials poorly transported from the lung.

-- For poorly transported material in the lung, lung lavage (i.e. internal lung washing under anesthesia) is the only recommended treatment, and it is only a consideration for intakes exceeding 100 times the ALI.

Because the dose associated with the ALI in the CEC/DOE Guidebook is 2-rem CED and because the upper administrative level recommended by the standard, Radiological Control, is 2 rem, intervention levels of 2 rem and 20 rem might be used for guidance in the manner presented in the CEC/DOE Guidebook:

-- When the CED for an estimated intake is below 2 rem, treatment is not generally recommended.

-- When the CED for an estimated intake is between 2 rem and 20 rem, treatment should be considered. Under these situations, short-term administration will usually be appropriate.

-- When the CED for an estimated intake exceeds 20 rem, then extended or protracted treatment is strongly recommended, except for poorly transported material in the lung.

Decorporation therapy should be administered immediately following any suspected intake or accidental internal contamination in excess of established action levels. The extent and magnitude of an internal plutonium contamination usually cannot be determined quickly; however, the usefulness of therapy will diminish if plutonium is allowed to translocate to bone where DTPA is ineffective. La Bone (1994b) has provided a recent approach to evaluating urine data enhanced by chelation (DTPA) therapy.

An initial prophylactic chelation therapy may be appropriate because bioassay measurements (particularly urinalysis) cannot usually be completed within the response time required for effective chelation therapy. Urinalysis becomes very helpful following administration of chelation therapy because there is a direct correlation between DTPA, urinary excretion, and dose averted because of plutonium excreted. Bihl (1994) has shown that about 2 mrem of CED is averted for every dpm of ^{239}Pu excreted. The averted dose would be less for assessment of dose in the newer quantity, per the 2007 amendment to 10 CFR 835, of CED. This is because the dose per intake is less under the newer models. For Material Type S compared to Class Y it is lower by approximately a factor of 10 for ^{239}Pu.

This provides useful information for measuring the effectiveness of DTPA therapy and determining if it is worthwhile to initiate or to continue therapy. For example, using the pre-2007 amendment to 10 CFR 835 models, if DTPA is administered when untreated excretion is 2 dpm/d, excretion should increase to 20 to 100 dpm for a dose savings of 40- to 200-mrem/d CED. However, the dose aversion would only be 4 to 20-mrem/d CED under the models required by the 2007 amendment to 10 CFR 835. Additionally, it is probable that the efficacy of treatment will decrease with continued administration as plutonium is removed from the liver and the rate of transfer into the systemic compartment decreases.

5.10 RESPONSE TO SUSPECTED INTAKES

Experience has shown that most intakes of plutonium are accidental. Plutonium facilities and operating procedures are designed to prevent intakes. Nonetheless, it is important for management to prepare for the possibility that workers might receive an intake of plutonium--even though the probability of an incident may be very small. Prompt and appropriate action following an accidental intake of plutonium will allow for therapeutic measures to be taken to minimize the internal contamination and lessen the potential for harmful effects. The health physicist and medical staff should work closely to ensure that the proper course of action is followed.

All employees suspected of having received an intake of plutonium should be referred for special bioassay measurements. Because a fraction of an intake by inhalation may be retained in the nasal passages for a few hours after exposure to airborne radioactive materials, any level of contamination on a nasal swab indicates an intake that should be followed up by a special bioassay measurement program. However, lack of detection on nasal smears cannot be taken as evidence that an intake did not occur either because the nasal passages can be expected to clear very rapidly or, alternatively, because the worker could be a mouth-breather. Special bioassay should also be initiated if plutonium contamination is found on the worker in the vicinity of nose or mouth.

For acute intakes, direct bioassay measurements should be taken before, during, and after the period of rapid clearance of activity. Urine and fecal samples collected after known or suspected inhalation incidents should also be used to estimate the magnitude of the intake. Initial assessments of intakes from contaminated wounds are based primarily on wound count and urinalysis data.

Guidance on evaluation of intakes of plutonium is found in DOE-STD-1121-2008, Internal Dosimetry (DOE, 2008d). DOE-STD-1121-2008 recognizes the difficulty in making final assessments of plutonium intakes and cautions that it "is not appropriate to place heavy reliance on the actual magnitude of the dose in the first few days following a suspected intake." Notwithstanding this difficulty, for various reasons there is a need to be able to make a timely evaluation of the potential magnitude of plutonium intakes. DOE Order 225.1B, Accident Investigations (DOE, 2011d), has a criterion for accident investigation based on a "confirmed monitoring result (workplace or individual) indicating an intake (via inhalation, ingestion, wound or absorption) of radioactive material by a general employee equivalent to 2 or more times the annual limit on intake." The Order also states that "Confirmation must be made within 3 working days following identification of monitoring results (workplace or individual monitoring) indicating an exposure exceeding one or more of the criteria in this section."

If a significant intake is indicated, the worker should not return to further potential exposure to plutonium until the intake has been thoroughly assessed and a predictable bioassay pattern established. This is particularly important because a new intake of a very low level may confound the interpretation of bioassay measurements for previous intakes of plutonium.

The health physicist shall make important decisions for prompt action at the site of an accidental or suspected intake of plutonium or other radioactive materials. Often, these decisions shall be based on limited data. Information that may be available for initially estimating the amount and type of intake may include the following:

-- levels of measured contamination in the work area

-- skin contamination levels, affected areas, and whether the skin is damaged or punctured

-- wound contamination levels

-- chemical form of the material involved

-- results of air monitoring

-- nasal smear activity levels

-- sputum and/or mouth contamination.

The special bioassay monitoring program is initiated following a known or suspected intake. This information is needed for dose assessment and future exposure management. The intake is confirmed if follow-up bioassay measurements indicate positive measurement results. Additional bioassay measurements may be needed to quantify the intake and provide data for determining the effective dose. The frequency of bioassay monitoring will depend on the specific case to be evaluated. Selection of the appropriate sampling frequency is based on the previously discussed performance capabilities for workplace monitoring program, consultations with internal dosimetry specialists, and the cooperation of the affected employee.

5.10.1 Planning

The management at the plutonium facility should be prepared to follow an emergency action plan for response to a plutonium intake. If a worker accidentally inhales or ingests plutonium or is injured by a plutonium-contaminated object, the action plan should be initiated immediately. A rapid response is important because any delay in implementing appropriate action could lessen the effectiveness of decorporation therapy and increase the probability for internalized plutonium to deposit on bone surfaces.

5.10.2 Medical Response Plan

The health physicist and medical staff shall establish an emergency action plan for the appropriate management of an accidental intake of plutonium. The elements of the plan should include the following:

-- Decision levels for determining when monitoring data or accident events require emergency medical response

-- responsibilities of the affected worker, health physicist, medical staff, and management or supervisory personnel

-- instructions for immediate medical care, decontamination, monitoring, and longer-term follow-up response

-- provisions for periodically reviewing, updating, and rehearsing the emergency action plan.

The sequence and priority of the emergency action plan may vary with the magnitude and type of accidental conditions and their severity. An initial early assessment of the incident should focus, first, on treatment of life-threatening physical injuries and, second, on the radioactive contamination involved. Minor injuries should be treated after decontamination.

A rapid estimate of the amount of internal contamination by plutonium or other alpha-emitters may not be possible. If a significant intake (meaning one that exceeds 10 times the ALI) is suspected, medical staff should proceed with decorporation therapy after first treating major injuries.

5.10.3 Responsibilities for Management of Internal Contamination

Responsibilities should be assigned for action in response to an accidental internal plutonium contamination. The affected worker has the responsibility to inform the health physicist, RCT, or his immediate supervisor as soon as an intake is suspected. (More broadly, all radiological workers have the responsibility to report conditions that could lead to an intake to their immediate supervisor and/or the health physics organization.) The health physicist or RCT should make an initial survey of the extent of the contamination and immediately contact his supervisor and, when action levels are exceeded, contact a member of the medical staff. He should continue to provide monitoring and radiation safety support to the medical staff and supervisors during the management of the contamination incident. Care should be taken to limit the spread of radioactive contamination.

The health physicist should immediately begin to gather data on the time and extent of the incident. Contamination survey results should be recorded. Radionuclide identity, chemical form, and solubility classification should be determined. Nasal smears should be obtained immediately if an intake by inhalation is suspected. When action levels are exceeded, all urine and feces should be collected and labeled for analysis. Decontamination should proceed with the assistance of the medical staff. Contaminated clothing and other objects should be saved for later analysis.

5.10.4 Immediate Medical Care

The medical staff should provide immediate emergency medical care for serious injuries to preserve the life and well-being of the affected worker. Minor injuries may await medical treatment until after an initial radiation survey is completed and the spread of contamination is controlled. However, the individual should be removed from the contaminated area as soon as possible. Chemical contamination and acids should be washed immediately from the skin to prevent serious burns and reactions.

Chelation

Chelation therapy, or chelation, is the process of removing unwanted metals from the body by administering an agent that binds to the metal and promotes its excretion. It is important to remove plutonium from the body because it is retained in the bones and liver for many years. Plutonium remaining in the body continues to irradiate nearby tissues. This results in increased risk of cancer. For over 60 years, chelation therapy has been practiced successfully and safely in treating lead and other heavy metal poisoning.

Chelating agents can be administered orally, intravenously, or as a mist, depending on the agent and the type of poisoning. Several chelating agents are available; each has different affinities for different metals. DTPA (diethylenetriaminepentaaceticacid) has been proven effective for the treatment of people accidentally contaminated internally with the transuranic nuclides plutonium, americium, and curium. Recently, based on additional clinical data and peer-reviewed articles, the Food and Drug Administration (FDA) has approved DTPA as a safe and effective compound to enhance elimination/excretion of radioactive materials from the body. There are two primary DTPA compounds: Ca-DTPA and Zn-DTPA. Ca-DTPA is more effective than Zn-DTPA in the first 24 hours after contamination. To avoid long-term depletion of essential metals, Ca-DTPA is administered initially, followed by Zn-DTPA if multiple doses are required.

The number of treatments is based on the results of the bioassay analyses. Most situations involve single treatments; however, a 2010 wound incident at a DOE facility involved 71 treatments. Possible side effects of such an extended chelation therapy regiment could include depletion of essential elements, which can be treated by administering supplemental minerals.

In addition, according to the Centers for Disease Control and Prevention (CDC) Web site, people who are given repeat doses of Ca-DTPA within a short period of time may have nausea, vomiting, diarrhea, chills, fever, itching, and muscle cramps. Other side effects may include headache, lightheadedness, chest pain, and a metallic taste in the mouth. Chelation therapy administered by nebulized inhalation may cause breathing difficulties in some individuals.

According to the FDA Web site at: **http://www.fda.gov/Drugs/EmergencyPreparedness/BioterrorismandDrugPreparedness/ucm130314.htm:**

- If Ca-DTPA is not available, or treatment cannot be started within the first 24 hours after contamination, treatment should begin with Zn-DTPA.
- If Zn-DTPA is not available, Ca-DTPA can be given for continued treatment, along with vitamin or mineral supplements that contain zinc.
- Ca-DTPA and Zn-DTPA can be administered by nebulizer or directly into the blood stream (i.e., intravenously). If the route of internal contamination is through inhalation alone, then nebulized chelation therapy will suffice. If the routes of contamination are multiple (e.g., inhalation and through wounds), then intravenous chelation therapy is preferred.
- The duration of treatment is dictated by the level of internal contamination and the individual’s response to therapy. Levels of internal contamination should be

ascertained weekly during chelation therapy to determine when to terminate treatment.

- Zn-DTPA is the preferred treatment for the pregnant woman with internal contamination.
- FDA recommends nebulized DTPA for patients whose internal contamination is only by inhalation.
- The safety and effectiveness of the intramuscular route has not been established for
Ca-DTPA or Zn-DTPA.
- The duration of Ca-DTPA and Zn-DTPA therapy depends on the amount of internal radioactive contamination and the individual's response to therapy.
- Ca-DTPA should be used with caution in patients suffering from a severe form of a disease called hemochromatosis.

Additional information is on the CDC Web site at:
http://www.bt.cdc.gov/radiation/dtpa.asp.

Some of this information includes:

- Radioactive materials chelated to DTPA are excreted from the body in the urine; therefore, DTPA shall be used carefully in people whose kidneys do not function properly.
- Breathing treatments using DTPA may not be safe for some people with asthma. If a person with asthma requires treatment with DTPA, the drug should be injected.
- DTPA should not be used to treat people who are internally contaminated with the radioactive materials uranium or neptunium.

The big advantage of chelation for radioactive metals, such as plutonium, is the radiation dose reduction for the patient. Substantial dose reductions can be achieved if DTPA is administered within a few hours (recommended within 1 hour) of the intake of plutonium. Dose reductions from 10 percent to 90 percent have been achieved for contaminated wound or burn cases and up to 30 percent for inhalation cases.

The decision to administer chelation therapy is made by the worker in consultation with a board-certified occupational medicine physician. In communicating information to the individual concerning the risks and benefits of chelation refer to the Health Physics Society policy paper which discusses providing individual risk estimates. The position paper states, in part: "*the Health Physics Society recommends against quantitative estimation of health risks below an individual dose of 5 rem in one year or a lifetime dose of 10 rem above that received from natural sources*". More information on this position paper is found at:
http://hps.org/documents/risk_ps010-2.pdf

Chelation is generally recommended when the estimated dose exceeds 2 rem CED. If a quick dose estimate cannot be made, indicators such as airborne radioactivity exposure, nasal/mouth smears, facial contamination, skin breaks, or bioassay measurements are used. Chest or whole-body counts and wound counts are used as well.

References and Web sites for this subject, in addition to the ones listed previously, include NCRP Report Number 161, Management of Persons Contaminated with Radionuclides (NCRP, 2008), and the Radiation Emergency Assistance Center/Training Site Web site at: http://orise.orau.gov/reacts/guide/internal.htm.

5.10.5 Contaminated Wounds

Medical treatment for contaminated wounds may include flushing with saline and decorporating solutions, debridement, and surgical excision of the wound. These measures are all the responsibility of trained medical staff operating under the direction of a physician. Health physics personnel can provide valuable assistance by prompt assessment of materials removed from the wound and identification of magnitude of residual activity as decontamination proceeds. Decontamination should continue until all radioactivity has been removed or until risk of permanent physical impairment is reached.

NCRP Report Number 156, Development of a Biokinetic Model for Radionuclide-Contaminated Wounds and Procedures for Their Assessment, Dosimetry and Treatment, provides additional information on evaluation of contaminated wounds (NCRP, 2006).

The Radiation Emergency Assistance Center/Training Site Web site provides dose coefficients for 38 radionuclides based on NCRP wound model and ICRP biokinetic models:

Dose Coefficients for Intakes of Radionuclides via Contaminated Wounds

6.0 EXTERNAL DOSE CONTROL

The purpose of an external dose control program is to protect the individual radiation worker by minimizing dose to levels as low as reasonably achievable (ALARA) and preventing exposures above prescribed limits. This also implies minimizing the collective dose by summing all the individual TEDs in a specified population. This section discusses methods to minimize exposures by characterizing the radiations emitted by plutonium and effective methods to shield or otherwise reduce exposures.

The Department of Energy provides a detailed explanation of the recommendations for external dosimetry in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). Specific applicable documents for external dosimetry are listed in the reference list of that Implementation Guide. Because the requirements and recommendations are explicitly given in these documents, they will not be discussed in any great detail in this section. Rather, the emphasis will be given to items that are unique to plutonium facilities and the radiological aspects for safe handling of plutonium.

Measuring the external radiation exposure and the resultant dose for personnel handling plutonium is a difficult task because of the many radiations involved. Examples of the radioactive decay schemes and radiations emitted were presented in Section 2.0 for the various plutonium isotopes and radioactive progeny. Plutonium has a wide distribution of gamma energies; literally hundreds of different photon energies are present. Fortunately, plutonium emits few high-energy photons, so photon dose rates are low. But plutonium also emits highly penetrating neutrons from spontaneous fission and alpha-neutron reactions from compounds and alloys.

In the past, most of the dose in plutonium facilities was the result of plutonium production and fabrication operations. Most of these operations involved physical contact with freshly separated plutonium in glove boxes during fabrication and assembly operations. With the reduction in weapons production, emphasis has shifted to dismantlement and storage operations and to D&D of plutonium facilities. Much of the material in these facilities is low-exposure plutonium containing 6% ^{240}Pu that is at least 20-years-old, so a significant fraction of the ^{241}Pu has decayed into ^{241}Am. The radioactive progeny have increased gamma dose rates, making dismantlement of plutonium facilities more difficult. Although many of the examples in this section involve higher-exposure plutonium, it is expected that most dosage in plutonium facilities will originate from clean-up and storage of weapons-grade plutonium.

6.1 DOSE LIMITS

Limits of interest used for control of external radiations are specified at various depths by 10 CFR 835 (DOE, 2007a) as well as the ICRP and the NCRP). The limits are given in Table 6.1 for the appropriate depths in tissue for the whole body, lens of the eye, skin and extremities.

Table 6.1. Effective Depth of Tissue for Various Organs

	Depth of Tissue mg/cm^2 .
Equivalent dose to the whole body	1000
Equivalent dose to the lens of eye	300
Equivalent dose to the extremity and skin	7

6.1.1 Limiting Quantities

Recently, DOE has made significant changes in the methodology used for radiation protection. Previously, DOE used the concept of *dose equivalent*. For whole body irradiations, dose equivalent was the product of absorbed dose multiplied by the quality factor, which was evaluated by Monte Carlo calculations in a cylindrical phantom of 30-cm diameter and 60-cm height. For monoenergetic neutrons or photons normally incident on the phantom model, the dose equivalent was the highest value calculated anywhere in the phantom. Dose equivalent was non-additive because the maximum values occur at different depths in the phantom for different energies. A detailed explanation of the calculations can be found in an article by Auxier et al. (1968).

ICRP Publication 60 used revised terms for *stochastic* and *nonstochastic* for radiation effects (i.e., *stochastic* and *deterministic*) and set limits for both types of effect. Stochastic effects are defined as those for which the probability of the effect occurring (as opposed to the degree or severity of effect) is a function of radiation dose. Deterministic effects were defined as those for which the severity of the effect is a function of the dose; a threshold may exist. Limits were established such that the risk of stochastic effects occurring was equivalent to about the same risks faced by workers in “safe” industries who were not occupationally exposed to radiation in the workplace. Limits were also established for deterministic effects that prevented these effects from occurring even if the exposure occurred at the annual limit over the lifetime of the worker.

The ICRP specified in Publication 60 that radiation exposure be limited by the *effective dose*, E, which can be expressed by the relation:

$$E = \sum w_T D_{T,R} w_R \qquad (6.10)$$

where

w_T = tissue weighing factor for the relevant organ or tissue T
D_{TR} = average absorbed dose in the tissue or organ of interest
w_R = radiation weighting factor for the type of radiation R

The weighing factors are given in Table 6.2, which is taken from 10 CFR 835 (DOE, 2007a). Effective dose has the benefit that it is additive, and internal and external radiations can be added numerically to drive an overall estimate of risk.

Table 6.2. Tissue Weighing Factors

Organs or tissues, T	Tissue weighting factor, w_T
Gonads	0.20
Red bone marrow	0.12
Colon	0.12
Lungs	0.12
Stomach	0.12
Bladder	0.05
Breast	0.05
Liver	0.05
Esophagus	0.05
Thyroid	0.05
Skin	0.01
Bone surfaces	0.01
Remainder[1]	0.05
Whole body[2]	1.00

[1] "Remainder" means the following additional tissues and organs and their masses, in grams, following parenthetically: adrenals (14), brain (1400), extrathoracic airways (15), small intestine (640), kidneys (310), muscle (28,000), pancreas (100), spleen (180), thymus (20), and uterus (80). The equivalent dose to the remainder tissues ($H_{remainder}$), is normally calculated as the mass-weighted mean dose to the preceding ten organs and tissues. In those cases in which the most highly irradiated remainder tissue or organ receives the highest equivalent dose of all the organs, a weighting factor of 0.025 (half of remainder) is applied to that tissue or organ and 0.025 (half of remainder) to

the mass-weighted equivalent dose in the rest of the remainder tissues and organs to give the remainder equivalent dose.

[2] For the case of uniform external irradiation of the whole body, a tissue weighting factor (w_T) equal to 1 may be used in determination of the effective dose.

The methodology of ICRP Publication 60 (ICRP, 1991a) has been incorporated into 10 CFR 835 (DOE, 2007a). The annual radiation dose limits for DOE and its contractors are presented in Table 6.3.

However, DOE contractors usually establish lower annual administrative control levels, typically 500 mrem/year.

In practice, it is very difficult to measure the effective doses specified in Table 6.3 because it is necessary to know not only the type of radiation but also its energy and direction. If the flux, energy, and direction of incidence are known, it is possible to calculate effective dose using fluence to effective dose conversion coefficients, which present the effective dose as a function of energy for various irradiation geometries. Conversion coefficients for photons in various irradiation geometries, including planar sources, can be found in a report by the Zankl et al. (1994). This will provide more accurate values of effective dose as opposed to numerically setting the value of effective dose equal to equivalent dose.

Table 6.3. Radiation Dose Limits for DOE and DOE Contractors

Type of Radiation Exposure	Annual Limit
Occupational Exposures	
Stochastic Effects	5-rem total effective dose from external sources and the committed effective dose from intakes received during the year
Deterministic Effects	
Lens of eye	15-rem equivalent dose
Extremity	50-rem equivalent dose
Skin	50-rem equivalent dose
Individual organ tissue	50-rem equivalent dose
Embryo/fetus of a Declared Pregnant Worker	
Gestation period	0.5-rem equivalent dose
Planned Special Exposure	
Event plus Annual Occupational exposure	5-rem total effective dose (TED)
Minors	0.1-rem TED

6.1.2 Operational Quantities

Because of the difficulties in determining effective dose from direct measurements, the concept of *operational quantities* has been introduced to be more closely related to measurable quantities. Operational quantities include *ambient dose equivalent* used for area monitoring and *personal dose equivalent* used for personnel dosimetry. Operational quantities should be a conservative estimator of effective dose, i.e., the values of the operational quantities should be equal or greater than the effective dose specified for the limiting quantities.

The ambient dose equivalent, H^* (d), is the dose equivalent at a depth, d, in a 30-cm-diameter sphere of tissue, where a) the radiation field has the same fluence and energy distribution as the point of reference for the measurement and b) the fluence is unidirectional (i.e., the sphere can be viewed as being in an aligned radiation field). Most survey instruments are designed to measure ambient dose equivalent, and international standards are based on the ambient dose equivalent concept. The depth of interest is 1 cm of soft tissue, as specified in 10 CFR 835.2.

The personal dose equivalent, H_p(d), is the dose equivalent in soft tissue at the appropriate depth, d, below a specified point on the body. Provided that personal dose equivalent is a conservative estimator of effective dose, personnel dosimeters should be calibrated in terms of personal dose equivalent. Otherwise a correction factor should be applied.

In reality, most instruments and personnel dosimeters used at DOE facilities are still calibrated in terms of dose equivalent. For example, consider the case in which personnel neutron dosimeters are calibrated on acrylic plastic phantoms at a specified distance from a calibrated neutron source. For Department of Energy Laboratory Accreditation Program for personnel dosimetry (DOELAP) testing, the dose equivalent is assessed in accordance with U. S. Department of Energy Laboratory Accreditation for External Dosimetry, DOE STD-1095- 2011 (DOE, 2011e) which incorporates ANSI/HPS N13.11, Personnel Dosimetry Performance-Criteria for Testing, ANSI/HPS N13.11 (ANSI, 2009).

In most instances, the present methods based on dose equivalent over-estimate effective dose. In cases where personnel are approaching dose limits, it may be prudent to more accurately evaluate effective dose using special calibrations.

6.2 RADIATIONS IN PLUTONIUM FACILITIES

As outlined in Section 2.0 of this report, plutonium emits a wide variety of radiations, including alpha and beta particles, as well as more penetrating X-rays and gamma rays. Because of the short half-life of ^{241}Pu, the radioactive decay progeny are also important sources of radiation. This section outlines methods to calculate doses from radiations emitted by plutonium and its progeny. Examples of measured dose rates are also included.

6.2.1 Alpha and Beta Doses

Plutonium is primarily an alpha-emitter and is of great concern if inhaled, ingested or injected into the body. However, the skin is an effective barrier to alpha particles, and external contamination is only a problem if there is a wound or break in the skin.

Plutonium-241 is a beta-emitter that produces low-energy beta particles with a maximum energy of 0.022 MeV. Both alpha and beta particles are completely shielded by thin rubber gloves or other protective devices. The dose rate through a rubber glove originates primarily from the X-rays and low-energy photons generated from plutonium and ^{241}Am, the decay progeny of ^{241}Pu.

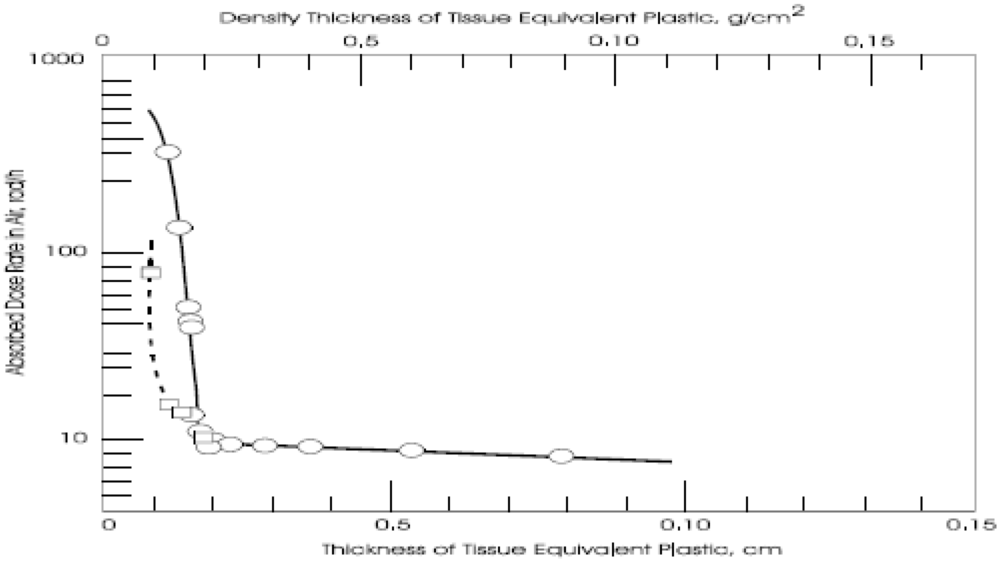

Figure 6.1. Absorbed Surface Dose Rate from Plutonium Dioxide as Measured with an Extrapolation Chamber

Figure 6.1 shows the dose rate as a function of tissue equivalent plastic absorber thickness, as measured by an extrapolation chamber in contact with a 3-in.-diameter plutonium dioxide source coated with a very thin layer of beryllium for contamination control. The plutonium was compressed to about 80% of its theoretical density and vitrified by a Dynapack process in which powder was compressed into a glassy solid by extreme pressure and heat evolved during the compression process. The plutonium oxide disk is mechanically stable and produces little smearable contamination. Even minute layers of tissue equivalent plastic reduce the dose rate significantly, as shown in Figure 6.1.

6.2.2 Gamma Doses

There can be substantial gamma doses involved in the processing and handling of plutonium, particularly in glove-box operations involving plutonium dioxide powders. Plutonium emits very few highly penetrating gamma rays; most of the radiations are L X-rays, which are very easily shielded. Because most of the photons emitted by plutonium are of low energy, plutonium sources are “infinitely thick” relative to their photon radiations, i.e., an additional thickness of plutonium does not appreciably increase the photon dose rate. A plutonium metal source of about 1-mm thickness or a plutonium oxide source about 6-mm-thick is “infinitely thick” due to self-shielding.

The age and isotopic composition are very important in determining the dose rate from plutonium because of the ingrowth of ^{241}Am from the decay of ^{241}Pu, which has a half-life of only 15 years. (The growth of plutonium daughters was discussed in detail in Section 2.1.1.) Old plutonium processing facilities can have high gamma dose rates, particularly from nearly invisible dust layers containing ^{241}Am, which has a 37% probability of emitting a 60-keV photon per alpha disintegration. A surprising amount of plutonium oxide powder can be found in dust layers on the interior surfaces of glove boxes because of the very high density of plutonium. For example, a 0.001-in.-thick layer of plutonium oxide dust on the 4-ft by 8-ft floor of a glove box can contain almost 200 grams of plutonium. Even though a glove box has additional iron or lead shielding, high gamma dose rates can persist because of the photons emitted by dust layers on the surface of gloves. Covers shall be placed over glove ports to reduce gamma dose rates around plutonium processing lines.

Doses to the extremities are usually dominated by gamma rays in typical glove-box operations. Extremity dosimeters shall be used by all personnel who perform hand contact operations with plutonium or who are involved in the manual decommissioning of plutonium facilities. Extremities are defined as the hands and forearms below the elbows and the feet and legs below the knees. In a plutonium facility, the contact doses to the hands and forearms are the most limiting cases. The extremity dose is more limiting than a whole body dose if the dose gradient is greater than 10:1 over a distance of 1 meter, the maximum distance from the fingers to the trunk of the body. In most cases, the source is not at arm's length and the dose gradient needs to be 10:1 or 20:1 for the extremity dose to be limiting (NUREG/CR-4297, Reece et al., 1985). But in highly shielded glove boxes, it is possible to have very high extremity doses from dust layers on gloves; the dose to the torso can be much lower because of shielding applied to the glove box.

6.2.2.1 Measured Gamma Dose Rates

There is a considerable amount of experimental data for measured photon dose rates from plutonium glove-box operations as recorded in progress reports issued by the Hanford Engineering Development Laboratory from the Personal Dosimetry and Shielding Program. For example, the photon dose rates were measured on an anthropomorphic Remab arm phantom inserted into gloves in a plutonium glove box. The arm phantom contains a human skeleton surrounded by tissue equivalent fluid inside a molded plastic “skin.” Thermoluminescent dosimeters (TLD-700s) were positioned at various locations along the surface of the arm phantom and inside tubes inserted into the bones.

Measurements were first made in a “clean” glove box before it was placed into service. The arm phantom was placed inside the glove and positioned in contact with a 1-quart steel can (nominal wall thickness of 10 mil or 0.25 mm), containing 1 kg of plutonium dioxide with the isotopic composition shown in Table 6.4. Measurements were made at the various locations with the arm phantom inside 20-mil Neoprene gloves (average thickness 0.021 in., 0.53 mm) and inside 37-mil (0.94-mm) lead-loaded Neoprene gloves.

The data shown in Table 6.5 are the average dose rates measured by three TLD-700s with the indicated one standard deviation in the measured values. As one would expect, the palm and fingers had the highest dose rates, approximately 300 mrad/h; the lowest dose rates of 1 mrad/h were measured at the top of the arm. Because the plutonium was "infinitely thick" and lower-energy photons were removed by the shielding provided by the steel can, the dose rates in the lead-loaded glove were only slightly lower than those in the Neoprene glove. The can of plutonium was removed, and the gloves dusted with high-exposure plutonium with an isotopic composition similar to that given in Table 6.4. The arm phantom was inserted into 20-mil Neoprene and 37-mil lead-loaded Neoprene gloves; the dose rates measured with TLDs are shown in Table 6.6.

Table 6.4. Isotopic Composition of the Plutonium Used in the Extremity Dosimetry Measurements

Isotope	Weight Percent
^{236}Pu	0.000003
^{238}Pu	0.58
^{239}Pu	72.1
^{240}Pu	19.15
^{241}Pu	6.29
^{242}Pu	1.88
^{241}Am	0.02

As expected, the highest dose rates were recorded on the hand, wrist and forearm, where the most PuO_2 dust had accumulated, and the lowest dose rates were on the upper arm and humerus. For thin dust layers, the dose rates inside the lead-loaded glove were generally much lower, typically a factor of 4 to 5 times less than the dose rates inside the Neoprene glove. The lead-loaded glove provided significantly better shielding for the 60-KeV photons from ^{241}Am and the L x-rays from plutonium, which were responsible for much of the dose. In these examples, the dose rates from the contaminated glove were about 10% of those from the 1 kg of plutonium dioxide inside the steel can. Additional experiments with 25% PuO_2- 75% normal UO_2 showed that dose rates increased as dust loadings increased with use; the dose rates on the hand and forearm increased to levels of about 30 mrem/h to 20 mrem/h, respectively.

Table 6.5. Gamma Dose Rates Along an Arm Phantom in Contact with a Steel Can Containing 1 kg of Plutonium Dioxide in an Uncontaminated Glove Box

	Gamma Dose Rates, mrad/h	
Position	Neoprene Glove	Lead-Loaded Glove
Ring Finger	330 ± 6	272 ± 25
Palm	292 ± 9	220 ± 16
Back of Hand	72 ± 2	65 ± 1
Wrist		
Inside	84 ± 6	56 ± 5
Outside	31 ± 1	24 ± 1
Forearm		
Inside	22 ± 0.4	12 ± 1
Outside	4.4 ± 0.1	3.8 ± 0.4
Elbow		
Inside	4.8 ± 0.1	2.6 ± 0.2
Outside	1.4 ± 0.1	1.8 ± 0.4
Front	2.9 ± 0.2	2.1 ± 0.1
Bottom of humerus	2.2 ± 0.1	2.5 ± 0.5
Lower mid-arm	7.1 ± 0.1	3.9 ± 0.3
Lower mid-humerus	3.8 ± 0.1	2.3 ± 0.2
Upper mid-arm	2.4 ± 0.1	2.5 ± 0.2
Upper mid-humerus	1.8 ± 0.1	1.8 ± 0.2
Top of arm	0.9 ± 0.03	2.2 ± 0.8
Top of humerus	1.1 ± 0.2	1.3 ± 0.1

The gamma energy spectra from plutonium sources are highly variable, depending on the amount of shielding present, including self-shielding. Small lightly shielded sources, such as dust layers on the interior of glove boxes, are dominated by L X-rays and the 60-keV photons from ^{241}Am, the decay progeny of ^{241}Pu.

But the gamma energy spectra are quite different in storage vaults and other facilities where the plutonium is encapsulated. In those cases, the low-energy photons have been shielded out, and the spectrum is dominated by higher photon energies. Note that plutonium metal buttons or cans of plutonium oxide prepared for storage are self-shielded, and high-energy photons from decay progeny such as ^{237}U become increasingly important.

Table 6.6. Gamma Dose Rates Measured with an Arm Phantom Placed Inside Gloves Dusted with Plutonium Dioxide Powder

	Gamma Dose Rates, mrad/h	
Position	Neoprene Glove	Lead-Loaded Glove
Palm	10.0 ± 0.4	9.5 ± 16
Back of Hand	21.8 ± 1.3	5.4 ± 0.3
Wrist		
Inside	22.6 ± 0.7	9.0 ± 0.6
Outside	22.5 ± 0.6	5.8 ± 0.4
Forearm		
Inside	34.5 ± 0.2	6.7 ± 0.4
Outside	16.7 ± 0.2	3.6 ± 0.6
Elbow		
Inside	17.5 ± 0.4	5.3 ± 0.4
Outside	11.4 ± 0.1	3.7 ± 0.4
Front	---	4.4 ± 0.3
Bottom of humerus	3.5 ± 0.2	3.6 ± 0.4
Lower mid-arm	6.7 ± 0.5	3.4 ± 0.2
Lower mid-humerus	2.1 ± 0.2	2.9 ± 0.3
Upper mid-arm	4.6 ± 0.1	2.3 ± 0.4
Upper mid-humerus	1.0 ± 0.1	1.4 ± 0.3
Top of arm	0.8 ± 0.2	3.7 ± 0.3
Top of humerus	---	1.1 ± 0.3

6.2.2.2 Calculated Photon Dose Rates

It is very difficult to accurately calculate dose rates from plutonium because of the wide range of photon energies and the relatively low abundance of photons. Most of the photons are of relatively low energies, usually below 425 keV, which are easily shielded. For heavily shielded spectra, the high-energy photons from decay progeny become very important, as well as the high-energy photons from plutonium, which have very low abundances.

For this reason, there are only a few computer codes that give accurate dose rates for plutonium. Many computer codes do not calculate the photons from progeny from radioactive decay. Others do not include the high-energy photons which have very low abundances, but which become very important for massive shields. One shall check the photon libraries to make certain that the higher-energy photons are included. Also, many point kernel codes may not give accurate results for thin shields because low-energy build-up factors are not very accurate.

There are only a few codes specifically designed for plutonium dose calculations in the Radiation Safety Information Computational Center (RSIC)(1); they include the following:

-- PUSHLD - Calculation of Gamma Radiation Dose Rates from Three- Dimensional Plutonium Sources and Shield Geometries at Various Distances, HEDL-TME 73-89, Hanford Engineering Development Laboratory (Strode, 1974).

-- BMC-MG - A Multigroup Monte Carlo Kernel Integration Neutron and Gamma-Ray Shielding Code System for Plutonium, BNWL-1855, Pacific Northwest Laboratory (Zimmerman, 1975).

-- PURSE - A Plutonium Radiation Source Code, PNCT 852-78-13, Japan Power Reactor and Nuclear Fuel Development Corp., Tokai-Mura, Japan.

The PUSHLD computer code has the advantage that the calculated results were experimentally verified to make certain that the low-energy build-up factors were correct. There are undoubtedly several other codes that could give accurate dose rates from plutonium, particularly if a radioactive decay code is used to calculate the amount of progeny as a function of time.

There are some empirical equations that can be used to calculate dose rate through simple shields, such as Neoprene, when plutonium is directly handled in a glove box. Because of the dominance of low-energy X-rays, the surface dose rates from plutonium sources can be quite high. Roesch and Faust have derived a formula for predicting the surface dose rate from plutonium through a 100-mg/cm^2 shield:

$$\begin{aligned} D_s(\text{rad/h}) = {} & 171\ f_{238} + 0.51\ f_{239} + 2.4\ f_{240} + 8.7\ f_{241} \\ & + 0.15\ f_{242}\,(0.074\ f_{241})t \end{aligned} \tag{6.2}$$

where D_s = the surface dose rate of plutonium metal or oxide, rad/h

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(1) RSICC, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6171, Telephone (865)-574-6176.

f_i = the weight fraction of the ith isotope of plutonium

t = the time since chemical separation of the plutonium, days.

This equation is only valid for a year or so after chemical separation, when the ingrowth of ^{241}Am can be represented linearly.

A similar equation has been derived for lead-loaded rubber gloves using the calculations from the computer code PUSHLD. The 80-mil lead-loaded glove is nominally 1.9-mm (0.076-in.) thick in the palm and forearm and contains the equivalent of about 1 mm of lead. The surface dose rate, D_{PbGl}, is given by the following equation:

$$D_{PbGl}(t) = 2.83\ f_{238}\ e^{-0.00789t} + 0.104\ f_{239} + 0.0315\ f_{240} + 6.35 \times 10^{-5}\ f_{242} + f_{241}\ (158.5\ e^{-0.0016t} - 152.5\ e^{-0.0457t}) \qquad (6.3)$$

where $D_{pb}Gl(t)$ = surface dose rate as a function of time, rad/h

f_i = weight fraction of the ith plutonium isotope

t = time since chemical separation of the plutonium, years.

This equation includes the radiations from plutonium, as well as the $_{237}$U and ^{241}Am progeny from the decay of ^{241}Pu. The expression is valid for times between 50 days and 100 years after the chemical separation of the plutonium. The formula predicts dose rates from 0% to +20% of those calculated by the computer code PUSHLD.

6.2.3 Neutron Doses

Neutron doses are significant in any process or decommissioning efforts involving kilogram quantities of plutonium or gram quantities of ^{238}Pu. Neutrons originate from three sources:

-- Spontaneous fission of even isotopes of plutonium

-- alpha-neutron reactions with low-atomic-number elements, including oxygen and fluorine in plutonium compounds and impurities in metals

-- neutron-induced fissions.

Experience has shown that only spontaneous fission and alpha-neutron reactions are important. Because of strict criticality controls, most forms of plutonium have very little neutron-induced multiplication. Induced fission seems to be a problem only in metal (1 kg or more) or in very large, high-density arrays of plutonium oxide with an additional moderator.

Plutonium-238 used for heat sources deserves special attention. Even sub-gram quantities of ^{238}Pu produce appreciable neutron doses because of the extremely high spontaneous fission rate in ^{238}Pu. Also, the high specific heat of ^{238}Pu creates handling problems; small microspheres of ^{238}Pu can melt through gloves in glove boxes and produce contamination problems.

Plutonium compounds created during the plutonium manufacturing process can produce very high neutron dose rates, especially PuF_4 created during the separation and purification of plutonium. Fluorinator glove boxes typically have the highest neutron dose rates in a plutonium processing line. Although PuO_2 is the preferred form because of its chemical stability, the oxide emits almost twice as many neutrons as pure metal. Neutrons are produced in alpha-neutron reactions with ^{17}O and ^{18}O. Some PuO_2 sources used in medical applications are prepared with enriched ^{16}O to reduce neutron dose rates, but isotopic enrichment is generally not used to reduce neutron doses from plutonium compounds.

6.2.3.1 Calculated Neutron Dose Rates

Neutron dose rates can be calculated accurately with computer codes. The MCNP code has the advantage that it can calculate both neutron and photon doses through shielding and in complex arrays. The Monte Carlo codes can also calculate the effects of neutron multiplication in systems containing large amounts of plutonium.

However, neutron dose rates can also be calculated from simple empirical formulas. Unlike gamma doses, there is very little self-shielding for neutrons in subkilogram masses of plutonium. In most instances, a canister containing plutonium can be treated as a point source at the geometric center of the plutonium. The neutron dose equivalent rate from a plutonium source can be calculated by:

$$H = 0.0097\ S/r^2 \qquad (6.4)$$

where H = dose rate, mrem/h

r = distance from the center of the source, cm

S = neutron emission rate from the plutonium source.

The total neutron emission rate, S, is the product of the mass of plutonium (in grams) times Y, the total neutron yield per gram of plutonium (neutrons/second/gram) from spontaneous fission, (α,n) reactions with low atomic number elements in contact with the plutonium, and fission-induced neutrons. But kilogram quantities of metals or compressed oxides can have significant multiplication and increased emission rates.

6.2.3.2 Neutron Emission Yields

The neutrons produced by spontaneous fission and α,n reactions can be estimated from the following information. Most neutrons from spontaneous fission originate from the even plutonium isotopes: ^{238}Pu, ^{240}Pu, and ^{242}Pu. Because it is the most abundant, the isotope ^{240}Pu is the most important source of spontaneous fission neutrons. Decay progeny of plutonium have very low spontaneous neutron emissions. Table 6.7 contains spontaneous fission yields for plutonium and other isotopes that may be found in plutonium facilities within the DOE complex. These data are taken from NUREG/CR-5550 (Reilly et al., 1991) and are believed to be more current than the previously published PNL values (Faust et al. 1977, Brackenbush et al., 1988). As a rule of thumb, nuclides with even numbers of protons and neutrons have the highest spontaneous fission neutron emission rates. The spontaneous fission rate for odd-even nuclides is about 1000 times less, and the rate for odd-odd nuclides is about 100,000 less. Spontaneous fission neutrons are emitted with a Maxwellian energy distribution given by the equation:

$$N(E) = (\sqrt{E})\ \mathrm{Exp}\ (E / 1.43\ \mathrm{MeV}) \qquad (6.5)$$

where N(E) is the number of neutrons as a function of the energy E in MeV.

Table 6.7.[1] Spontaneous Fission Neutron Yields

Isotope	Total Half-Life	Spontaneous Half-Life, years	Fission Yield, n/sec-gram
^{232}Th	1.41×10^{10} y	$>1 \times 10^{21}$	$>6 \times 10^{-8}$
^{232}U	71.7 y	8×10^{13}	1.3
^{233}U	1.59×10^{5} y	1.2×10^{17}	8.6×10^{-4}
^{234}U	2.45×10^{5} y	2.1×10^{16}	5.02×10^{-3}
^{235}U	7.04×10^{8} y	3.5×10^{17}	2.99×10^{-4}
^{236}U	2.34×10^{7} y	1.95×10^{16}	5.49×10^{-3}
^{238}U	4.47×10^{9} y	8.20×10^{15}	1.36×10^{-2}
^{237}Np	2.14×10^{6} y	1.0×10^{18}	1.14×10^{-4}
^{238}Pu	87.74 y	4.77×10^{10}	2.59×10^{3}
^{239}Pu	2.41×10^{4} y	5.48×10^{15}	2.18×10^{-2}
^{240}Pu	6.56×10^{3} y	1.16×10^{11}	1.02×10^{3}
^{241}Pu	14.35 y	2.5×10^{15}	5×10^{-2}
^{242}Pu	3.76×10^{5} y	6.84×10^{10}	1.72×10^{3}
^{241}Am	433.6 y	1.05×10^{14}	1.18
^{242}Cm	163 days	6.56×10^{6}	2.10×10^{7}
^{244}Cm	18.1 y	1.35×10^{7}	1.08×10^{7}
^{249}Bk	320 days	1.90×10^{9}	1.0×10^{5}
^{252}Cf	2.646 y	85.5	2.34×10^{12}

[1] Adapted from NUREG/CR-5550 (Reilly et al., 1991)

Energetic alpha particles can overcome coulomb barriers in low-atomic-number elements and create an unstable nucleus that emits neutrons. Because of the high alpha activity of plutonium, this can be a significant source of neutrons. There are two nuclear reactions that are of importance:

$$\alpha + {}^{18}\mathrm{O} \rightarrow {}^{21}\mathrm{Ne} + \mathrm{n} \tag{6.6}$$

$$\alpha + {}^{19}\mathrm{F} \rightarrow {}^{22}\mathrm{Na} + \mathrm{n}. \tag{6.7}$$

Table 6.8 contains the alpha-neutron yields for oxides and fluorides for the most common plutonium and transuranic nuclides. Note that the neutron yields are normalized per gram of nuclide, not per gram of compound. To obtain the yields per gram of compound, multiply by 0.88 for PuO_2 and 0.76 for PuF_4. These data are taken from NUREG/CR-5550 (Reilly et al., 1991).

Table 6.9 contains the neutron yields for trace amounts of elemental impurities in plutonium metal or oxide. These data are also from NUREG/CR-5550 (Reilly et al., 1991) and are derived from thick target yields from accelerator data. The data in Table 6.9 differ from previous values in BNWL-2086 (Faust et al., 1977), and the authors have not experimentally checked the accuracy of these values. Two sets of data are included: one for alphas emitted from enriched uranium and the other for alphas emitted from ^{239}Pu. To determine the neutron yield from trace impurities, it is first necessary to determine the specific alpha activity from Table 6.8, and the neutron yield per parts per million per 10^6 alphas from Table 6.9 for either enriched uranium or plutonium. The specific neutron yield from impurities can be estimated from the following formula:

$$Y_{imp} = 10^{-12} A_{\alpha} \sum_{j}^{n} P_j \; I_j \qquad (6.8)$$

where A_{α} = alpha activity of the plutonium nuclides

P_j = specific neutron yield from the jth element (neutrons/alpha part per million) from Table 6.9

I_j = elemental impurity concentration in plutonium (parts per million).

Note that this formula is valid only if the impurities are uniformly distributed with the plutonium so that the alpha particles directly interact with the impurities. Dust layers of plutonium oxide can also produce high neutron yields. For example, plutonium oxide dust layers on HEPA filters with borosilicate glass can produce neutron emission rates 10 times higher than those for pure oxide because of alpha-neutron reactions with boron in the glass fibers and aluminum spacer plates.

The total neutron yield per gram of plutonium can be found by summing the contributions from:

- Spontaneous fission (from Table 6.7)
- alpha-neutron reactions in oxides or fluorides (from Table 6.8)
- neutrons from low-atomic-number impurities (from Table 6.9).

Table 6.8. Neutron Yields from Alpha-Neutron Reactions for Oxides and Fluorides

Isotope	Alpha Decay Half-Life	Alpha Yield α /s-g	Average Alpha Energy MeV	α , n Yield in Oxides n/s-g	α , n Yield in Fluorides n/s-g
^{232}Th	1.41 x 10^{10} y	4.1 x 10^{3}	4.00	2.2 x 10^{-5}	
^{232}U	71.7 y	8.0 x 10^{11}	5.30	1.49 x 10^{4}	2.6 x 10^{6}
^{233}U	1.59 x 10^{5} y	3.5 x 10^{8}	4.82	4.8	7.0 x 10^{2}
^{234}U	2.45 x 10^{5} y	2.3 x 10^{8}	4.76	3.0	5.8 x 10^{2}
^{235}U	7.04 x 10^{8} y	7.9 x 10^{4}	4.40	7.1 x 10^{-4}	0.08
^{236}U	2.34 x 10^{7} y	2.3 x 10^{6}	4.48	2.4 x 10^{-2}	2.9
^{238}U	4.47 x 10^{9} y	1.2 x 10^{4}	4.19	8.3 x 10^{-5}	0.028
^{237}Np	2.14 x 10^{6} y	2.6 x 10^{7}	4.77	3.4 x 10^{-1}	
^{238}Pu	87.74 y	6.4 x 10^{11}	5.49	1.34 x 10^{4}	2.2 x 10^{6}
^{239}Pu	2.41 x 10^{4} y	2.3 x 10^{9}	5.1	3.81 x 10^{1}	5.6 x 10^{3}
^{240}Pu	6.56 x 10^{3} y	8.4 x 10^{9}	5.15	1.41 x 10^{2}	2.1 x 10^{4}
^{241}Pu	5.90 x 10^{5} y	9.4 x 10^{7}	4.89	1.3	1.7 x 10^{2}
^{242}Pu	3.76 x 10^{5} y	1.4 x 10^{8}	4.90	2.0	2.7 x 10^{2}
^{241}Am	433.6 y	1.3 x 10^{11}	5.48	2.69 x 10^{3}	
^{242}Cm	163 days	1.2 x 10^{14}	6.10	3.76 x 10^{6}	
^{244}Cm	18.1 y	3.0 x 10^{12}	5.80	7.73 x 10^{4}	
^{249}Bk	6.6 x 10^{4} y	8.8 x 10^{8}	5.40	1.8 x 10^{1}	
^{252}Cf	2.646 y	1.9 x	10^{13}	6.11	6.0 x 10^{5}

Multiplying the specific neutron yield (neutrons/second-gram of plutonium) by the mass of plutonium (grams) gives S, the neutron emission rate (neutrons/second). The dose rate is then calculated using Equation 6.4.

Table 6.9. Neutron Yields for Trace Impurities in Plutonium and Uranium

Element	Neutron Yield Per 10^6 Alphas at 4.7 MeV (^{234}U)	Neutron Yield Per 10^6 Alphas at 5.2 MeV (^{234}U)	Average Neutron Energy in MeV For 5.3-MeV Alphas from Pu
Li	0.16 ± 0.04	1.13 ± 0.25	0.3
Be	44. ± 4	65. ± 5	4.2
B	12.4 ± 0.6	17.5 ± 0.4	2.9
C	0.051 ± 0.002	0.078 ± 0.004	4.4
O	0.040 ± 0.001	0.059 ± 0.002	1.9
F	3.1 ± 0.3	5.9 ± 0.6	1.2
Na	0.5 ± 0.5	1.1 ± 0.5	
Mg	0.42 ± 0.03	0.89 ± 0.02	2.7
Al	0.13 ± 0.01	0.41 ± 0.01	1.0
Si	0.0028 ± 0.002	0.076 ± 0.003	1.2
Cl	0.01 ± 0.01	0.07 ± 0.04	

2.3.3 Radiation Weighting Factors for Neutrons

Approved Radiation Weighting Factors for neutrons are provided in 10 CFR 835.2. As used here Radiation Weighting Factor means the principal modifying factor used to calculate the equivalent dose from the absorbed dose; the absorbed dose (expressed in rad or gray) is multiplied by the appropriate Radiation Weighting Factor (wR).The Radiation Weighting Factors to be used for determining equivalent dose in rem for neutrons are as follows:

RADIATION WEIGHTING FACTORS[1], w_R

Type and energy range	Radiation weighting factor
Photons, electrons and muons, all energies	1
Neutrons, energy < 10 keV[2, 3]	5
Neutrons, energy 10 keV to 100 keV[2, 3]	10
Neutrons, energy > 100 keV to 2 MeV[2, 3]	20
Neutrons, energy > 2 MeV to 20 MeV[2, 3]	10
Neutrons, energy > 20 MeV[2, 3]	5
Protons, other than recoil protons, energy > 2 MeV	5
Alpha particles, fission fragments, heavy nuclei	20

[1] All values relate to the radiation incident on the body or, for internal sources, emitted from the source.

[2] When spectral data are insufficient to identify the energy of the neutrons, a radiation weighting factor of 20 shall be used.

[3] When spectral data are sufficient to identify the energy of the neutrons, the following equation may be used to determine a neutron radiation weighting factor value:

$$w_R = 5 + 17 \exp\left[\frac{-(\ln(2E_n))^2}{6}\right]$$

Where E_n is the neutron energy in MeV.

6.3 RADIATION DETECTION AND EVALUATION

This section describes the response of portable instruments, personnel dosimeters, and nuclear accident dosimeters to the radiations emitted by plutonium, with a focus on photons and neutrons. Data are also included on special spectrometry instruments used to calibrate dosimeters in the field.

6.3.1 Response of Portable Survey Instruments

The energy and angular responses of almost all portable gamma survey instruments have been well characterized and published in the instruction manuals available from the manufacturers. Because of the preponderance of low-energy photons, especially the 60-keV photons emitted by ^{241}Am, particular attention should be given to the low-energy response.

It is not generally well known that neutron survey instruments have a severe energy dependence. In fact, some manufacturers claim a ±15% response per unit dose equivalent extending over an energy range of thermal to 15 MeV. The energy dependence of several commercially available neutron survey meters has been experimentally measured at the PTB in Germany (Liesecki and Cosack, 1984). Their measurements made with monoenergetic neutrons in low-scatter conditions demonstrate that a typical moderator-based neutron survey meter underestimates the dose equivalent by a factor of 2 at an energy of 14 MeV and overestimates dose equivalent by a factor of 2 to 3 at an energy of 20 keV. Survey instruments also exhibit changes in response with the direction of incidence of the neutrons due to absorption and scattering of the neutrons by the electronics package attached to the moderator/detector. This can also result in 40% variation in response, depending on the direction of incidence. Fortunately, plutonium compounds emit neutrons in the MeV range, where the problems with energy and angular responses are minimal. Accuracies of ±15% can be achieved with careful calibration with ^{252}Cf or other fission sources.

6.3.2 Personnel Dosimetry

The detailed requirements of an external dosimetry program are given in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). Explicit guidance and requirements are given and need not be repeated here. This section will focus on dosimetry problem areas specific to plutonium facilities and possible solutions.

Personnel working in plutonium facilities are exposed to both photon and neutron radiations, and plutonium processing is one of the largest contributors to neutron exposure in the United States. The response of beta-gamma personnel dosimeters is well documented and will not be discussed here.

The response of neutron dosimeters to the neutron fields encountered in the workplace shall be evaluated. All existing neutron dosimeters have a severe energy response problem and shall be carefully calibrated for the specific radiation field in which the neutron dosimeter is worn. Typically, neutron dosimeters are calibrated to either bare ^{252}Cf or D_2O-moderated ^{252}Cf sources in a low-scatter calibration facility. Then, the neutron dosimeters are worn in plutonium facilities under high-neutron-scatter conditions, which produce a completely different energy spectrum than that in which the dosimeter was calibrated. Accordingly, a typical approach is to perform neutron energy field characterization surveys at selected areas in a facility. Based on these surveys a field correction factor is determined which corrects for the difference between the dosimeter response to the calibration source and the response to the neutron fields in the workplace. Because of the large response of TLD-albedo dosimeters to low-energy neutrons (with energies below about 20 keV), the response of the dosimeter usually depends on the scattering conditions rather than the initial neutron energy spectrum. These problems are discussed in detail in several documents, including PNL-3213, Personnel Neutron Dosimetry at Department of Energy Facilities (Brackenbush et al., 1980) and PNL-7881, Response of TLD-Albedo and Nuclear Track Dosimeters Exposed to Plutonium Sources (Brackenbush et al., 1991).

Thermoluminescent dosimeters are the most widely used neutron dosimeters in plutonium facilities. The energy response of a typical TLD-albedo dosimeter is shown in Figure 6.2. At neutron energies below about 20 keV, the energy response is almost constant. Above 20 keV, the response per unit dose drops dramatically by almost three orders of magnitude at 10 MeV. Almost all neutrons emitted by plutonium have energies in the MeV range. However, about 50% of the neutrons striking a thick concrete wall or floor are reflected back into the room at lower energies, and neutrons typically are reflected two or three times before being absorbed. Thus, the low-energy scattered neutrons are often more important in determining TLD-albedo dosimeter response than the high-energy neutrons emitted by the plutonium source. The TLD-albedo dosimeters are often calibrated in specific facilities by exposing them on phantoms at locations where the dose equivalent has been carefully determined from dose and spectrometric measurements (Brackenbush et al., 1991).

Nuclear track dosimeters are also being used for personnel dosimetry in plutonium facilities. These dosimeters have the advantage of a much more constant response per unit dose equivalent, as shown in Figure 6.3. Nuclear track dosimeters operate on the principle that a fast neutron interacts with plastic to produce a proton recoil that damages the polymer. Under special etch conditions, the damaged areas are removed to produce a distinct track, which is easily observed under a microscope. The neutron dose equivalent is then determined from the track density. Nuclear track dosimeters have a distinct threshold, usually about 100 keV.

In conclusion, the combination of TLD-albedo and nuclear track dosimeters can provide a more uniform response with energy and more accurate personnel dosimetry. This combination of dosimeters may be an appropriate solution to neutron dose monitoring in DOE facilities with significant neutron exposure.

It is important to verify and document that personnel dosimetry systems provide accurate measurements and records of the occupational radiation doses received by workers in plutonium facilities (McDonald et al., 1992). To provide a level of confidence in dosimetry services in DOE facilities, the DOELAP accreditation program was established. 10 CFR 835 402(b) (DOE, 2007a) requires that personnel dosimetry programs implemented to demonstrate compliance with the dose limits established in Subpart C shall be accredited in accordance with the requirements of the DOELAP for Personnel Dosimetry (ANSI, 2009) (DOE, 2011e). NIST has also established the National Voluntary Laboratory Accreditation Program (NVLAP) for testing and accreditation of dosimeter processors serving commercial industry and medical facilities. Because the dosimetry needs at many DOE facilities, particularly those processing plutonium, are different from commercial industries, the DOE initially established a broader and more stringent accreditation program. Both DOELAP and NVLAP accreditation programs use performance tests that evaluate the accuracy and precision of personnel dosimetry measurements. The accuracy is determined by comparing the measured dose equivalent to the "conventionally true dose equivalent" derived from calibration standards directly traceable to NIST in carefully controlled conditions.

Two laboratories conduct the performance test irradiations for the DOELAP and NVLAP programs: Pacific Northwest National Laboratory of Richland, Washington, and the Radiological and Environmental Sciences Laboratory (RESL) of Idaho Falls, Idaho. Dosimeters are submitted for testing to the performance testing laboratories in specified categories. If the dosimeter passes certain accuracy and tolerance testing criteria, a team of dosimetry experts visit the processor and/or site and assess the operation of the dosimetry program, including dosimetry records and data retrieval systems, before the dosimeter processor or DOE site is accredited. DOE requirements are given in the DOE STD-1095- 2011 (DOE, 2011e).

6.3.3 Extremity Dosimetry

Doses to the extremities from plutonium processing and handling can involve significant exposures to the skin of the hands and forearms. For information on performance testing of extremity dosimeters refer to DOE STD-1095- 2011 (DOE, 2011e) which incorporates ANSI/HPS N13.32-2008, Performance Testing of Extremity Dosimeters (ANSI, 2008b). Doses over small areas of the skin are discussed in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a) and will not be discussed here. That chapter discusses skin contamination including hot particles, and the determination of skin dose from these events.

Highly accurate measurement of the dose to the hands and forearms is especially difficult because of the low-energy photons (L x-rays and 60-keV photons from ^{241}Am). Small variations in shielding, such as differences in the thickness of gloves used in glove boxes or non-uniform distribution of plutonium oxide dust on the surface of gloves, can produce large variations in the dose rate. Examples of these variations were given previously.

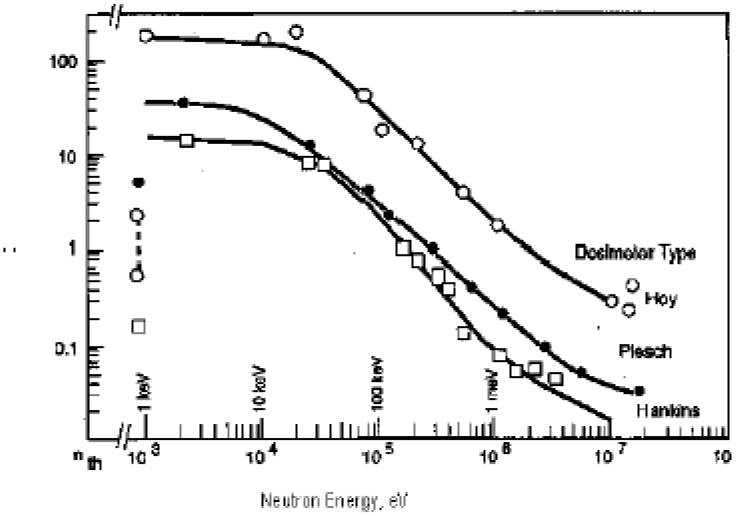

Figure 6.2. Energy Dependence of Various TLD-Albedo Dosimeters (Source: Piesch and Burgkhardt, 1978)

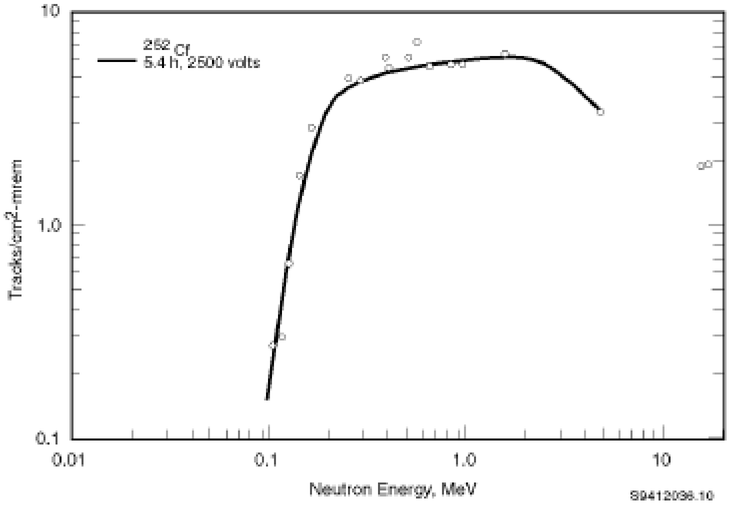

Figure 6.3. Response of Electrochemically Etched CR-39 Used in Nuclear Track Dosimeters as a Function of Neutron Energy

Extremity doses are typically determined by TLD finger rings, which are usually worn with the TLD chip facing the radiation source on the palm side of the hand. In glove-box and in D&D operations, the photon dose is usually significantly higher than any neutron dose. However, neutron dosimeters are sometimes worn to estimate extremity doses. Two types of neutron extremity dosimeters have been used: nuclear track dosimeters worn in special finger rings and specially calibrated TLD-albedo dosimeters worn on the wrist or forearm. DOE STD-1095-2011 (DOE, 2011e) is currently applicable to personnel dosimeters for whole body irradiation.

There is some question about the correct radiation weighting factor to apply to extremity neutron dosimeters. Radiation weighting factors were derived from biological experiments on cancer induction, especially leukemia in blood-forming organs. There are no blood-forming organs in the extremities, so there is no biological basis for large values of radiation weighting factors for extremity exposures. However, regulatory agencies typically apply radiation weighting factors derived for whole-body exposures to the extremities, thus for compliance purposes radiation weighting factors should be applied for extremity exposures.

6.3.4 Criticality Accident Dosimetry

A criticality safety program, which includes material control, criticality alarms, and criticality accident dosimetry, is required as outlined in DOE Order 420.1C (DOE, 2012b). The requirements in 10 CFR 835.1304 require that fixed nuclear accident dosimeters (NADs) and personnel nuclear accident dosimeters (PNADs) shall be worn by all individuals entering a controlled area that contains certain quantities of fissile materials, such as those required in DOE Order 420.1C (DOE, 2012b); which requires installed criticality alarms. The criticality accident dosimetry system should follow the provisions of ANSI N13.3, Dosimetry for Criticality Accidents (ANSI, 1969); this standard, although currently withdrawn from ANSI/HPS, is being revised. Information on criticality accident dosimetry is also available from the International Atomic Energy Agency (IAEA, 1982).

The criticality accident program should contain the following items:

- a method and procedure to conduct an initial screening of individuals involved in a nuclear accident to determine whether significant exposures to radiation occurred (10 CFR 835.1304(b)(1))

- methods, procedures, and equipment for obtaining and analyzing biological materials (including ^{24}Na activity from blood samples and ^{32}P activity in the hair)(10 CFR 835.1304(b)(2)), as well as metal coins, jewelry, and articles of apparel that may have become activated from neutrons

- a system of fixed dosimeters (i.e., NADs) (10 CFR 835.1304b(3)) capable of furnishing estimated radiation dose within an accuracy of ±25% and the approximate neutron spectrum at the installed locations to allow conversion from rad to rem

-- an operating range for the fixed dosimeters' neutron component 10 rad to 10,000 rad

-- measurement capabilities for the fixed dosimeters' gamma ray component of fission gamma rays in the presence of neutrons with an accuracy of ±20%, and an operating range for the gamma component operating range extending from 10 rad to 10,000 rad

-- PNADs capable of furnishing sufficient information to determine neutron and gamma dose with an accuracy of ±25% over a range of 10 rad to 1000 rad without dependence upon fixed NAD data

-- a radiological counting laboratory with the methodology, analytical procedures, and quality assurance program in place to count the activated samples from the criticality accident and provide results quickly

-- counting of activities in persons with significant exposures to assess the activation products in the body if a whole body counter is available (this is one of the more accurate methods for dose estimation)

-- a health physicist designated to coordinate and evaluate the dosimetry information and provide dose estimates shortly after the accident

-- means to obtain medical treatment for personnel who receive a high radiation dose

-- a quality assurance program in place to help assure the accuracy and validity of the dosimetry results.

As mentioned in Section 6.1, the concept of equivalent dose was used to quantify exposures to different radiations. The radiation weighting factors used to determine equivalent dose are based on stochastic effects, primarily cancer induction some years later. But the doses in criticality accidents are typically so large that acute symptoms, including death, may occur within relatively short times, and radiation weighting factors are usually not applicable. For this reason, it is usually more appropriate to determine absorbed dose rather than equivalent dose if a person receives more than about 25 rem. These absorbed dose estimates to the torso are much more important for triage and treatment considerations.

The NADs are used to determine the neutron and photon dose at various locations in the plutonium facility, as well as providing spectral and calibration data for PNADs. A typical NAD used at the Hanford Site is shown in Figure 6.4. This unit is fixed to the wall or posted at locations around plutonium storage areas where it is easily recovered in the event of a criticality. The “candle” insert contains neutron- and gamma-sensitive TLDs as well as activation foils positioned at the center of the detector. Tests at the Health Physics Research Reactor at Oak Ridge have shown that this arrangement gives accurate estimates of “deep” dose for both neutrons and gamma rays. A set of foils identical to those used in the PNAD dosimeter is positioned above the moderator. These foils provide an estimate of the average cross-section or response per unit dose, so that the neutron dose from the foils in the PNAD can be more accurately evaluated for the incident neutron spectrum.

The PNAD dosimeter typically consists of several activation foils. In the case of the Los Alamos/Hanford design (Vasilik and Martin, 1981), the activation foils consist of ½-in.- diameter foils of bare and cadmium-covered gold, bare and cadmium-covered indium, cadmium-covered copper, and a sulfur pellet. Algorithms have been developed to unfold an approximate neutron energy spectrum from the measured neutron activation products, so that neutron doses can be calculated. Criticality dosimeters containing various activation foils are available from vendors, but some of the commercial products do not contain sufficient material to measure neutron doses as low as 10 rad, which is the recommended lower detection limit for personal criticality accident dosimeters.

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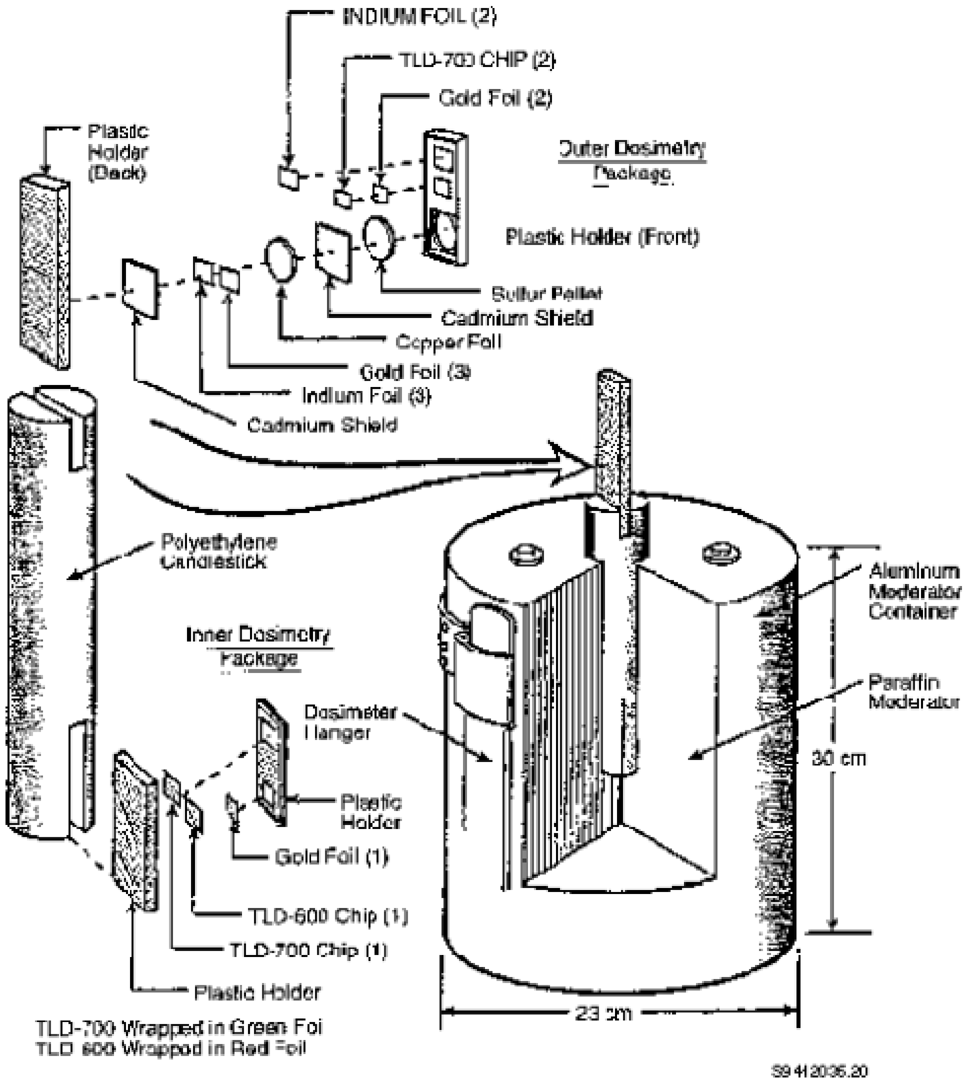

Figure 6.4 Fixed Nuclear Accident Dosimeter Used at Hanford to help Assess Doses from Criticality Accidents

6.3.5 Dose to Lens of Eye

The dose to the lens of the eye is not generally a problem in plutonium facilities because whole body exposures are generally the limiting case. Dosimeters to measure the dose to the lens of the eye are seldom used. However, equivalent doses to the extremity and skin at depths of 3 mm can be appreciable in cases where there is an abundance of low-energy photons, such as during visual inspection of machined plutonium pieces on laminar-flow tables or other situations where the plutonium is not shielded. In these cases, the eyes are generally protected by requiring safety glasses to be worn.

6.3.6 Spectrometry Measurements

Personnel neutron dosimeters used at DOE plutonium facilities include TLD-albedo and nuclear track detectors. The response per unit equivalent dose for TLD-albedo dosimeters is a sensitive function of incident neutron energy (see Figure 6.2). These dosimeters are typically calibrated under low-scatter conditions in a calibration laboratory, such as the facility at PNL used for exposing dosimeters for DOELAP accreditation. The dosimeters are calibrated to a fission spectrum from ^{252}Cf or a degraded fission spectrum from D_2O-moderated ^{252}Cf. However, the neutron energy spectrum of the workplace is significantly different from that of the calibration facility and the response per unit equivalent dose is also different, primarily because of the number of low-energy neutrons produced by scatter within process equipment, glove boxes, and the walls and floor of the facility. To achieve accurate results, the TLD-albedo dosimeter results shall be corrected for the specific neutron energy spectrum in which they are exposed. One method to achieve accuracy is to expose neutron dosimeters on a phantom in the workplace in neutron fields where the equivalent dose rate has been carefully measured using neutron spectrometers.

There are several neutron energy spectrometers available to make accurate neutron spectrum measurements and dose estimations, as outlined in the document A Field Neutron Spectrometer for Health Physics Applications (Brackenbush et al., 1992). Neutron spectrometers that are useful for dose determinations in plutonium facilities include:

- Multisphere or Bonner sphere spectrometers
- tissue equivalent proportional counters (to determine linear energy transfer (LET) spectra)
- liquid scintillator spectrometers
- proton recoil spectrometers.

6.3.6.1 Multisphere Spectrometer System

The multisphere or Bonner sphere spectrometer (Bramblett et al., 1960) is the neutron spectrometer system most often used by health physicists for neutron energy spectrum measurements, perhaps because it is simple to operate. Multisphere spectrometers are typically used for measuring neutron energy spectra over a wide energy range from thermal energies to over 20 MeV although detailed energy spectra are not obtained. With the use of an appropriate spectrum unfolding code, the multisphere system will determine the average neutron energy, dose rate, total flux, kerma, and graphical plots of differential flux versus energy and dose distribution versus energy.

The multisphere spectrometer consists of a set of polyethylene spheres of different diameters, typically 3 in. to 12 in. A thermal neutron detector, such as a ^{3}He proportional counter or a ^{6}LiI scintillator is positioned at the center of each sphere, and the count rate measured. The neutron energy spectrum can be determined from the ratio of counts from different detectors. However, the spectral unfolding algorithms do not provide mathematically unique solutions. The most appropriate solutions are obtained by making an initial guess that the spectrum consists of a fission spectrum with a 1/E “tail.” Multisphere spectrometers have demonstrated accuracies of ±15% when exposed to ^{252}Cf sources with calibrations directly traceable to NIST (Brackenbush et al., 1991).

Figure 6.5 demonstrates the type of neutron energy spectra measured by the multisphere spectrometer. The plot shows the logarithmic plots of four multisphere spectrometer measurements made at a distance of 50 cm from 1 kg of plutonium for “bare” plutonium fluoride (i.e., no intervening shielding), plutonium fluoride shielded with 10 cm (4 in.) of acrylic plastic, “bare” plutonium oxide, and “bare” plutonium metal. The plutonium fluoride has the highest neutron emission rate and corresponds to the highest peak in the graph. The lowest peak corresponds to the moderated plutonium fluoride spectrum with 4 in. of acrylic plastic shielding. These measurements are typical of the neutron energy spectra in plutonium processing areas containing glove boxes.

The spectra contain a significant fraction of low-energy scattered neutrons from the glove boxes and the thick concrete floor and walls of the facility. The spectra are distinctly different from neutron emission spectra (see Section 6.2), which do not contain scattered or background neutrons.

6.3.6.2 Tissue Equivalent Proportional Counter

The tissue equivalent proportional counter (TEPC) is not often used by health physicists, but it can provide highly accurate estimates of dose. The TEPC consists of a hollow sphere or cylinder of tissue equivalent plastic filled with low-pressure equivalent gas. The pressure is so low (a few torr) that the TE gas cavity has the same mass stopping power as a 2-μm sphere of tissue at unit density. Because the TEPC actually measures the energy absorption in a known mass of tissue equivalent material, it provides an absolute measure of absorbed neutron dose. The TEPC also measures the pattern of microscopic energy distributions from any penetrating ionizing radiation. With appropriate algorithms, LET distributions, hence radiation weighting factors, can be calculated. Thus, the TEPC provides absorbed dose, radiation weighting factor, and dose from a single spectral measurement of the event size distribution from the TEPC.

The TEPC can provide highly accurate measurements of dose under laboratory conditions. However, it suffers from stability problems, and its accuracy decreases with time as impurities diffuse from the TE plastic walls and temperature changes cause gain shifts in the proportional counter. Nevertheless, the TEPC can provide reasonably accurate measurements of dose in the workplace (±15%) over extended time periods of 6 months or more, and can be used to monitor dosimeter irradiations on phantoms in the workplace.

6.3.6.3 Liquid Scintillator Spectrometer

The liquid scintillator spectrometer typically consists of a 2-in. by 2-in.cylindrical cell of hydrogenous scintillator solution in contact with a photomultiplier. Neutrons interact in the scintillator to produce proton recoils, which interact with the scintillator to produce light. With careful calibration, the incident neutron energy spectrum can be unfolded from the measured distribution of scintillation events.

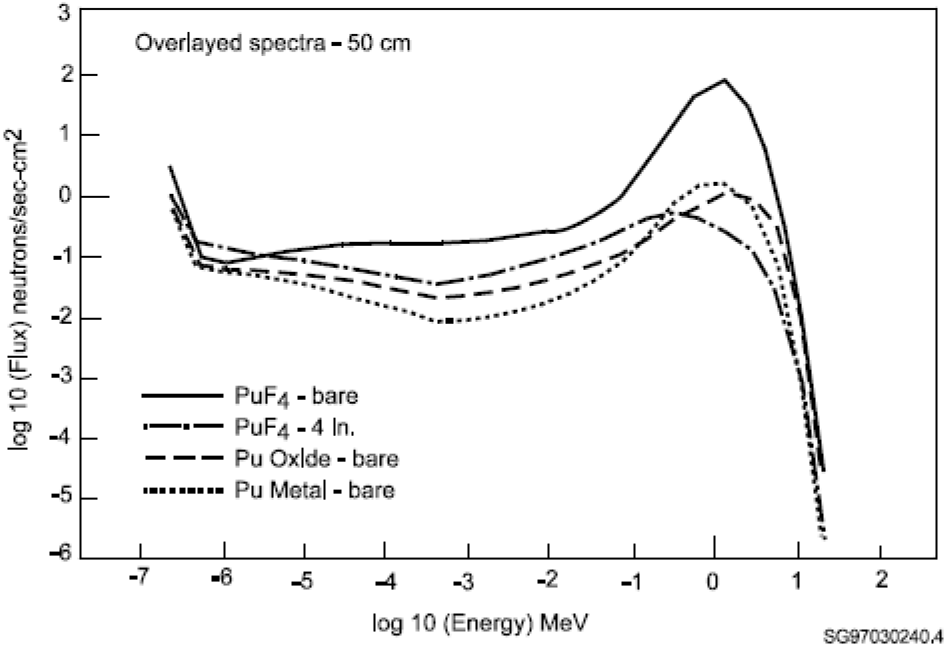

Figure 6.5. **Neutron Energy Spectra as Measured by the Multisphere Spectrometer at 50 cm from Plutonium Metal, PuO_2, and PuF_4 Sources**

The liquid scintillator spectrometer has the advantage that it is very sensitive and can operate at low dose rates. It is useful over an energy range extending from about 1 MeV to 20 MeV. Neutron equivalent dose can be calculated from the measured spectra using the conversion factors referenced in Chapter 6 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). The doses calculated from liquid scintillator measurements are reasonably accurate (±10% to ±20%) for lightly moderated plutonium spectra. Because of the lower energy cut-off of liquid scintillator spectrometers, they may not provide accurate dose equivalent values outside heavily shielded facilities, such as plutonium storage vaults with thick concrete walls.

6.4 EXTERNAL DOSE REDUCTION

The traditional methods of using time, distance, and shielding are typically employed in plutonium facilities to reduce exposures to ALARA levels. However, other considerations may be just as important. Good housekeeping practices are vital to keep dose rates low. Even invisible dust layers on the interior surfaces of glove boxes can create gamma radiation fields of 10 mrem/h or more, especially through lightly shielded glove ports. The practice of pulling gloves outside for storage should not be condoned in operations that generate dust or powders. Dose rates of 30 mrem/h have been measured in facilities processing high-exposure oxide powders. A factor of 30 reduction in dose rate was achieved by merely storing the gloves inside the glove box when not in use and placing lightweight “pie plate” shields over the glove-port openings.

6.4.1 Time

Obviously, reducing the time a worker is exposed in a radiation field will reduce the dose. Any operation which involves elevated dose rates (more than a few mrem/hour) or long exposures should be reviewed for possible reductions in a worker's exposure time. For example, a worker should minimize the time spent near a fluorination operation. After the equipment has been set up, the worker should leave the area during the actual fluorination step.

6.4.2 Distance

Because of the inverse square relationship with discrete radiation sources, significant dose reductions can be achieved by increasing the distance between the worker and the plutonium source. Also, the low-energy photons emitted through glove ports and bag-out ports can be attenuated by several feet of air. Most plutonium operations involve contact work, so increasing the distance may not always be practical. But significant reductions in doses can be achieved by reducing plutonium inventories in glove boxes. It is good practice not to store plutonium samples in glove boxes, but to remove them to storage vaults or other shielded locations. In many cases, the plutonium samples can be stored in the glove box in “wells” or specially shielded areas at some distance from the work areas where the plutonium technicians spend most of their time. The best method of reducing neutron dose is simply to remove the plutonium from the glove box and minimize inventories in the glove box.

6.4.3 Shielding

The most practical method of reducing doses in plutonium operations is to apply shielding. Plutonium emits both neutrons and photons, which require different types of shielding materials to be effective. There are also additional constraints that shall be met, such as the maximum thickness of shielding that can be placed on glove boxes and still retain worker mobility. It has been found that more than about 8 cm (4 in.) of shielding on the exterior surface of a glove box greatly reduces the worker's manual dexterity and efficiency. It is also important to place the shielding close to the plutonium source and not to try to shield personnel. Because neutrons scatter around shadow shields, it is usually best to shield all surfaces of glove boxes or storage areas. The following sections describe the shielding effectiveness of common photon and neutron shielding materials.

6.4.3.1 Photon Shielding

Because of the preponderance of low-energy photons, significant reductions in gamma doses can be achieved by even modest shielding. It is important to note that there is a significant amount of self-shielding in plutonium samples. A 1- mm-thick plutonium metal sample is “infinitely thick” and additional thicknesses will not appreciably increase the dose rate. For this reason, the photon dose is more dependent on the surface area rather than on the mass of plutonium. Invisible dust layers on gloves and interior surfaces of glove boxes can produce high exposure rates, especially if the gloves are pulled outside the glove box for storage to prevent them from being caught in machinery. Simple iron or lead shields placed over the glove ports can reduce the dose rates near the glove box by an order of magnitude. Modest gamma shields of 6 mm (0.25 in.) of lead and 13 mm (0.5 in.) of lead-loaded x-ray glass are usually sufficient to reduce photon dose rates from plutonium to acceptable levels.

Table 6.10 gives examples of how effective various gamma shielding materials are in reducing the dose rates from low-exposure (6% ^{240}Pu) and high-exposure (19% ^{240}Pu) sources. The sources consist of cylinders containing 1 kg of plutonium oxide; the dose rates are given at a distance of 2 m from the source. This example is typical of the shielding effectiveness for cans of plutonium containing kilogram quantities of plutonium oxide, as might be found in storage vaults.

In contrast, Figure 6.6 shows the reduction in photon dose rates from a small sample of plutonium oxide power weighing about 100 grams. The dose rates were measured at a distance of 3 cm from the surface of the plutonium, which was contained in polyvinyl chloride plastic bags (a total thickness of 33 mil or 0.85 mm) for radiation measurements. The isotopic composition of the plutonium was similar to that given in Table 6.11.

Photon radiation is a significant source of exposure, especially during D&D activities, when most of the plutonium has already been removed. Much of the photon exposure problem originates from thin dust layers, as described in the preceding paragraphs. High photon doses often originate from “streaming” through glove ports from dust layers on gloves. But there also can be appreciable neutron dose rates, even in supposedly “empty” glove boxes, from plutonium hold-up, especially in fluorinator glove boxes where there is a high neutron emission rate from alpha-neutron reactions. Wearing lead-loaded aprons can reduce dose rates by a factor of 2 in plutonium fuel manufacturing. High-exposure plutonium (>10% ^{240}Pu) should be handled in glove boxes with lead-loaded Neoprene gloves although some loss of mobility and dexterity may result. The photons from plutonium are easily shielded by several millimeters of lead or iron, but it requires almost 15 cm (6 in.) of polyethylene or hydrogenous moderator to reduce neutron doses by a factor of 10. Simplistically stated, the gamma dose rate is a function of surface area, while neutron dose rate is a function of the mass of the plutonium and its chemical form.

6.4.3.2 Neutron Shielding

The neutron radiations from plutonium are much more difficult to shield than the photon radiations. As a rule of thumb, it requires about 15 cm of hydrogenous shielding to reduce the neutron dose rate by an order of magnitude.

Figure 6.7 shows the reduction in dose equivalent rate for various shielding materials for plutonium tetrafluoride sources, which have an average neutron energy of 1.3 MeV. For practical purposes, the shielding thickness for glove boxes is limited to about 4 in.; it is not possible to operate machinery through thicker shields. Figure 6.8 shows the reduction in dose equivalent rate through various slab shields for plutonium dioxide. These data were obtained from measurements of the neutron dose using a TEPC. Figures 6.7 and 6.8 show the reduction in dose equivalent rates, the reduction in equivalent dose rates will differ.

Table 6.10.[1] Photon Dose Rates at 2 Meters from Cylinders of Plutonium Containing 1 kg of Plutonium at 5 Years After Chemical Separation

Shield Material	Shield Thickness cm	Photon Dose Rate, mrad/h 19% ^{240}Pu Source	 6% ^{240}Pu Source
Polyvinyl chloride (PVC)	0.005	19.3	1.74
	0.038	8.72	0.570
	0.084	6.29	0.391
Lead Glove	0.094	1.85	0.105
Heavy Lead Glove	0.152	0.54	0.0464
Lucite	0.612	7.03	0.447
	2.54	3.30	0.190
Steel	0.025	2.69	0.144
	0.038	2.41	0.131
	0.051	2.19	0.121
	0.317	0.42	0.0418
	0.635	0.221	0.0299
	1.33	0.134	0.0205
	2.43	0.0766	0.0119
Lead	0.635	0.0701	0.0103
	1.27	0.0380	0.00288
	2.57	0.0156	0.000391
	5.08	0.00429	0.00023
	10.16	0.000467	0.0000001
X-ray Glass	0.645	0.135	0.0251
	1.30	0.0841	0.0144
	2.60	0.0463	0.00534
Lead Apron	0.16	0.306	0.0346
Safety Glass	1.30	1.94	0.109
	2.60	1.50	0.0886

[1] Adapted from NUREG/CR-5550 (Reilly et al., 1991)

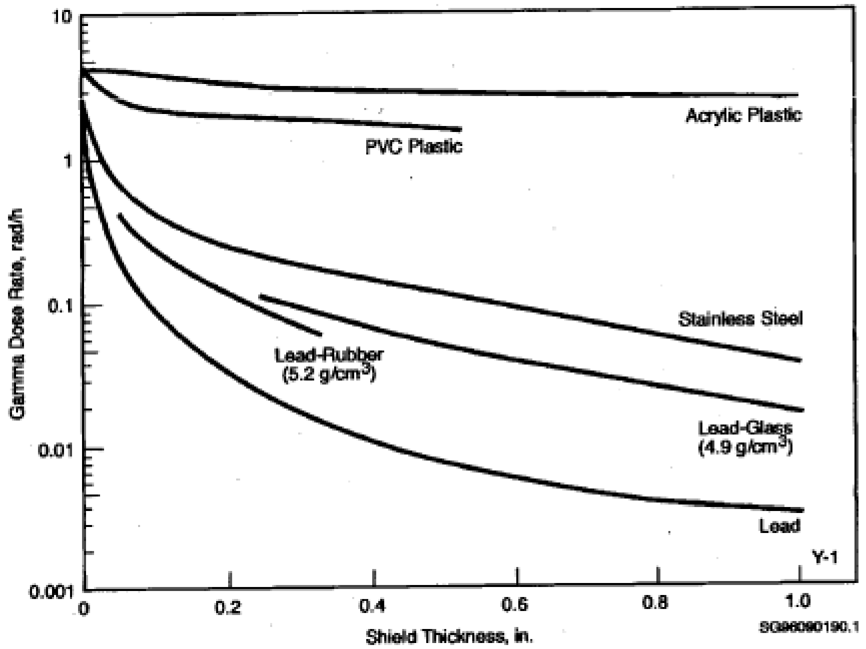

Figure 6.6. Reduction in Photon Dose Rate with Various Shielding Materials at a Distance of 3 cm from a 100-gram Disk of Plutonium Oxide

Table 6.11. Isotopic Composition of Plutonium Sources at 5 years After Chemical Separation of the Plutonium

	Weight-Percent of Isotope	
	Low-exposure Pu	High-exposure Pu
^{235}Pu	0.001	1.85
^{239}Pu	93.5	63.3
^{240}Pu	5.99	19.2
^{241}Pu	0.397	9.27
^{242}Pu	0.001	3.88
^{241}Am	0.103	2.40

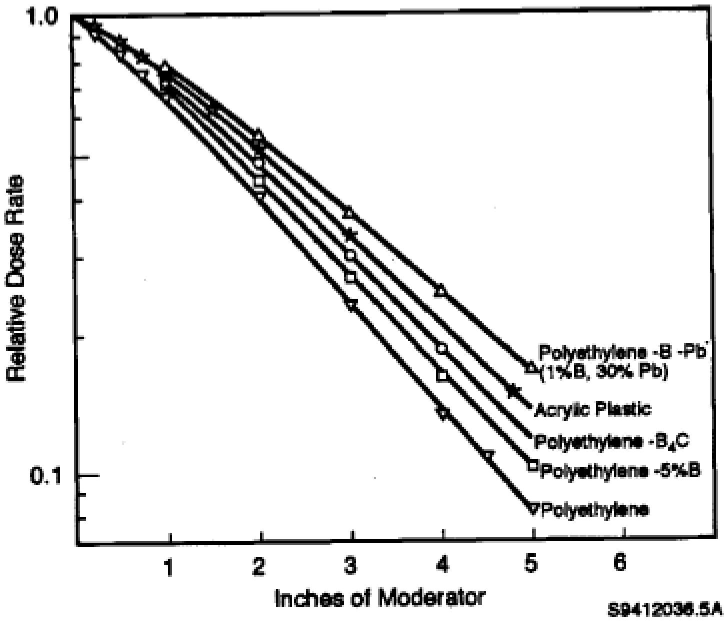

Figure 6.7. Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Tetrafluoride Sources

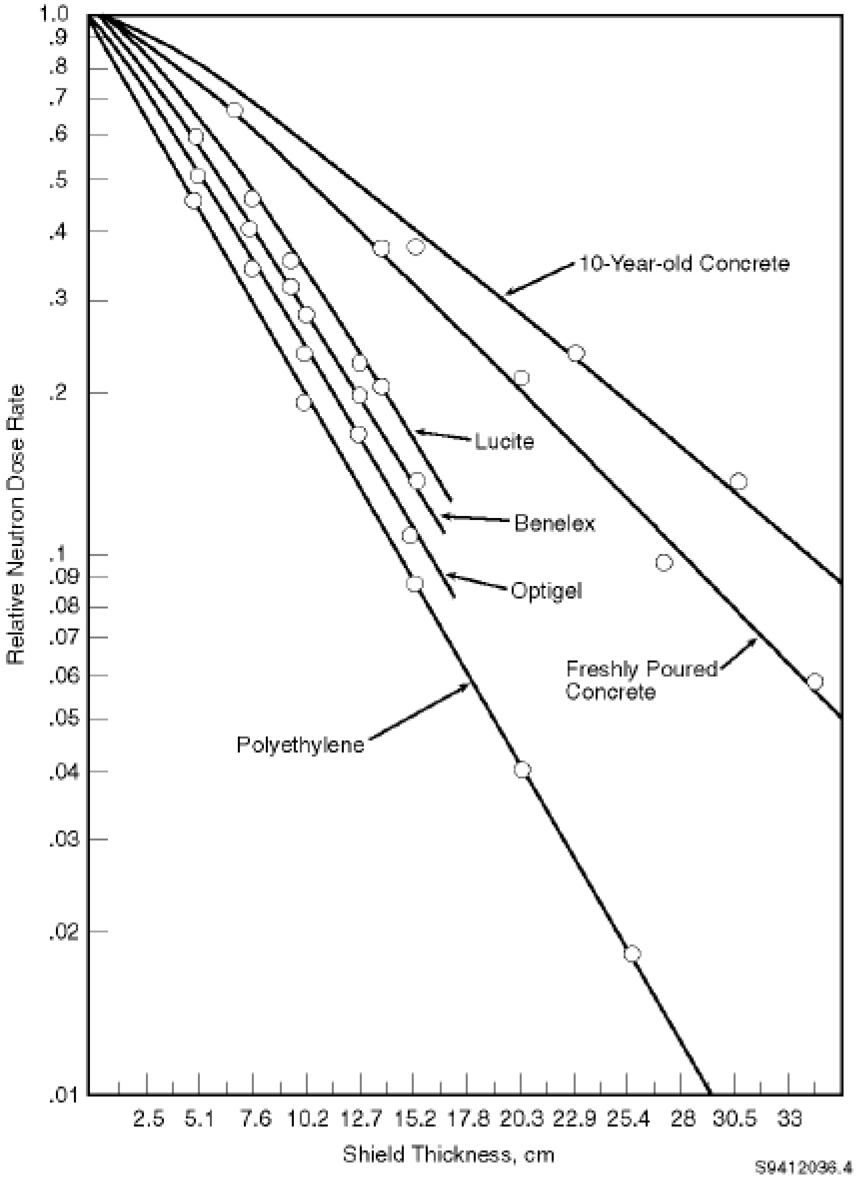

Figure 6.8. Reduction in Neutron Dose Equivalent Rate for Various Slab Shields for Plutonium Oxide Sources

7.0 NUCLEAR CRITICALITY SAFETY

This chapter will emphasize present-day criticality concerns from the standpoint of what health physics personnel need to know to ensure that the DOE mission is accomplished in a safe and cost effective manner. It provides an overview of the administrative and technical elements of current nuclear criticality safety programs. It does not provide a definitive discourse on nuclear criticality safety principles or repeat existing guidance. For health physics personnel who require a greater understanding of nuclear criticality safety, the references contained here provide a source of such detailed requirements and information.

DOE Policy 420.1, Department of Energy Nuclear Safety Policy, documents DOE's nuclear safety policy (DOE, 2011f).

Nuclear criticality safety issues at DOE facilities historically have been concerned with manufacturing plutonium, processing plutonium into weapon components, and storing weapon components and weapons in safe arrays. With DOE's newly identified mission of concluding much of the plutonium production and decommissioning of production reactors and processing facilities, today's nuclear criticality safety concerns have changed. While the historic nuclear criticality safety issues remain with the storage of weapons and associated components, current concerns include the disassembly of weapons, processing, and disposition of unique plutonium materials (commonly referred to as “legacy materials”), and decommissioning of production reactors and processing facilities.

Radiation protection personnel should understand nuclear criticality principles and the impact of these principles on radiological conditions that result from the processing, handling, and storage of fissionable materials. Radiation protection personnel provide an additional knowledgeable resource to help recognize workplace situations that might lead to the violation of a nuclear criticality control parameter that could contribute to an inadvertent nuclear criticality event. There have been occasions in the history of the nuclear industry when radiation protection personnel have observed and stopped unsafe actions by facility personnel that, if allowed to continue, might have resulted in an inadvertent nuclear criticality. Radiation protection personnel shall also be aware of the potential impact of actions that may be routine for normal radiation protection practice, but which could result in the violation of a nuclear criticality control parameter. Finally, radiation protection personnel provide significant support in emergency response actions should an inadvertent nuclear criticality occur. These actions include use of emergency instrumentation, accident dosimetry, radiological dose assessment, and recovery.

This chapter reviews (1) nuclear criticality safety regulations and standards applicable to DOE facilities, (2) criticality control factors, (3) past criticality accidents and associated lessons learned, (4) roles, responsibilities, and authorities of health physics staff with regard to nuclear criticality safety, (5) the content of an acceptable nuclear criticality safety program, and (6) a summary of the criticality safety issues identified in DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

7.1 REGULATIONS AND STANDARDS

Nuclear criticality safety program requirements for DOE facilities are presented in DOE Order 420.1C (DOE, 2012b). The order also addresses nuclear safety design criteria, fire protection, natural phenomena hazards mitigation and the cognizant system engineer program.

DOE Order 420.1C requires that the criticality safety program describe how the contractor will satisfy the requirements of the ANSI/ANS-8 consensus nuclear criticality safety standards in effect as of the date of DOE Order 420.1C, including why any recommendation in applicable ANSI/ANS-8 standard is not implemented.

7.2 CRITICALITY CONTROL FACTORS

For a criticality accident to occur, there has to be a critical mass of fissionable material. The critical mass is a function of the radionuclides in the material as well as its density, chemical and physical form, shape, and surroundings (i.e., moderators, reflectors, neutron absorbers). Nuclear criticality safety is achieved through the control over both the quantity and distribution of fissile materials and other materials capable of sustaining a chain reaction as well as the control of the quantities, distributions, and nuclear properties of all other materials with which fissile materials are associated. For new facilities, DOE requires that design considerations for the establishing controls should be mass, density, geometry, moderation, reflection, interaction, material types, and nuclear poisons (neutron absorbers). The use of administrative controls should be minimized (DOE, 2002).

Nuclear criticality control factors can be classified as technical (e.g., geometry controls and mass limitation controls) or administrative (e.g., operating procedures).

7.2.1 Technical Control Factors

Plutonium isotopes include ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. Although a tremendous amount of criticality safety work centered around uranium and its manufacture at Oak Ridge. This standard addresses plutonium facilities. All these radionuclides are fissionable materials; however, ^{239}Pu and ^{241}Pu are referred to as fissile materials, a subset of fissionable materials. Fissile materials are capable of sustaining a neutron chain reaction with thermal neutrons and fast neutrons and, as such, have lower critical masses than other plutonium isotopes.

Single-parameter limits for plutonium solutions, oxides, and metals are presented in ANSI/ANS-8.15 (ANSI, 1981) and are summarized in Table 7.1. A single-parameter limit means that if any one of the parameters for a given material is maintained less than its limit, then a criticality event is impossible. For example, for a $^{239}\mathrm{Pu(NO_3)_4}$ solution, as long as the ^{239}Pu mass in the solution is less than 0.48 kg, the other parameters can exceed their limits (e.g., the solution concentration could be greater than 7.3 g/L) and a criticality incident is not possible.

For plutonium solutions and metals in an isolated system, use of favorable geometry is the preferred method of criticality control. An isolated system is far enough removed from other systems such that neutron leakage from a nearby system will not contribute to the likelihood of a criticality excursion. Where geometry control is not feasible, the preferred order of controls is (1) other passive engineering controls (e.g., mass control), (2) active engineering controls, and (3) administrative controls

Other technical control factors used to control nuclear criticality risks include density controls, spacing controls (sometimes referred to as interaction), neutron absorbers, moderation controls, and neutron reflection. Spacing controls become particularly important in the storage and transport of fissionable materials.

7.2.2 Double Contingency

The concept of double contingency in nuclear criticality safety applies technical control parameters to ensure nuclear criticality safety. The following table identifies some single parameters.

Table 7.1. Subcritical, Single Parameter Limits for Plutonium Solutions and Metals (AMSI, 1983b)

	Plutonium Solutions and Metals			
Parameter	$^{239}Pu(NO_3)_4$	Metallic ^{239}Pu	$^{239}PuO_2$[(a)]	$^{239}PuO_2$[(b)]
Mass of fissionable nuclide, kg	0.48	5.0	10.2	27
Diameter of cylinder of Solution, cm	15.4	-	-	-
Volume of solution, L	7.3	-	-	-
Concentration of fissionable Nuclide, g/L	7.3	-	-	-
Cylinder, diameter, cm	-	4.4	7.2	12.6
Slab thickness, cm	5.5	0.65	1.4	2.8
Maximum density for which Mass and dimension limits are valid, g/cm^3	-	19.82	9.92	-

(a) Oxides containing no more than 1.5% water by weight at full density.
(b) Oxides containing no more than 1.5% water by weight at no more than half density.

Double contingency requires that process designs incorporate sufficient factors of safety to require at least two unlikely, independent, and concurrent changes in process conditions before a nuclear criticality accident is possible.

Protection, or defense in depth, should be provided by either (a) the control of two independent process parameters (which is the preferred approach, if practical) or (b) a system of multiple controls on a single parameter. The basis for selecting either approach should be fully documented.

The two parameters that are controlled in the double contingency analysis process shall not be related by common mode failures. Judgment is required in determining whether two events are related and, consequently, whether they represent two contingencies or a single contingency. For example, exceeding a storage limit and then flooding an area with water would constitute two independent events. However, a fire followed by the flooding of a storage area with fire suppression water would constitute a single event.

The double contingency principle should be applied to all nuclear criticality safety analyses for processes, systems and equipment, storage, and transportation of fissile materials. Should contingencies be determined to be related, efforts are to be made to separate the contingencies.

7.2.3 Administrative Control Factors

Administrative control factors are the combination of personnel, programs, plans, procedures, training, audits and reviews, and quality assurance practices which are used to administer a nuclear criticality safety program. Administrative controls are used in addition to engineered controls or design features to ensure nuclear criticality safety of facility operations. Administrative control factors are outlined in ANSI/ANS-8.19 (ANSI, 2005). An effective nuclear criticality safety program requires a joint effort by managers, supervisors, plutonium workers, and nuclear criticality safety staff and relies upon conformance with operating procedures by all involved personnel. The following sections describe the key requirements of a nuclear criticality safety program from ANSI/ANS-8.19.

7.2.3.1 Nuclear Criticality Safety Program

Management shall develop a nuclear criticality safety policy and ensure that it is distributed to fissile material workers. They also delegate authority to implement the policy, monitor the nuclear criticality safety program, and periodically participate in audits of the program. Supervisory staff shall ensure that nuclear criticality safety procedures are written and that staff is trained in those procedures. The nuclear criticality safety staff shall provide technical guidance for the design of equipment and processes and for the development of operating procedures. A nuclear criticality safety evaluation shall be performed by the nuclear criticality safety staff before starting a new operation with fissile materials or before an existing operation is changed. An independent evaluation of the technical adequacy of the nuclear criticality safety program shall also be performed periodically.

7.2.3.2 Nuclear Criticality Safety Organization

Like the radiation protection program, the nuclear criticality safety organization should have a reporting line to the highest level of facility management independent of operations. The nuclear criticality safety organization shall have the responsibilities and authorities of its staff clearly delineated and communicated to the other facility personnel. Lines of interaction and interfaces with other facility organizational components should be clearly defined, both organizationally and procedurally. In any case, the responsibility for nuclear criticality safety should be assigned in a manner that is compatible and consistent with the other safety disciplines. The organization should also contain an independent nuclear criticality safety review committee and have access to consultants to assist in the conduct of the criticality safety program.

7.2.3.3 Plans and Procedures

Facility nuclear criticality safety plans and procedures are important components of the overall facility operation. These documents provide the means by which the program is conducted and prescribe how nuclear criticality safety is to be achieved. These plans and procedures identify how both the administrative activities are to occur and how the technical aspects of nuclear criticality safety analysis are conducted. The purpose of procedures is to facilitate the safe and efficient conduct of operations. The processes of procedure development, review, training, and approval have sufficient controls to ensure that nuclear criticality concerns are properly addressed. These controls include the periodic review and reaffirmation of these procedures, ensuring that procedure deviations are properly investigated and reported to facility management and, if appropriate, to DOE. The controls should also mitigate the possibility of such deviations recurring.

Procedures should exist that address the determination and posting of nuclear criticality safety parameters. These procedures should include a description of how the limits are to be determined and how workstations are to be posted as to form, geometry controls, mass limits, moderator limits, etc.

Inspections and audits are performed to assess the success of the nuclear criticality safety program. The audits shall be performed by qualified individuals who are independent of the operation. They are conducted to verify that operating procedures and other safety standards are being followed and to identify any weaknesses in the nuclear safety program. Deficiencies identified in these inspections and audits shall be formally addressed, tracked, reported, and resolved.

ANSI/ANS-8.20 (ANSI, 1991) provides guidance for development of nuclear criticality safety training plans and procedures for personnel working with or near fissile materials. This program and its associated procedures should describe the program, training requirements, recordkeeping, content, responsibilities, and objectives of a facility nuclear criticality safety program.

7.3 CRITICALITY ACCIDENT EXPERIENCE

Criticality accidents, sometimes called criticality excursions, can either be short-duration pulse-type excursions or continuous excursions. In the history of plutonium handling and processing, there have been five criticality accidents involving plutonium materials. Three of the accidents occurred during research activities and the other two accidents during plutonium-processing operations. The two processing accidents are reviewed in this section.

7.3.1 Types of Criticality Accidents

In a pulse-type criticality accident, there is an initial pulse of typically 10^{15}-10^{17} fissions over a short time-period (less than 1 sec), sometimes following by additional lower-intensity pulses. In a fissionable material solution, the pulse or spike is terminated by the heating and consequent thermal expansion of the solution and by bubble formation that serves to reconfigure the fissile mass into a noncritical configuration (Paxton, 1966). If the initial pulse results in a loss of solution from the container (e.g., by splashing) or redistribution of material, the criticality event may conclude without further pulses. However, if there is no loss of material as the solution cools, it may form a criticality mass once again and pulse with slightly lower fission yield (Paxton, 1966).

Criticality accidents can result in lethal doses of neutron and gamma radiation at considerable distances from the accident site (on the order of tens of meters). There can also be high beta-gamma residual radiation levels from fission products after the excursion is concluded. The heat generated during the excursion can melt parts of the system that held the fissionable material (Moe, 1988).

Moe (1988) reviewed estimations of prompt radiation doses from excursions in a moderated system and a metallic system, as well as dose rates from residual contamination left by a criticality excursion. Assuming a burst of 10^{18} fissions in an unshielded, water-moderated system, the total absorbed dose is estimated to be >600 rad up to 6 m and >100 rad up to about 15 m. The gamma/neutron ratio of the total absorbed dose was 2.8. For an unmoderated configuration, the gamma/neutron absorbed dose ratio was 0.1. In general, for a moderated system, the gamma dose would be expected to be higher than the neutron dose and, for a metal system, the neutron dose would be expected to be higher than the gamma dose.

Moe (1988) noted that for an excursion of $>10^{18}$ fissions, dispersion of the fissile material and the fission products would occur, resulting in heavy local contamination and a subsequent high residual dose rate. This dose rate was estimated at >1000 rad/h at 100 ft shortly after the burst and >10 rad/h at 30 ft an hour after the burst. This is the basis for instructing workers to immediately vacate the work area when the criticality alarm is sounded. Seconds can save significant dose, if not from the excursion itself, then from any residual radiation that is in the area.

7.3.2 Summary of Past Criticality Accidents

Stratton summarized five criticality accidents involving plutonium prior to 1967 (Stratton, 1967). Three of the accidents involved plutonium in solutions, with the other two involving metallic forms. Three of the accidents involved early research activities and the other two were plutonium-processing accidents. Summaries of these two accidents follow as derived from Stratton (1967) or Paxton (1966). No criticality accidents have occurred regarding mechanical processing, storage of plutonium materials, or transportation of plutonium materials.

7.3.2.1 Los Alamos Accident - December 30, 1958

A nuclear criticality accident occurred on December 30, 1958, at the Los Alamos Scientific Laboratory, killing one worker and overexposing two other workers. The criticality occurred in a 225-gal, 38-in.-diameter stainless steel tank, with a thick organic layer containing 3.27-kg plutonium floating on a dilute aqueous solution of 60-g plutonium in 330 L. The tank was cylindrical and water-reflected. The tank contents were stirred, mixing the contents into a critical configuration. Microbubbles, thermal expansion, and continued mixing of the tank eliminated the critical configuration. The excursion consisted of a single pulse of 1.5 x 10^{17} fissions. The operator near the tank received a lethal dose of 12,000 rem (±50%), while two workers who assisted the operator received doses of 134 rem and 53 rem. The tank was supposed to have only 0.125 kg of plutonium; however, a gradual accumulation of solids during the 7.5-year operating history of the plant resulted in 3.27-kg plutonium in the tank.

7.3.2.2 Hanford-Recuplex Plant Accident - April 7, 1962

On April 7, 1962, a criticality accident occurred at a multipurpose plutonium-recovery operation at the Recuplex Plant, Hanford, Washington. During a clean-up operation, about 46 L of solution containing 1400- to 1500-g plutonium was directed into a 69-L glass transfer tank that led to the criticality accident. The tank was 93% full and unreflected. Solutions in the tank generally contain only a fraction of a gram per liter; however, in this situation apparently the solution was drawn from a sump through a temporary line that was being used for cleanup. The excursion had an initial pulse of about 10^{16} fissions. Following this spike, the tank was fissioning for 37.5 hours with the power level steadily decreasing (Stratton, 1967). The total yield of the accident was about 8.2 x 10^{16} fissions distributed over a 37-hour time period with about 20% in the first half-hour. Three workers in the vicinity of the tank during the initial spike received doses greater than regulatory limits. One worker about 5 to 6 ft from the tank received 110 rem, another approximately 9 ft away received about 43 rem, and the final worker about 26 ft away received about 19 rem.

7.4 CRITICALITY ALARMS AND NUCLEAR ACCIDENT DOSIMETRY

Requirements for criticality alarm systems and nuclear accident dosimetry are presented in this section. Criticality alarm systems provide rapid warning to individuals in the immediate accident location and nearby locations to evacuate to a predesignated assembly location. Specifications for the criticality alarm system are found in DOE Order 420.1C (DOE, 2012b) and ANSI/ANS-8.3 (ANSI, 1997c). Key requirements that may be of interest for the health physics staff are summarized in Section 7.4.1. Paxton (1966) noted that lives have been saved in past criticality accidents by radiation alarms coupled with effective evacuation procedures. Nuclear accident dosimetry, discussed in Section 7.4.2, provides the means of determining the dose to workers in the vicinity of the excursion.

7.4.1 Criticality Alarm System

The nuclear criticality safety program evaluation and documentation should include an assessment of the need for criticality accident detection devices and alarm systems, and installation of such equipment where total risk to personnel will be reduced.

Per the criticality safety program, the basic elements and control parameters of programs for nuclear criticality safety shall satisfy the requirements of the applicable American Nuclear Society's ANSI/ANS nuclear criticality safety standards:

As specified in ANSI/ANS-8.3, the need for criticality alarm systems shall be evaluated for all activities in which the inventory of fissionable material in individual unrelated work areas exceeds 700 g of ^{235}U, 520 g of ^{233}U, 450 g of ^{239}Pu, or 450 g of any combination of these three isotopes.

ANSI/ANS-8.3 provides several additional requirements regarding criticality alarm systems. The alarm signal shall be for immediate evacuation purposes only and of sufficient volume and coverage to be heard in all areas that are to be evacuated. Information on sound levels of the alarm can be found in ANSI/ANS-8.3. The alarm trip point shall be set low enough to detect the minimum accident of concern. The minimum accident of concern may be assumed to deliver the equivalent of an absorbed dose in free air of 20 rad at a distance of 2 m from the reacting material within 60 sec. The alarm signal shall activate promptly (i.e., within 0.5 sec) when the dose rate at the detectors equals or exceeds a value equivalent to 20 rad/min at 2 m from the reacting material. A visible or audible warning signal shall be provided at a normally occupied location to indicate system malfunction or loss of primary power. Each alarm system should be tested at least once every three months. An evacuation drill shall be conducted at least annually.

Criticality alarm systems may consist of one to several detectors per unit. In multi-detector units (e.g., three detectors), at least two detectors shall be at the alarm level before initiating the alarm; in redundant systems, failure of any single channel shall be into the trip state (ANSI, 1997c).

7.4.2 Nuclear Accident Dosimetry

Nuclear criticality safety program evaluation and documentation should include:

Assessment of the need for criticality accident detection devices and installation of such equipment where total risk to personnel will be reduced.

Per the criticality safety program, nuclear accident dosimetry is required when the fissionable material mass exceeds the ANSI/ANS-8.3 limits discussed in Section 7.4.1 and the probability of criticality is greater than 10^{-6} per year.

Requirements for nuclear accident dosimetry programs at DOE facilities are found in 10 CFR 835.1304 (DOE, 2007a). A nuclear accident dosimetry program shall include the following:

-- A method to conduct initial screening of personnel involved in a nuclear accident to determine whether significant exposures to radiation occurred;

-- methods and equipment for analysis of biological materials;

-- a system of fixed nuclear accident dosimeter units (sometimes referred to as area dosimeters); and

-- personnel nuclear accident dosimeters (PNADs) worn by all individuals who enter locations with specified quantities of fissile material.

Additional desirable features of a nuclear accident dosimetry program include:

-- Facilities to evaluate fixed dosimeters and/or PNADs;

-- a method to determine the approximate neutron spectrum;

-- a method to determine the activity of ^{24}Na in blood and ^{32}P in hair, such as a calibration coefficient determination for a common site survey instruments (such as a pancake GM or a NaI scalar counter) to count in an individual's armpit or other similar gross assay estimation techniques; and

-- a method to correct dosimeter results for actual spectrum (if known).

7.4.2.1 Initial Screening Evaluation

A nuclear accident dosimetry program should provide absorbed dose information within 24 hours after the incident. A nuclear accident dosimetry program shall include a method to conduct initial screening of personnel involved in a nuclear accident to determine whether significant exposures to radiation have occurred (10 CFR 835.1304)[also see ANSI N13.3 (ANSI, 1969)]. Discussions on initial screening evaluations to segregate exposed from unexposed individuals (sometimes referred to as “quick sort techniques”) are found in several references (Moe, 1988;

Delafield, 1988; Petersen and Langham, 1966; Hankins, 1979; Swaja and Oyan, 1987).

A common initial screening method is to provide all workers in areas requiring nuclear accident dosimetry with an indium foil in their personnel dosimeter or security badge. During a criticality excursion the foil will become activated by neutrons per the ^{115}In (n, gamma) ^{116m}In reaction and can be measured with a portable beta-gamma survey instrument or ion chamber. The ^{116m}In has a 54-min half-life and releases a 1-MeV beta (maximum energy) and a 1.3-MeV gamma (80% of the time).

An alternate screening is to measure body activity due to neutron activation of the sodium in the blood via the ^{23}Na (n, gamma) ^{24}Na reaction. Sodium-24 has 15-hour half-life and releases a 1.4-MeV beta (maximum energy) and two gammas (1.37 MeV and 2.75 MeV). A beta-gamma survey meter is used to measure the ^{24}Na activity in the blood by placing the detector probe against the individual's abdomen and having the individual bend forward to enclose the detector (Moe, 1988). Alternatively, the probe can be positioned under the armpit with the open window facing the chest area. Moe (1988) noted that this method is less sensitive than the use of indium foils and even a small reading can indicate a significant exposure. An approximate equation to calculate worker dose (D) based on body weight and instrument reading is shown in Equation 7.1:

$$D\,(\text{Gy}) = \frac{80\,(\text{instrument reading in mR / h})}{\text{Body weight (lb)}} \qquad (7.1)$$

Differences in incident neutron energy spectrum, orientation, and measurement techniques relative to conditions used to develop activity-dose correlations can cause significant errors in estimated radiation dose based on quick-sort surveys. Swaja and Oyan (1987) showed that radiation doses estimated from induced body activity can vary by a factor of about 2 due to neutron energy spectrum or orientation effects and by as much as 30% due to probe position. Doses based on indium foil activity can vary by a factor of about 9 due to neutron energy spectrum effects, a factor of 3 depending on foil orientation relative to the incident field, and a factor of about 2 due to probe window setting. Swaja and Oyan (1987) recommended that those count rates above background during quick-sort techniques should be initially interpreted only as an indication that the person has been exposed.

7.4.2.2 Fixed and Personnel Nuclear Accident Dosimeters

A comprehensive nuclear criticality dosimetry system should consist of stationary (fixed-location, area) dosimeters, neutron and gamma dosimeters worn by personnel (i.e., PNADs), and specialized laboratory equipment to evaluate the dosimeters.

Fixed nuclear accident dosimeter units should be capable of determining neutron doses in the range of 10 rad to 10,000 rad with an accuracy of ±25%. They should also be capable of providing the approximate neutron spectrum to permit the conversion of rad to rem. The gamma-measuring component of the dosimeter should be capable of measuring doses in the range of 10 rem to 10,000 rem in the presence of neutrons with an accuracy of about ±20%. The number of fixed dosimeter units needed and their placement will depend on the nature of the operation, structural design of the facility, and accessibility of areas to personnel. Generally, dosimeters should be placed such that there is as little intervening shielding and as few obstructions as possible (ANSI, 1969). The number and placement of dosimeters should be periodically reverified to reflect changes in building design and operations. Ease of dosimeter recovery after a criticality event should be considered in their placement, including the possible need for remote retrieval.

10 CFR 835.1304 requires that PNADs be worn by all individuals who enter a controlled area with specified quantities of fissile material. The PNADs should be capable of determining gamma dose from 10 rad to 1000 rad with an accuracy of ±20% and neutron dose from 1 rad to 1000 rad with an accuracy of ±30% without dependence upon fixed-unit data.

ANSI N13.3 (ANSI, 1969) provides general criteria for nuclear accident dosimeters that are reviewed below. Dosimeters, both fixed and personnel, should be protected against radioactive contamination to avoid false measurements. Periodic inventory methods should be established and audits made to ensure that the dosimeters are not removed or relocated without appropriate approvals. Techniques for estimating the effect of body orientation at the time of the exposure should also be developed.

Neutron-Measuring Component of Dosimeter. Criticality accidents create a wide range of neutron energies. Since the neutron dose per unit fluence is strongly dependent on neutron energy, knowledge of the neutron energy spectrum is important in accident dosimetry. In criticality accidents, neutrons with energies greater than about 100 keV contribute most of the dose; therefore, measurement of the fast neutron dose is of most importance. See Delafield (1988) for a review of the different types of neutron dosimeters available for accidents.

Gamma-Measuring Component of Dosimeter. Delafield (1988) noted that the ratio of the gamma rays to neutron dose will vary according to the type of critical assembly and whether or not additional shielding is present. For unshielded assemblies, the gamma-to-neutron ratio can range from 0.1 for a small heavy metal system up to about 3 for a small hydrogen-moderated solution system. A concrete or hydrogenous shielding material will increase the gamma-to-neutron ratio. Gamma dose can be determined by TLD, film, or radiophotoluminescent glass.

Dosimeter Comparison Studies. Sims and Dickson (1979) and Sims (1989) present a summary of nuclear accident dosimetry intercomparison studies performed at the Oak Ridge National Laboratory Health Physics Research Reactor. A summary (Sims, 1989) showed that of the 22 studies conducted over 21 years, 68% of the neutron dosimeter results were within the ±25% accuracy standard and 52% of the gamma dosimeter results were within the ±20% accuracy standard. Most measurements that failed to meet the accuracy standards overestimated the actual dose. Some of their other findings include the following:

- Doses from hard neutron energy spectra are more accurately measured than those from soft energy spectra
- The threshold detector unit is the most accurate type of nuclear Accident neutron dosimeter; however, its use is declining due to increasingly strict control of small quantities of fissionable materials
- Activation foils (ACT) are the most popular nuclear accident neutron dosimeter
- For gamma dosimeters, TLDs are the most popular and the least accurate, and film is the least popular and the most accurate.

7.4.2.3 Biological Indicators

Earlier in this section, a quick-sort method was described using neutron activation of sodium in the blood as an indicator of worker exposure. More sophisticated laboratory analysis of blood samples can be performed to obtain a more accurate estimate of worker dose, as discussed in Delafield (1988) and Hankins (1979). The use of neutron activation of sulfur in hair ($^{32}S(n, p)^{32}P$) is another method to estimate absorbed dose for workers involved in a criticality accident (Petersen and Langham, 1966). The orientation of the subject can also be determined by taking samples of hair from the front and back of the person. Hankins (1979) described a technique for determining neutron dose to within ±20-30% using a combination of blood and hair activations. Their evaluation was independent of the worker's orientation, of shielding provided by wall and equipment, and of neutron leakage spectra.

7.5 RESPONSIBILITIES OF HEALTH PHYSICS STAFF

The health physics staff should have a basic understanding of program structure, engineering criteria, and administrative controls as related to nuclear criticality safety as reviewed in earlier sections of this chapter. Additionally, the health physicist's responsibilities include emergency instrumentation and emergency response actions.

7.5.1 Routine Operations

During routine operations the health physics staff's responsibilities related to nuclear criticality safety include calibrating, repairing, and maintaining the neutron criticality alarm detectors and nuclear accident dosimeters, and maintaining appropriate records. The health physics staff should be knowledgeable of criticality alarm systems, including alarm design parameters, types of detectors, detector area coverage, alarm set-points, and basic control design. The staff should also be familiar with plans for emergency response.

The health physics staff should maintain an adequate monitoring capability for a nuclear criticality accident. In addition to the criticality alarm systems and the fixed nuclear accident dosimeters discussed above, remotely operated high-range gamma instruments, personal alarming dosimeters for engineering response/rescue teams, neutron-monitoring instrumentation (in case of a sustained low-power critical reaction), and an air-sampling capability for fission gases should be maintained.

Other support activities may include assisting the nuclear criticality safety engineer or operations staff in performing radiation surveys to identify residual fissionable materials remaining in process system or ventilation ducts.

7.5.2 Emergency Response Actions

The priorities of line management (which could include involving the health physics staff) during a criticality event should be to rescue personnel, prevent further incidents or exposures, and quickly determine those who have been seriously exposed (Moe, 1988). To support these emergency response actions, the health physics staff should be trained in facility emergency procedures. These emergency procedures include evacuation routes, personnel assembly areas, personnel accountability, care and treatment of injured and exposed persons, a means for immediate identification of exposed individuals, instrumentation for determining the radiation levels at the assembly area, and the re-entry and formation of response teams.

Emergency response procedures for conducting the initial quick sort of workers should specify measurement techniques and require that surveyors record methods and instrument settings used for quick-sort operations to ensure proper interpretation of the results. Field results should be compared to pre-established activity-dose relationships developed as part of emergency response procedures to determine if a worker was exposed. Other indicators such as a discharged self-reading dosimeter could also be an indication of a possible exposure.

As an immediate follow-up action for workers identified as being exposed during a quicksort procedure, a more accurate dose estimate should be made using PNADs, fixed-location accident dosimeters, or biological activity analyses (^{24}Na in the blood or ^{32}P in the hair). Part of these more accurate analyses should include: 1) better definition of source characteristics, 2) location of moderating materials, and 3) location and orientation of the person(s) at the time of exposure and action of the person following the irradiation. The health physics staff can provide valuable information to support this analysis, particularly regarding the location and orientation of workers to the excursion if they are involved in the rescue and initial monitoring procedures.

Health physics staff will be responsible for retrieving fixed nuclear accident dosimeters and ensuring that PNADs from any exposed workers are submitted for analysis.

7.5.3 Special Considerations During Decommissioning Activities

Before decommissioning or disposal of any facilities or equipment, an evaluation should be performed to assess the potential holdup of fissionable material in any equipment. These types of measurements may require the assistance of health physics staff.

Some strippable coatings and surface fixing films are good neutron moderators. Nuclear criticality safety specialists should be consulted when using these coatings to decontaminate surfaces because criticality could be a concern, depending on the geometry of the removed coating when in the disposal unit.

7.6 DEPARTMENT OF ENERGY PLUTONIUM VULNERABILITY ANALYSIS STUDY

In March 1994, Department of Energy Secretary Hazel R. O'Leary commissioned a comprehensive assessment to identify and prioritize the environment, safety and health vulnerabilities that arise from the storage of plutonium in the DOE facilities and determine which are the most dangerous and urgent. The following, provided for historic perspective, summarizes the results of that study.

Vulnerabilities identified included degradation in plutonium materials and packaging, and weakness in facilities and administrative controls that can expose workers and public, or contaminate the environment. The summary of the results presented in this section is taken from DOE/DP-0123T, Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a).

The assessment was commissioned because of recent ruptures of stored plutonium packages and the need to store safely the large amount of plutonium-bearing materials held by the DOE in its aging facilities. The ultimate goal of the assessment was to facilitate safe and stable interim storage until its final disposition, which is not expected to take place for at least 10 to 20 years. The assessment covered 166 facilities at 35 sites and employed a Working Group process. The Plutonium Working Group combined the talent of DOE federal staff, site management and operations contractors, consultants and stakeholders. The Working Group developed plans and technical approaches for the assessment and evaluated the assessment results. Overall, this assessment took more than six months and 80,000 person-hours.

During the assessment, the DOE discussed information about vulnerabilities with stakeholders. About 45 stakeholder groups were involved in either the Working Group meetings or local activities associated with site assessments.

Excluding the classified mass of plutonium contained in nuclear weapon pits at the Pantex Plant in Texas, these sites held 26 metric tons of plutonium. Most of this was located in Rocky Flats, Colorado; Hanford, Washington; Argonne-West, Idaho; Los Alamos, New Mexico; and Savannah River Site, South Carolina. The report detailed the most significant vulnerabilities within each site and across all sites. The Working Group categorized and classified vulnerabilities based on possible effects on workers, the public or environment.

The DOE-wide assessment identified 299 environment, safety and health vulnerabilities at 13 sites, consisting of 91 material/packaging vulnerabilities, 140 facility condition vulnerabilities and 68 institutional vulnerabilities.

In general, the vulnerabilities identified in this assessment posed the greatest hazards to workers. Packaging, which the Working Group found to be widely deficient for long term storage, was often the only barrier that separates the workers from the plutonium. Plutonium solutions were the form most difficult to store and present unique hazards. Plutonium scrap and residue forms are reactive, and some are corrosive enough to degrade containers. Plutonium metals and oxides generally present fewer problems, but much of this material is stored in plastic, which can react with plutonium and cause container failure.

Facility conditions that cause vulnerabilities include aging safety systems, holdup of plutonium in process systems, and design problems that weaken the ability to mitigate accidents like fires or earthquakes. In addition to their impact on workers, such large-scale events have the potential to release plutonium that could affect the public and environment. Institutional vulnerabilities involve incomplete safety analyses, loss of experienced staff, and operational problems such as a backlog of maintenance items on systems that are important to safety.

The assessment found Rocky Flats Buildings 771 and 776 were the most vulnerable facilities, based on combinations of their vulnerabilities and amount of plutonium they hold. These buildings were more than 35 years old and had design deficiencies. The next group of most vulnerable facilities were the Savannah River Site's Building 235-F, FB-Line and Old HB-Line; Hanford's Plutonium Finishing Plant; and Rocky Flats Building 779, 707, and 371. The material in these facilities included plutonium solutions and reactive materials.

This assessment provided the information base to improve the Department's plan for safely managing the future disposition of its plutonium. While most vulnerabilities were already known, this assessment improved DOE's understanding of the issues. It has also enabled the Department to document vulnerabilities, identify new ones and set priorities which will establish a systematic approach to corrective action. DOE began formulating corrective action plans to achieve safe and stable interim storage in September 1994.

The assessment reached several conclusions. Plutonium package failures and facility degradation will increase in the future unless problems are addressed in an aggressive manner. The Department needs a strong, centrally coordinated program to achieve safe interim storage of plutonium. Priority shall be given to plutonium solutions, chemically reactive scrap/residues and packaging with plastics or other organic compounds. Much of the Department's plutonium inventory, including plutonium in holdup, shall be better characterized and site-specific programs shall be implemented to establish package design lives. Management priorities at some site should be reassessed to provide proper attention to those facilities identified as most vulnerable by this assessment. Sites shall evaluate institutional vulnerabilities such as the loss of qualified staff, and compensate for them. Standards or guidelines for packaging, storage and surveillance of plutonium scrap/residues and solutions shall be developed and implemented.

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8.0 WASTE MANAGEMENT

A material is a waste once there is no identified use or recycle value for it. Normally, wastes are considered by their physical form as either solids, liquids, or gasses, except that containerized liquids are considered solid waste under some of the current regulations. Although these forms are each processed differently, there are interrelationships. For example, it may be possible to reduce solid waste by replacing disposable protective clothing with reusable clothing that shall be laundered. The laundry will produce liquid waste. In treating liquid waste, solids may be generated, for example, filters or ion exchange resins. By careful engineering, waste generation, and treatment alternatives, a site can minimize the total waste volume and elect to generate types of waste that can be disposed of. The following sections address potentially contaminated waste and waste terminology and handling of airborne waste, solid waste, and liquid waste. The treatment of excess materials to reclaim plutonium is not a waste treatment process and is not discussed here. Refer to DOE O 435.1. Ch. 1, Radioactive Waste Management, for requirements to ensure that radioactive waste is managed in a manner that is protective of worker and public health and safety, and the environment (DOE, 1999).

8.1 POTENTIALLY CONTAMINATED WASTES

This section discusses the generation, processing, storage, and disposal of wastes in plutonium facilities. It is divided by waste types, treatability groups, and waste disposal.

8.1.1 Waste Types

In addition to the classification of waste by physical form, regulatory definitions determine how waste can be disposed. The Secretary of Energy Notice 37-92, “Waste Minimization Crosscut Plan Implementation” (SEN, 1992), requires annual reports of waste generation by type, waste stream, site, and program. The waste classifications used in the DOE Annual Reports are defined in Table 8.1.

A plutonium facility may generate any of these types of waste, except that high-level waste (HLW) will be generated only from irradiated reactor fuel. Any waste containing at least 100 nCi/g of transuranics (TRU), including plutonium, will be classified as TRU or TRU mixed waste. Waste containing detectable quantities of radioactive materials but less than 100 nCi/g of transuranics will be low-level waste (LLW).

The distinction between sanitary waste and very low-level radioactive waste can be technically a difficult one. Sometimes, material is designated LLW waste because the conditions of use could have resulted in contamination that would be difficult to detect. Techniques and limitations for doing this are discussed below with reference to solid waste.

	Table 8.1. Waste Types[(a)]
HLW	High-level waste (HLW) is the material that remains following the reprocessing of spent nuclear fuel and irradiated targets from reactors. The HLW is highly radioactive and generates heat on its own. Some of its elements will remain radioactive for thousands of years. Because of this, HLW shall be managed very carefully and all handling shall be performed from behind heavy protective shielding.
LLW	Low-level waste (LLW) is any radioactive waste that is not HLW, spent nuclear fuel, TRU waste, or uranium mill tailings. The LLW is typically contaminated with small amounts of radioactivity dispensed in large amounts of material. The LLW is generated in every process involving radioactive materials in the DOE including decontamination and decommissioning projects.
MW	Mixed waste (MW) is waste that contains both radioactive and hazardous wastes. Any of the types of radioactive waste described can be a mixed waste if it contains any hazardous wastes. In fact, all of DOE's HLW is mixed waste because of the chemicals used to reprocess the fuel that resulted in the generation of the material or because it is suspected to contain hazardous materials.
TRU	Transuranic (TRU) waste refers to waste materials containing elements with atomic numbers greater than 92. These elements are generally alpha-emitting radionuclides that decay slowly. The TRU waste contains a concentration of these elements greater than 100 nCi/g. The TRU waste is not as intensely radioactive as HLW. The TRU waste also decays slowly, requiring long-term isolation.
Sanitary Waste	Sanitary waste is waste that is neither hazardous nor radioactive.
Hazardous Waste	Because of its quantity, concentration, and physical, chemical, or infectious characteristics, hazardous waste may cause or significantly contribute to an increase in mortality, or an increase in serious irreversible or incapacitating reversible illness; it may pose a potential hazard to human health or the environment when improperly treated, stored, transported, disposed of, or otherwise managed.

Table 8.1. Waste Types[(a)] (continued)

RCRA (USC, 1976a) Regulated Waste	Solid waste, not specifically excluded from regulation under 40 CFR 261.4 (EPA, 1994a), or delisted by petition, that is, either a listed hazardous waste (see 40 CFR 261.30 - 261.33) or waste exhibiting hazardous characteristics.
State Regulated Waste	Any other hazardous waste not specifically regulated under TSCA or RCRA, which may be regulated by a State of Local authority. An example of such waste is used oil.
TSCA (USC, 1976b) Regulated Waste	Hazardous chemical wastes, both liquid and solid, containing more than 50 parts per million of polychlorinated byphenyls.

(a) Definitions from DOE/S-0101, *U.S. DOE Annual Report on Waste Generation and Waste Minimization Progress, 1991-1992*, February 1994 (DOE, 1994c).

8.1.2 Treatability Groups

In addition to being classified by type, as discussed above, wastes are classified by treatability group, depending on the treatment the waste receives. The common treatability groups are defined in Table 8.2. These are reported in each site's annual waste management report.

Table 8.2. Treatability Groups

LLW
Contact-handled LLW: exposure rate of 200 mR/h or less on contact Remote-handled LLW: exposure rate greater than 200 mR/h on contact
TRU
Contact-handled TRU: exposure rate of 200 mR/h or less on contact Remote-handled TRU: exposure rate greater than 200 mR/h on contact
Mixed Waste (Mixed LLW and Mixed TRU)
Treatable mixed waste has an existing treatment that will eliminate or encapsulate the hazardous constituents of the mixed waste, rendering it LLW or TRU. Treatable includes treatment of mixed waste that results in volume reduction. Non-treatable: no treatment exists

Plutonium facilities generate mostly contact-handled TRU waste (even though they may do much of the handling and processing within glove boxes for contamination control). The most common treatment will be compaction although some facilities have incinerators available.

Depending on the treatment methods available, waste streams may be tailored to be amenable to treatment. Some facilities are able to incinerate TRU waste. Facilities with this capability may need to eliminate halogenated, nitrogenated, or sulfur-containing materials to maximize incinerator acceptance and minimize hazardous effluents from the incinerator.

8.1.3 Waste Disposal

Waste classifications and treatability groups are important because they determine waste disposal options. Sites needing to characterize Pu waste for ultimate disposal at the Waste Isolation Pilot Plant (WIPP) at Carlsbad, New Mexico should do so as that waste is generated. It is generally more expensive to characterize the waste after the fact to meet the WIPP's waste acceptance criteria.

Sanitary waste is by far the least costly and easiest to dispose of. Liquid sanitary waste is disposed of in sanitary sewerage systems or septic systems. Sanitary solid waste is nearly always disposed of by landfill disposal or by incineration with landfill disposal of ash. Because sanitary waste disposal facilities still face various siting and permitting requirements, it is desirable to minimize waste volumes.

Hazardous waste is second in ease of disposal for most DOE facilities. Hazardous waste can be treated to eliminate the hazard only if a permit for the particular waste stream has been granted by the EPA. Hazardous waste treatments permitted in DOE facilities are usually limited to pH adjustment, precipitation, and ion exchange for liquid waste and compaction or incineration for solid waste. Combustible liquids may be incinerated either onsite or offsite, as conditions permit.

Low-level waste is still disposable at most sites. For NRC and state-licensed facilities, commercial disposal is an option, but subject to the requirements of the Low-Level Waste Policy Amendments Act (USC, 1985), which requires individual states or groups of states, called compacts, to develop local disposal facilities. In general, local facilities have not been developed, so disposal volumes are severely limited and/or significant surcharges are imposed in addition to the already high disposal cost.

Several DOE sites are currently permitted to dispose of their own low-level waste by burial. Other DOE sites have long-term storage facilities. In some cases, DOE waste is being placed in retrievable storage in the hopes that the classification of the facility can be changed and the waste allowed to remain permanently.

Mixed waste disposal facilities require all of the permitting for radioactive waste disposal facilities plus all of the permitting for hazardous waste disposal facilities. For this reason, there are very few such facilities in operation, and in general they are rather restricted in the type of waste they can accept. If possible, it is generally better to treat the waste than to destroy or chemically alter the hazardous component. In some cases, mixed waste may be treated to encapsulate the hazardous component so that it no longer has the leachability or other characteristics that cause it to exhibit hazardous properties. Mixed waste requires special permits for treatment, so it is generally preferable to avoid generating it or to treat it in connection with some other process while it is a useful material (before it becomes a waste). For example, if the hazardous component is a metal with some recycle value, or it there are recycle metals in the material, it may be best to alter the process to plate or precipitate the material as a final step in the process line, before it is declared a waste.

Most plutonium facilities will produce TRU waste or TRU mixed waste. According to national policy, DOE TRU waste is supposed to be permanently disposed of at WIPP. Volume allocations have been given to each DOE site for the waste to be placed there, so limiting the quantity of high-level TRU waste is extremely important.

Therefore, volume reduction of TRU waste is highly desirable. Incineration offers the greatest volume reduction and has the added advantage of destroying some types of hazardous constituents (flammable and other organic compounds).

High-level wastes are slated to be disposed of at a high-level waste repository. In the interim, TRU waste is being stored either at the sites that are generating it or, for some DOE facilities, at the Nevada National Security Site, until a final repository is available. Long-term maintenance of interim storage facilities and the prospect of later moves to the final disposal site and burial at that site make high-level waste very costly.

8.2 AIRBORNE WASTE

The only airborne plutonium likely to arise from either normal operations or decommissioning of DOE facilities will be in a particulate form. Although plutonium vapors are possible during cutting and perhaps some grinding operations, they will soon condense to particulate material.

8.2.1 Design Objectives

Plutonium particulates are notoriously difficult to confine and extensive use is made of glove boxes, local ventilation systems, fixatives, and other means to minimize generation of particulates and to confine them. The high-efficiency particulate air (HEPA) filter is the backbone of plutonium air-cleaning systems. Such filters are certified to have a 99.93% removal efficiency for particulates of 0.3 µm and larger and are normally used with at least two in series.

Because confinement systems are subject to component failures and other accidents, differential air pressures are normally maintained so that a breach of containment will not affect occupied areas or the environment. Glove-box lines are at the lowest pressure, plutonium laboratories at a higher pressure, and other occupied areas at the highest pressure but still negative with respect to the outside.

Because plutonium air-cleaning systems are usually expensive to service (requiring workers to be dressed in multiple layers of protective clothing and respiratory protection), and plutonium waste is expensive to dispose of, measures are taken to protect the life of plutonium air-cleaning systems. Extraneous particulates are eliminated by HEPA filtration of incoming air. (These HEPA filters may be disposed of as sanitary waste.) Roughing prefilters are used to capture the bulk of particulates and prolong the life of HEPA filters.

Care shall be taken in designing HEPA filter installations for plutonium facilities so that provisions are made to safely change the filters while maintaining contamination control. Such measures normally include redundant banks of filters (in parallel) that can be valved out for filter change, location of HEPA filter banks in enclosed rooms that are themselves HEPA-filtered, and appropriate provisions for filter bag-out.

New filters shall be tested after they are installed to ensure proper gasketing, etc. Once in place, they shall be periodically retested to ensure that HEPA efficiency is maintained. For this reason, HEPA filter installations shall have ports for the introduction of a challenge aerosol upstream of the filter and collection of a representative sample in a region of laminar flow downstream of the filter. The HEPA filters in plutonium use sometimes fail from mechanical fatigue and vibration rather than plugging or being subject to some other mechanical failure. The proper design of HEPA filtration systems and proper sampling provisions are discussed in DOE Order 6430.1A, General Design Criteria (DOE, 1989b); ANSI N510-1989, Testing of Nuclear Air-Cleaning Systems (ANSI, 2007b); ANSI/UL 586-1990, High-Efficiency Particulate Air Filter Units (ANSI, 1990); ANSI/ASME N509- 1989, Nuclear Power Plant Air Cleaning Units and Components (ANSI, 2002); Chapter 10 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a); and ANSI N13.1-2011, Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities (ANSI, 2011a).

In addition to the above features of the air-handling system, there may be process-selection features that will minimize the generation of airborne plutonium. If at all possible, plutonium compounds should be handled in sealed containers or, in the case of a metallic solid, the material encapsulated. Wet mechanical processes, such as cutting and grinding, usually generate fewer particulates than dry ones, so they are often preferred. However, it is also important to minimize the use of chemicals that will attack the air-cleaning system or contaminate the filters with hazardous chemicals, making them mixed waste. Even moisture will shorten the life of HEPA filters, so wet processes should be enclosed to the extent practicable and demisters and/or heaters used to pretreat the air from wet processes prior to HEPA filtration.

The final consideration in the design of air cleaning systems for plutonium operations is the probability and consequences of accidents. In general, plutonium air-handling systems are designed so that all probable accidents, including the failure of a single HEPA filter, do not have measurable consequences offsite. It will be necessary to design the system for all probable meteorological conditions, including (for some regions of the country) tornados. The system shall also be designed so that some improbable (but not impossible) events (accidents) have consequences that are less than catastrophic. For example, the simultaneous failure of two HEPA filters in series is highly unlikely (without a common cause such as high differential pressure from an explosion or meteorological event) but facilities shall be designed so that these events are not likely to cause fatalities offsite. The minimum performance criteria for the air-cleaning systems are dictated by DOE design criteria. Other design parameters are finalized during the Environmental Impact and Safety Analysis processes. They will differ from facility to facility.

8.2.2 Operational Controls

Plutonium air-handling systems shall be operated within the design safety envelope of the system. Beyond that, there are measures that can further reduce the potential for airborne plutonium, even in glovebox operations. Even within glove boxes, plutonium should be containerized, preferably doubly encapsulated whenever possible. Spills should be cleaned up promptly. If rags or tissues are contaminated, they should be bagged as soon as possible.

8.2.3 Waste Treatments

The principal treatment for cleaning plutonium from air is HEPA filtration. There are other technologies that can be used for pretreatment, but the most common is filtration. Electrostatic precipitation, wet scrubbing, demisters to remove moisture, and other technologies may have specific applications. (Treatment of the HEPA filters, a solid waste, and the wet scrubber effluent, a liquid waste, are discussed in Sections 8.3 and 8.4, respectively.)

8.2.4 Sampling and Monitoring

Sampling is the primary method used to achieve a complete and accurate legal record of releases after they have occurred. The primary requirement for a particulate air sample of any type is that it be representative of the stream being measured. This translates into isokinetic sampling in a laminar flow section of the exhaust duct. The parameters needed to achieve such a sample are given in numerous references such as Chapter 10 of Implementation Guide G 441.1-1C, Ch. 1(DOE, 2011a). For sampling, the analytical methods are the same as those discussed in Chapter 3 of this document for workplace sampling.

Monitoring is used to determine if current conditions are within expected parameters and to initiate corrective action if they are not. For monitoring, the system design should conform to ANSI N42.18, Specification and Performance of On-site Instrumentation for Continuously Monitoring of Radionuclides in Effluents (ANSI, 2004). The choice of the filter medium will depend on the analysis that will be done on the sample. For samples containing only plutonium particulate, a non-absorbing filter such as a membrane filter will have the highest efficiency for alpha counting. In all cases, the final count shall be done after any residual radon has decayed because it will often result in a large amount of alpha on the filter that is not plutonium. If there are other radionuclides in the waste stream that cannot be decayed in a reasonable time, either alpha spectroscopy or chemical separation shall be done. Chemical analysis shall also be done if there are stable contaminants of interest such as beryllium or heavy metals. The nature of these procedures is beyond the scope of this document.

8.2.5 Disposal

Airborne effluents are not stored. Disposal of the airborne effluent, possibly containing traces of plutonium, is generally arranged by the design of the facility and the existing air quality permits. Normally, the design of the facility is such that the method of disposal of the cleaned effluent should be unimportant during normal operation. However, the facilities are designed to minimize the impact of a filter failure or operational difficulty that results in a release. Disposal of airborne effluents is handled at the design, environmental impact assessment, and safety analysis stages of facility construction. Disposal of secondary waste from air cleaning is covered in the sections that follow.

8.3 SOLID WASTE

Solid waste will come from all phases of operation and from decommissioning of plutonium facilities. Because most plutonium solid waste will be TRU (containing more than 100 nCi/g), containerization of the waste will be done in anticipation for transportation to the WIPP, which is an expensive and detailed process. Thus, it is highly desirable to minimize the generation of solid waste in the design, operation, maintenance, and decommissioning of plutonium facilities.

8.3.1 Design Objectives

One of the principal means of minimizing solid waste is to minimize the area that becomes contaminated by plutonium and to ensure that all surfaces contaminated by plutonium are readily cleanable.

Glove boxes are often used to contain contamination and permit work in minimal protective clothing that can be reused to minimize waste volumes. By assuring that these are in isolated areas that are covered with easily cleanable materials and maintained at negative pressure with respect to the rest of the facility, waste is minimized even during minor accidents.

The choice of surface materials is extremely critical. For example, concrete floors will become impregnated by plutonium particulates or solutions and will require fixatives or scabbing to control contamination. Relatively large quantities of solid waste will be generated when facilities are decommissioned or major modifications are done. Conversely, electropolished stainless steel is easily cleaned, even to releasable levels generating only small quantities of TRU waste.

Choosing components that can be easily maintained rather than totally replaced may also be an effective strategy at minimizing waste. Whenever possible, choose equipment for which high-maintenance components can be located outside of contaminated areas. For example, many mixers, saws, and other such components have been adapted so that the motor is located outside the glove box where it can be maintained or replaced without concern for contamination status, while the working or tool end operates in a contaminated environment.

8.3.2 Operational Controls

Operational controls for waste-management purposes in plutonium facilities serve two distinct purposes: waste volume reduction (waste minimization) and waste classification control. Each of these is discussed briefly below. Operational controls to reduce the probability of accidents or minimize their consequences are also important but are not directly addressed as part of waste management.

8.3.2.1 Waste Minimization

Plutonium facilities should have a waste minimization program. The objective of a waste minimization program is the cost-effective reduction in the generation and disposal of hazardous, radioactive, and mixed waste. The preferred method is to reduce the total volume and/or toxicity of hazardous waste generated at the source, which minimizes the volume and complexity for waste disposal.

The waste minimization program applies to all present and future activities of the facilities that generate hazardous, radioactive, and/or mixed wastes. Furthermore, waste minimization is to be considered for all future programs and projects in the design stages, and should be included in all maintenance and/or construction contracts.

All managers of facilities or activities that generate hazardous, radioactive, and mixed waste are responsible for:

-- Minimizing the volume and toxicity of all radioactive, hazardous, and radioactive mixed waste generated, to the extent economically practicable

-- preparing and updating waste minimization plans for their waste-generating facilities or activities. Small waste generators in a larger facility may be grouped with others in a facility or activity plan

-- implementing the facility-specific or activity-specific waste minimization plan

-- providing input to the organization responsible for waste characterization and minimization, to support the waste minimization program

-- communicating waste minimization plans to their employees, and ensuring that employees receive appropriate training

-- ensuring that existing system/equipment replacement or modification is designed and installed to minimize generation of waste

-- developing new waste minimization strategies, and identifying cognizant staff for waste minimization communications between facility personnel

-- identifying new waste generating facilities or activities and significant process changes to existing facilities or activities to the waste characterization and waste minimization organization.

Waste volume control, or waste minimization, involves limiting the amount of material that becomes contaminated, segregating clean and contaminated material, and prolonging the useful life of equipment and material to minimize replacement. Sometimes, materials can be completely cleaned so that disposal as sanitary waste (or refurbishment in clean areas) is an option.

Program design decisions can affect TRU waste-generation. For example, the quantity of protective clothing may be a significant factor. If an incinerator is available, combustible protective clothing may be selected to have a low ash content and generate a minimum of harmful effluents such as oxides of nitrogen or halogenated compounds. In other facilities, water-washable, reusable protective clothing may minimize waste disposal.

In many nuclear facilities, contamination of packaging materials is a problem. For example, if a tool or material (e.g., a pump or some ion exchange resin) is to be used in a contaminated area, as much of the packaging material shall be removed as possible before the material enters the radiological area.

Another opportunity for waste minimization occurs when materials are used as a contingency protection against contamination. For example, strippable coatings may be applied to an area that is not expected to become contaminated or may receive only minor contamination so that it can be easily cleaned. Another example involves the disposition of disposable surgeons' gloves, which are routinely worn inside glove-box gloves. Unless there are serious contamination control problems in the facility, these can be surveyed and disposed of as sanitary waste rather than LLW or TRU waste.

If a piece of equipment is to have more than a single use in a contaminated environment, every possible measure should be taken to ensure its continued reliability rather than relying on frequent replacements. Tools should be of the highest quality and maximum flexibility consistent with the situation. For example, if a wrench is needed to maintain a piece of equipment in a glove-box, consideration should be given to future needs and storage provisions. A socket set with interchangeable sockets may ultimately create less waste than a box-end wrench of each size that is needed.

Likewise, all tools and equipment to be placed in a contaminated environment should be tested for reliability and preferably used on a clean mock-up to ensure their serviceability before they become contaminated. There is often a temptation to put the equipment into the plutonium service when it first arrives rather than test it completely first. This can result in unnecessary waste volume.

8.3.2.2 Waste Classification Control

Many operational controls involve measures to ensure that the waste generated is TRU waste rather than mixed-TRU waste, or that if it is mixed-TRU, it is of a composition that can be treated. Tight controls in the following areas are necessary to minimize mixed waste (and hazardous waste) problems: procurement of hazardous chemicals, actions of subcontractors and vendors, and training of workers. In some cases, decontamination processes have been used that result in mixed waste, such as Freon cleaning, electropolishing, and chemical decontamination. These should be used only after due consideration of the waste management consequences. In some cases, these mixed wastes can be readily treated; in other cases, their use needs to be avoided. Some new techniques are designed specifically for waste minimization and waste classification control. For example, one method involves abrasive blasting with solid carbon dioxide (dry ice), which sublimes after use and can be exhausted through a HEPA filter, leaving no added material to the waste. Decontamination with high-pressure water has some similar advantages, but care shall be taken to ensure that used decontamination solutions do not spread contamination.

8.3.3 Waste Treatments

Available treatments for solid waste include compaction and incineration. In specific cases, there may be decontamination options available, as well.

Compaction, with pressures in the range of 40,000 to 60,000 psi, is most often used on paper, fabric, and plastic although it is effective on glass, sheet metal, and some other materials. With such ordinary materials, one commercial reactor has approached up to 800 pounds of waste per 55-gallon drum, although an average of 500 pounds per drum is considered to be very good.

Compaction is done by drum compactor or box compactor. Compacting into a drum or a 4- by 4- by 8-ft box is normally a labor-intensive operation and often involves some risk of personnel exposure, even though the better compactors are equipped with HEPA-filtered ventilation systems. Supercompaction uses considerably higher pressures than compaction, normally 200,000 psi or greater. Supercompaction usually involves compacting filled waste drums into a box or overpack. Supercompaction has been success-fully used on piping and other materials that are normally considered noncompactable.

It is really a choice of words whether incineration is considered a disposal technique or a volume-reduction technique. All carbon, oxygen (except for any that becomes bound in oxide ash), nitrogen, hydrogen, and sulfur present in the incinerator feed will be converted to gasses and disposed to the atmosphere. Plutonium and most metals will remain as a solid material. As a volume-reduction technique, incineration is very successful, with volume-reduction factors up to 200:1 or greater achieved on some waste streams. There have been licensing delays for some incinerators, and often there are limitations brought about by air quality restrictions. There is also the possibility that incinerator ash may be a mixed waste due to the concentration of other impurities such as heavy metals in the waste. If a facility has an incinerator, a quantity of the feed material can be incinerated to determine if the waste will have hazardous characteristics before the material is contaminated. In some cases, it is desirable to size-reduce or repackage in combustible packaging before incineration.

Decontamination is most successful when the material can be recycled for use in a nuclear facility since the need to prove releasability (cleanliness) is eliminated. Nevertheless, cleaning material for unrestricted release is also possible in some cases. It may also be possible to decontaminate an item enough to change its classification from TRU waste to LLW, thereby allowing immediate disposal of the item, while a relatively small quantity of decontamination waste is stored as TRU waste.

Electropolishing to remove the thinnest metal surface has been very effective and produces a relatively small waste volume, especially when one of the wetted sponge units is used rather than an emersion tank. Surface scabbling has been used in decontamination of concrete, and various abrasive blasting methods have also been effective. Strippable and self-stripping coatings may be used to decontaminate surfaces, even though the primary application of strippable coatings has been in preventing contamination of surfaces.

There are occasionally mixed strategies that work well. Used HEPA filters may be removed from their frame for compaction. Metal frames may be decontaminated and wood frames may be incinerated. Whatever treatment or disposition path is chosen, insure that during waste generation, all waste is characterized, accumulated and packaged in direct readiness for final disposal or reuse.

8.3.4 Sampling and Monitoring

Solid waste is monitored for several reasons: to determine if it can be released as sanitary (or hazardous) waste; to distinguish its classification as either LLW or TRU waste, depending on the concentration of transuranic isotopes; and to obtain defensible values for documenting shipping and disposal quantities. See section 4.2.4.2 for guidance on release surveys.

8.3.5 Storage and Disposal

Solid sanitary waste, hazardous waste, and LLW can normally be disposed of using existing procedures. Transuranic waste, HLW, and most mixed waste may have to be stored for a period of time awaiting approval of disposal facilities; they will have to be stored in a manner that prevents routine and accidental impact on the environment. They shall be protected from unauthorized access, fire, flood, or water damage. Containers shall be protected from corrosion or other deterioration and an accurate inventory of the material shall be kept. Most facilities prefer to store such material in a form that they believe will be shippable. Refer to the WIPP waste acceptance criteria for venting requirements for TRU waste containers.

Existing storage and packaging requirements for plutonium metal and oxide are addressed in DOE Order 460.1C (DOE, 2011b). The DOE's existing storage practices for plutonium and plutonium-containing materials and wastes were evaluated at a DOE Workshop in May 1993 [see Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities (DOE, 1994a)]. The draft recommendations from this workshop for metals and oxides that are not in containment vessels with certified hermetic seals [per ANSI N14.5 (ANSI, 1997a)] are given in Table 8.3. In 2008 DOE published the Nuclear Material Packaging Manual, DOE M 441.1-1 (DOE, 2008e). The manual provides detailed packaging requirements for protecting workers from exposure to nuclear materials stored outside of an approved engineered contamination barrier. The variety of plutonium-containing materials is illustrated by the inventory information for the Hanford Site contained in documents by Christensen et al. (1989) and Hoyt (1993).

8.4 LIQUID WASTE

Liquid waste from plutonium facilities includes various aqueous waste streams such as cooling water, laundry waste, and floor-drain waste, and numerous organic and inorganic chemical wastes. The design criteria and operational controls to make these streams treatable and disposable, and the methods to treat them are beyond the scope of this document and are highly facility-specific. General considerations are given below.

8.4.1 Design Objectives

If a facility process requires the generation of plutonium-contaminated liquids, it is probably best to ensure that the mother liquid is demineralized water and that plutonium is the only contaminant added. In this case, the liquid can be filtered, demineralized, and recycled. Any other chemicals added to the water will complicate treatment, increase the volume of secondary waste, and diminish the opportunity for recycle. Organic contaminants such as oils, solvents, and detergents will likely foul the ion exchange resin, greatly increasing resin volume.

A pure organic solvent has many of the advantages of demineralized water, especially if it does not chemically degrade or evaporate under the conditions of use. (Solvents are not usually amenable to purification by ion exchange; however, filtration, extraction into aqueous solutions, and distillation are possible.) Unfortunately, most organic solvents are classified as hazardous materials and any material that comes in contact with them is likely to be a hazardous (or mixed) waste when it is disposed of. If the solvent is combustible and the facility includes an approved incinerator of sufficient capacity to handle the secondary waste, then the organic solvents are highly desirable.

While such guidance may be helpful in facility design, there will be waste streams that do not conform to either of the situations above. Most decontamination wastes, laundry wastes, and floor-drain wastes are examples. In decontamination, it is important that the process is selected with provisions to manage the waste. In many cases, the nature of the facility determines that the waste will be a mixed waste. In these cases, minimizing the volume is most important. For example, if a plutonium-contaminated surface has been painted with a lead-based paint, the decontamination waste will be mixed waste unless it is further treated to ensure that the lead is not in a leachable form. In this example, removing the paint by dry ice blasting, high-pressure water blasting, heat, or a similar method would be preferable to sand blasting in which the sand would be added to form an additional mixed waste that could require storage for many years.

Laundry wastes are a special problem because radioactive contamination, body oils, and odors shall be removed from protective clothing. For a time, dry cleaning was extremely popular, because the solvents were easily redistilled and recycled. However, because the solvents were usually chloroflorocarbons and because the small volume of waste generated was mixed waste, this method is now rarely used. Incineration of disposable protective clothing is an outstanding choice if an incinerator of sufficient capacity is available, but this is rarely the case. Water washing is often the method of choice. In a few cases, plutonium in the waste stream is removed adequately by filtration and the effluent can be disposed to a sanitary sewer or to the environment under a National Pollutant Discharge

Elimination System permit. It is important to select a detergent for water washing that does not foul or plug the filter and that has a minimal impact on ion exchange resins if they shall be used. Many household laundry detergents have fillers such as wood fiber to give them greater bulk. These should never be used because the fiber has no beneficial use and will end up as solid waste. As a general rule, extensive testing on clean material should be done to optimize disposal of laundry waste.

Floor-drain wastes are much more of a problem in some facilities than in others. In some facilities, there is a culture that says, “if you don't know what to do with it, pour it down the floor drain.” Such practices can lead to a mixture of water, detergent, oil, antifreeze, and other substances that clog filters and foul ion exchange resins and lead to environmental compliance issues. In the worst cases, solidification with Portland cement is the only alternative, and this increases an already large volume. The use of catch basins under chemical and lubricating systems and extensive training of personnel minimize the probability of such occurrences. Oil skimmers on floor drain collection tanks are sometimes advisable, as well.

8.4.2 Operational Controls

Once the facility is properly designed, training of personnel is the primary operational control against generating excessive volumes of waste or against generating waste with contaminants that interfere with treatment or change the classification.

Some facilities have used color codes to prevent materials from entering an area where they will adversely affect waste management. For example, certain electronic contact cleaners may be banned from some radiologically contaminated plant areas because they would generate mixed waste. The procurement organization might code all such materials red and certain areas would be posted to indicate that the materials were not allowed.

Whatever the system, it is important that each employee be trained to effectively use the system and that well-intentioned housekeeping efforts do not result in excessive waste volumes.

8.4.3 Waste Treatments

The primary treatments for aqueous waste are

- pH adjustment
- precipitation
- liquid-solid separation such as flocculation and filtration
- ion exchange
- distillation
- purification by reverse osmosis
- solidification.

The primary treatments for organic solvents are:

- Solvent extraction
- filtration
- incineration.

Virtually all of these processes (except pH adjustment) are likely to result in secondary waste that requires treatment and/or disposal. In all cases, recycling of the primary solution is desirable because it reduces monitoring cost and waste-disposal liability and cost. A brief description of the use of each of these treatment methods is given below. Extensive design and engineering should be done before any method is selected in order to ensure meeting design objectives.

8.4.3.1 pH Adjustment

This treatment is used on aqueous systems to meet discharge limitations or to make the solution amenable to other treatment. A mineral acid, such as sulfuric, hydrochloric, or nitric, is normally used to lower the pH. A base, such as sodium, potassium hydroxide, or occasionally ammonia, is used to raise the pH. The solubility of some contaminants will be affected by the pH of the solution. For example, an acidic solution containing iron may show a copious precipitate of ferric hydroxide upon the addition of a base.

8.4.3.2 Precipitation and Co-precipitation

Precipitation and co-precipitation are used to decrease the solubility of some compounds. Precipitation involves making the contaminant into an insoluble material by the adjustment of pH or the addition of a chemical. For example, nickel may be rendered insoluble by the addition of sodium dimethylglyoxime. Co-precipitation is similar but is used when the contaminant is not present in sufficient quantity to form a filterable solid but will incorporate into another precipitate as it forms or will adhere to the surface of another precipitate. In some waste treatment processes, a stable isotope of the radioactive contaminate is added to co-precipitate the radioactive material that is not present in sufficient quantity to form a precipitate on its own. Precipitation is always followed by some liquid/solid separation technique.

8.4.3.3 Liquid-Solid Separation Techniques

Treatments such as flocculation and filtration are used to remove solid and colloidal contaminates either directly from the waste stream or following a precipitation or co-precipitation process. Centrifugation or settling are sometimes used to remove gross quantities of solids preceding some filtration processes. These processes separate the waste into a concentrated and dilute waste stream, both of which will probably require further treatment. The bulk liquid fraction may be subject to filtration before recycling or disposal. The fraction with the high concentration of solids may be subject to evaporation, or drum or filter-press filtration to remove excess water, or it may be solidified as discussed below.

Where the contaminant is present as a colloid or extremely fine particulate, co-precipitation or flocculation may be required before settling, centrifugation, or filtration. Flocculation involves the addition of an extremely small quantity of a long chain molecule that has the appropriate electrostatic affinity for the contaminant present. The flocculent molecules gather the contaminant into rather large particles that are amenable to settling and filtration. The flocculent and dosage (addition ratio) are usually selected by trial and error. Flocculents do not add appreciably to the waste volume and usually do not add a contaminant that results in a mixed waste. Residual flocculent may, however, foul ion exchange resins or reverse osmosis membranes, so it is important that the quantity added be closely controlled.

8.4.3.4 Ion Exchange

Ion exchange is one of the most useful waste treatment techniques. Aqueous wastes that are free of oil and other organics and contain only very minimal quantities of solids may be subject to ion exchange on cation resin, anion resin, or specialty resins, either alone or in combination. If the contaminant is present as a cation, such as sodium, ammonia, or calcium, a cation resin can be used to replace the cation in solution. The cation from the resin will go into the solution to replace the contaminant cation. If the water stream is being recycled, the cation resin will probably be in the hydrogen form so that only hydrogen ions will enter the solution. If a hydrogen form of cation resin is used by itself, the water solution will likely become more acidic (lower pH). If an anion resin is used, anions in solution will be replaced with anions from the resin. Although resin may be in a chloride or other form, the hydroxyl form of the resin is often used so that anions are replaced with hydroxyl anions (-OH). If only a hydroxyl anion resin is used, the solution will drop in pH, becoming more basic. If both a hydrogen form of cation resin and a hydroxyl form of anion resin are used, the ions they add combine to form water, so both resins are used on demineralized water systems that are recycled. One disadvantage of most ion-exchange resins for waste treatment is the fact that they remove all ionic contaminants, not just the radioactive ones, and so are exhausted earlier than they might be. Selective resins are available for a few materials, most notably cesium, but are not available for plutonium.

In some applications, radionuclides pass through both cation and anion resin beds. This is assumed to happen because they are not present in an ionic form. They are either colloidal or are present in a molecule or complex that is neutral. In these cases, pretreatment or multiple treatment steps may be required.

Unfortunately, plutonium may be present as a cation, anion, neutral chemical complex, or colloid. Testing is almost always required to optimize plutonium removal. One additional limitation in the use of most ion exchange media for plutonium and other alpha-emitting radionuclides is that the radiation degrades the resin over time. Organic ion exchange media loaded with large quantities of plutonium may emit hydrogen and may become unstable when exposed to oxidizing materials such as nitric acid.

In some applications, ion exchange resins are “recharged” by the addition of large quantities of a particular ion (e.g., hydrochloric acid may be used to reconvert spent cation resin to the hydrogen form). In nuclear applications, this is rarely feasible because of the need to dispose of the recharge solution and because of the large quantity of rinse water used to remove the excess recharge solution from the resin.

8.4.3.5 Distillation

Distillation (including vacuum distillation) is at least conceptually simple. It removes all but volatile contaminants. In practice, some contaminants will cause foaming, and evaporator maintenance is often a problem. If laundry waste or other waste-containing detergents are to be evaporated, it may be necessary to add an antifoaming compound. Although these are sometimes effective, they often degrade with heat faster than the detergents or other compounds causing the foaming. Few evaporators take the product to dryness, as this often creates a scale build-up. If the evaporator bottoms are removed as a solution, they shall be solidified, usually with some increase in volume.

8.4.3.6 Purification by Reverse Osmosis

This process is highly effective on relatively pure water streams. The water is passed through a semipermeable membrane by mechanical pressure, leaving contaminants behind. The result is generally 80% to 99% of the influent water released as pure water, with the remainder containing all of the contaminants. Reverse osmosis has the advantage over ion exchange in that it will remove nonionic contaminants although these often shorten the life of the membrane. It is much more energy-efficient than distillation and requires much less equipment for the same volume of water treated. It is sometimes used as a “polishing” technique to further treat relatively clean water.

8.4.3.7 Solidification

Solidification is often a last-resort treatment because, while the other treatments described reduce the volume of solid waste requiring disposal, solidification increases it. Nevertheless, it is useful for some waste. Portland cement is the most common solidification medium for water solutions, aqueous suspensions, and resins. However, there are other proprietary materials, including some especially for oils and other organic compounds.

8.4.3.8 Solvent Extraction

Solvent extraction is used exclusively with organic solvents and involves mixing the solvent with an immiscible aqueous solution in which the contaminant is soluble. In this way, the contaminant is transferred to the aqueous solution for further treatment. (Solvent extraction may also be used in the other mode, in which the contaminant is transferred to the organic solvent solution, but this has fewer applications in waste management.) The organic solution is usually recycled.

8.4.3.9 Incineration

Incineration is an ideal waste-management technique for combustible solvents and other liquids that do not yield toxic or hazardous combustion products. The volume reduction from feed material to ash is usually outstanding. Incinerators are usually equipped with wet scrubbers, demisters, and filters to ensure that the effluent released to the environment is acceptable and ALARA. These features create secondary waste that shall be dealt with appropriately, but the disposal efficiency usually makes them well worthwhile.

8.4.4 Sampling and Monitoring

Sampling and monitoring of liquid waste streams are usually straightforward. Bulk liquid in tanks shall often be mixed, usually with a recirculating pump, before dip sampling to ensure a representative sample. Liquid effluent streams are often sampled with a flow-proportional sampler. For on-line monitoring, a small ion exchange column is used to concentrate ionic contaminants, and a detector is placed on the column for gamma analysis.

8.4.5 Storage and Disposal

Sanitary liquids and those meeting disposal criteria may be released to the environment or to sanitary waste treatment systems (sewerage systems). Hazardous liquid waste may be shipped, with excess absorbent material in compliance with 40 CFR, to a licensed treatment facility. Small quantities of radioactive-contaminated liquids, such as samples, may be shipped in a similar way, but most liquid waste shall be solidified prior to shipment or disposal. It is preferable to store only solid waste, as well. The recommendations of Table 8.3 are applicable to the storage of plutonium-containing liquids as well as solids. In particular, where long-term storage of plutonium solutions may occur, even within glove boxes, it is advisable to avoid plastic containers unless one can be certain that the alpha radiation will not have degraded the container.

Table 8.3. Recommendations for Storage of Plutonium Metal and Plutonium Oxide at Department of Energy Facilities

The following recommendations are made to improve current plutonium storage safety practices. Until new equipment and facilities become available to package plutonium based upon long-term standards, these recommendations are applicable to plutonium metal or plutonium oxide stored outside of glovebox lines in containers that do not have certified hermetic seals (i.e., per ANSI N14.5 (ANSI, 1997a)). These should be used in addition to the applicable requirement in DOE M 441.1-1 (DOE, 2008e).

1.	Plutonium solutions, metal turnings, or particles with specific surface areas greater than 1 cm^2/g should not be stored outside of glove boxes.
2.	All packages containing plutonium metal should be taped, re-taped, and placed in plastic bags prior to handling.
3.	Inspections should incorporate use of adequate personnel protection. Inspection practices should be codified in surveillance plans. These plans should reflect current facility operating status. There shall be personnel radiological surveillance during all handling operations. Personnel protection during operations should include protective clothing and gloves and, if necessary, respiratory protection.
4.	Inspection of containers should be integrated with audits for materials control and accountability (MC&A) to minimize container-handling and attendant radiation exposure to ALARA levels.
5.	Containers should be inspected for abnormalities (e.g., mass change, container deformation, or discoloration) using visual inspection, weighing, or video surveillance where such capability exists. Findings should be recorded for safety and MC&A evaluations. Visual inspections should be made at intervals of 1 week and 1 month after the material's initial containment and annually thereafter.
6.	Packages containing more than 0.5 kg of plutonium metal should undergo an annual surveillance in which the total mass of the package is determined to an accuracy of ±0.5 g and compared with the preceding year's mass and with the initial (reference) mass at the time of packaging. A storage package should be evaluated (e.g., opened and inspected, radiographed) if any of the following conditions are evident:
	a. The outer storage vessel is bulged or distorted. b. Hydride-catalyzed oxidation is suspected. Such reaction is indicated by a mass increase in either of two circumstances:

	Table 8.3 (cont'd)
	i) For packages whose masses continue to increase since initial packaging or for which historical mass data are unavailable (see item 6 above), a mass increase greater than 15 g per kilogram of plutonium over a one-year period indicates a hydride-catalyzed oxidation reaction.(a) ii) For a package whose mass has remained constant over a period of several years (less than ±0.5 g change) from its reference value, then undergoes an annual mass increase of more than 2 g per kilogram of plutonium, hydride-catalyzed reaction is indicated. Such a package is particularly suspect. The indications are that a previously sealed container may now be breached and that the continuing reaction may lead to rapid containment failure within 12 to 24 months
	c. The measured package mass, relative to the reference mass, corresponds to the mass that indicates formation of oxide with a volume exceeding 10% of the free volume of the inner vessel. Each 1-g increase in mass corresponds to formation of 1.5 cm^3 of oxide with a density of 50% of the theoretical value of 11.46 g/cm^3.
7.	Inspected containers exhibiting abnormalities (e.g., external contamination, bulging, discoloration, or other anomalies) should be repackaged in accordance with well-defined procedures (see items 3 and 4 above). Handling such containers outside of a glove box or conveyor confinement requires respiratory protection until the package is placed in an overpack container (e.g., taped metal can or sealed plastic bag) before further handling and transport.
8.	As an interim measure, material that is repackaged may be placed in a food pack can or slip-fit (Vollrath) container with a secured lid. If possible, metal should be repackaged in a configuration containing at least one gas-tight seal. No plastic material should be in direct contact with plutonium metal or oxide, and use of plastic in outer layers of packaging should be minimized.
9.	When packaging metal, hazardous or pyrophoric material such as plutonium hydride should be removed. However, it is not necessary to remove protective oxide film. Metal should be packaged in as dry and inert an environment as possible to minimize corrosion (<100 ppm H_20).

	Table 8.3 (cont'd)
10.	Impure oxide from sources other than metal should be thermally stabilized at 1000±100°C for at least an hour, or placed in a combination of a slightly lower temperature (850°C) for longer heating time to result in the lowest loss on ignition practicable with existing equipment. This ensures complete conversion of substoichiometric material and aids small-particle coalescence, which diminishes dispersal risk.
11.	Because plutonium oxide has greater potential for dispersion in severe accidents, it should have priority over metal for storage in structurally robust vaults. Metal should be characterized to ensure that it has not converted to oxide while in storage. Stored plutonium will have an increasing radiation level because of the build-up of ^{241}Am. Therefore, characterization of metal should be done as soon as possible and should make full use of small-sample statistical methods to minimize worker exposure. The results of characterization should be integrated with a site's surveillance plan, as well.
12.	Quality assurance measures, labeling, and material characterization are essential. Material and storage packaging specifics should be thoroughly documented.

(a) A higher oxidation rate may occur if the contained metal exhibits a high surface-area configuration, such as sheet or foil. The maximum annual increase for normal (uncatalyzed) oxidation of a given metal geometry can be calculated using a reaction rate of 3×10^{-7} g oxygen/cm^2-minute measured for alpha-phase plutonium under moist conditions at 50°C.

9.0 EMERGENCY MANAGEMENT

It is DOE policy that all DOE facilities and activities be prepared to respond to operational emergencies in a way that minimizes consequences to workers, the public and the environment. Formal emergency management programs are the final element of DOE's defense-in-depth against adverse consequences resulting from its operations.

9.1 EMERGENCY MANAGEMENT IN DOE

DOE Order 151.1C (DOE, 2005) requires DOE elements and contractors to plan and prepare for the management of emergencies. The following discussion of emergency management principles, requirements and guidance is generally applicable to DOE plutonium facilities. Specific facility requirements are in accordance with the individual facility DOE contract. The Emergency Management Guides (EMG), DOE G 151.1-1A, DOE G 151.1-2, DOE G 151.1-3, DOE G 151.1-4, provide guidance for implementing DOE Order 151.1C (DOE, 2007d, 2007e, 2007f, 2007g).

9.1.1 Basis for DOE Emergency Management Policy

DOE emergency management policy and direction is based on the following: planning and preparedness commensurate with hazards; integrated planning for health, safety and environmental emergencies; classification of and graded response to emergencies, and; multiple levels (tiers) of emergency management responsibility.

NOTE ON TERMINOLOGY: Within the Emergency Management System (EMS), “planning” includes the development of emergency plans and procedures and the identification of personnel and resources necessary to provide an effective response. “Preparedness” is the procurement and maintenance of resources, training of personnel, and exercising of the plans, procedures, personnel and resources. “Response” is the implementation of the plans during an emergency to mitigate consequences and to initiate recovery.

(a) **Planning and Preparedness Commensurate with Hazards**. Because of the wide range of activities and operations under DOE's authority, standards and criteria suited to one type of facility or hazard may be inappropriate for another. To deal with this diversity, while assuring an adequate overall state of preparedness, DOE Order 151.1C requires that the <u>details of each feature be tailored to the unique hazards of the specific facility.</u> This approach ensures a more complete and quantitative understanding of the hazards while providing for focused and cost-effective emergency planning and preparedness.

(b) **Integrated Planning for Health, Safety and Environmental Emergencies**. A wide variety of different types of Operational Emergencies can occur at DOE operations. Some may involve loss of control over radioactive or other hazardous materials unique to DOE operations, while others may involve security, transportation activities, natural phenomena impacts, environmental damage, or worker safety and health concerns. Planning, preparedness and response requirements applicable to DOE facilities and activities for some types of emergency conditions are specified by other agencies. For example, Federal regulations on occupational safety, environmental protection and hazardous waste operations have consequent "emergency planning" requirements. Rather than meet these requirements piecemeal through separate programs, each DOE/NNSA site/facility shall have an Operational Emergency Base Program that implements the requirements of applicable Federal, State, and local laws/regulations/ordinances for fundamental worker safety programs (e.g., fire, safety, and security). These requirements are not unique to DOE/NNSA operations.

(c) **Classification of Emergencies and Graded Response**. Operational Emergencies involving the airborne release of hazardous materials are grouped into one of three classes according to magnitude or severity. Classification of events is intended to promote more timely and effective response by triggering planned response actions generally appropriate to all events of a given classification. This principle, termed "graded response", is embodied in DOE Order requirements and is important to the effective management of response resources.

(d) **Tiers of Emergency Management Responsibility**. Within the EMS, responsibility for emergency management extends from the individual facility level to the cognizant DOE Field Element, and culminates at the cognizant Headquarters Program Secretarial Office (PSO). The responsibilities vested at each level of the hierarchy are specified in DOE Order 151.1C (DOE, 2005). The responsibility and authority for recognizing, classifying, and mitigating emergencies always rests with the facility staff. The head of the cognizant Field Element oversees the response of contractors and supports the response with communications, notifications, logistics, and coordination with other DOE elements. The DOE Headquarters (HQ) Emergency Operations Center (EOC) receives, coordinates, and disseminates emergency information to HQ elements, the cognizant PSO, Congressional offices, the White House, and other Federal Agencies.

9.1.2 Requirements Pertaining to All DOE Operations

DOE Order 151.1C (DOE, 2005) identifies program elements that comprise each DOE facility emergency management program. The elements form a standard framework, with the details of each program element varying according to the nature and magnitude of the facility hazards and other factors. The Order requires that a Hazard Survey be used to identify the generic emergency events or conditions that define the scope of the emergency management program. Where hazardous materials, such as plutonium, are present in quantities exceeding the quantity that can be "easily and safely manipulated by one person" and whose potential release would cause the impacts and require response activities characteristic of an Operational Emergency, the Order requires a facility-specific Emergency Planning Hazards Assessment (EPHA) be conducted and the results used as the technical basis for the program element content. Using the results of an objective, quantitative, and rigorous hazards assessment as a basis, each program is configured to the specific hazards and response needs of the facility. Detailed guidance on the implementation of the Order requirements has been published by the DOE Office of Emergency Management (DOE, 2007d, 2007e, 2007f, 2007g). These EMGs specify acceptable methods of meeting the EMS Order requirements.

Individual guides have been published for the technical planning basis (i.e., Hazards Survey/EPHA) processes and for programmatic and response program elements.

9.2 SPECIFIC GUIDANCE ON EMERGENCY MANAGEMENT FOR PLUTONIUM FACILITIES

This section provides technical guidance that is specifically applicable to the development and implementation of emergency management programs for plutonium facilities. It is intended to supplement, not replace, the more general recommendations provided in the EMG.

9.2.1 Technical Planning Basis

9.2.1.1 Hazards Survey

The Operational Emergency Base Program shall be based on a Hazards Survey. A Hazards Survey is an examination of the features and characteristics of the facility or activity to identify the generic emergency events and conditions (including natural phenomena such as earthquakes and tornadoes; wild land fires; and other serious events involving or affecting health and safety, the environment, safeguards, and security at the facility) and the potential impacts of such emergencies.

Each Hazards Survey shall—

(a) identify (e.g., in matrix or tabular form) the emergency conditions (e.g., fires, work place accidents, natural phenomena, etc.);

(b) describe the potential health, safety, or environmental impacts;

(c) indicate the need for further analyses of hazardous materials in an EPHA, based on the results of a hazardous material screening process; and

(d) identify the planning and preparedness requirements that apply to each type of hazard.

A Hazardous Material Screening Process shall identify specific hazardous materials and quantities that, if released, could produce impacts consistent with the definition of an Operational Emergency. The potential release of these materials to the environment requires further analysis in an EPHA. The release of hazardous materials less than the quantities listed below does not require quantitative analysis in an EPHA.

(a) In general, to meet the definition of an Operational Emergency, the release of a hazardous material shall: immediately threaten or endanger personnel and emergency responders who are in close proximity of the event; have the potential for dispersal beyond the safety of onsite personnel or the public in collocated facilities, activities, and/or offsite; and have a potential rate of dispersal sufficient to require a time-urgent response to implement protective actions for workers and the public.

(b) The hazardous material screening process shall identify all hazardous materials in a facility/activity that require further analysis in an EPHA. Specifically, for radioactive materials:

   (1) All radioactive materials in a facility/activity shall be subjected to a hazardous material screening process.

   (2) Radioactive materials that may be excluded from further analysis in an EPHA include: sealed radioactive sources that are engineered to pass the special form testing specified by the Department of Transportation (DOT) or ANSI; materials in solid form for which there is no plausible dispersal mechanism; materials stored in DOT Type B shipping containers with overpack, if the Certificates of Compliance are current and the materials stored are authorized by the Certificate; and, materials used in exempt, commercially available products.

   (3) Radioactive hazardous materials that require further analysis in an EPHA include the radioactive materials listed in DOE-STD-1027-92 in quantities greater than the Category 3 values given in Attachment 1, Table A.1., of that Standard.

9.2.1.2 Emergency Planning Hazards Assessment (EPHA)

Unique properties and characteristics of plutonium and its compounds may need to be considered at certain steps in the EPHA process.

(a) **Description of Facility and Operations.** The properties of the hazardous material do not significantly affect the manner in which this step of the EPHA is performed, except to the extent that plutonium safety considerations may mandate more detailed descriptions of certain facility physical or operational features.

(b) **Characterizing the Hazards.** The objective of this step is to describe the hazardous materials in sufficient detail to allow accurate modeling of releases and calculation of consequences. The following properties of plutonium and its compounds influence the release potential and consequences.

   - Chemical and physical form. The chemical toxicity of plutonium and its compounds is of much less concern than the radiotoxicity of the plutonium. However, the chemical and physical form may strongly influence the release potential. Plutonium metal oxidizes readily in humid air at elevated temperatures to form loosely-attached oxide particles, a source of readily dispersible airborne and surface contamination. Plutonium metal fines and turnings can ignite spontaneously in the presence of air, creating aerosol-size oxide particles and providing energy to disperse them. Also, some plutonium compounds may ignite violently on contact with air, water or hydrocarbons (Benedict, et al., 1981).

- Solubility. The CED per unit activity inhaled is about three times greater for plutonium of material type M than for material type S. No plutonium compounds of material type F are generally recognized.

- Particle size. Particle size distribution has a large effect on the radiotoxicity of inhaled materials. Larger particles tend to be cleared rapidly from the upper respiratory regions and swallowed, thereby delivering little radiation dose to the lung tissues. Because plutonium is poorly absorbed in the gut, very little dose is attributed to the larger particles that are cleared from the body by this process. Small particles are deposited deeper in the lung and are cleared very slowly, producing a much larger dose per unit activity inhaled. Extremely small particles tend to be exhaled and not deposited.

- Isotopic mixture. Characterization of the isotopic mixture is important to the accuracy of both dose calculations and contamination measurements. When the inventory or quantity released is expressed as the total activity (Ci or Bq) of a mixture of isotopes, the total often includes the ^{241}Pu activity. Because ^{241}Pu decays almost exclusively by beta emission, it contributes little to the internal dose from a mixture of Pu isotopes. Also, the fraction of ^{241}Am (from decay of ^{241}Pu) in plutonium can vary greatly, depending on the degree of irradiation and the time since the plutonium was chemically separated from the reactor fuel. Characterization of contamination from a plutonium mixture is often done by detecting the low-energy photons emitted by ^{241}Am, which requires knowledge of the activity of ^{241}Am compared to the other isotopes in the mixture.

(c) **Developing Event Scenarios.** The properties of the hazardous material do not significantly affect the manner in which this step of the hazards assessment is performed.

(d) **Estimating Potential Event Consequences.** For the scenarios developed in the previous step, this step determines the area potentially affected, the need for protective actions, and the time available to take those actions. The way these consequences are determined depend on properties of the hazardous material.

For plutonium and its compounds, inhalation during plume passage is the most important exposure process in the early phase of an emergency. After passage of a plume, exposure to material deposited on the ground will dominate. Therefore, the following features should be considered when selecting and applying calculation models:

- Inhalation pathway dose. For any realistic mixture of plutonium isotopes, the great majority of the dose will be by the inhalation pathway. Therefore, the model selected to estimate consequences of an atmospheric plutonium release shall be able to calculate the TED to an individual exposed by inhalation.

- Plume depletion during transport. As it is transported downwind, an aerosol plume will be depleted by gravitational settling of particles. Because of the high density of plutonium and its compounds, this depletion effect can be very significant in reducing the dose. Therefore, a consequence model that accounts for plume depletion by gravitational settling should be used. When analyzing consequences of any postulated accidental criticality, any model selected should account for the decay during transport of short-lived fission product gases.

- Ground deposition. Following passage of a plume, the amount of plutonium deposited on the ground will determine whether long-term intervention to minimize the dose to the resident population will be required. The consequence model selected should calculate ground deposition to support protective action planning.

9.2.2 Program Elements

Properties and characteristics of plutonium and its compounds shall be considered in formulating the emergency management program elements. Following are specific program element considerations related to the hazardous properties of plutonium.

9.2.2.1 Programmatic Elements

The specific properties of the hazardous material do not significantly affect the content of the programmatic program elements: Program Administration, Training and Drills, Exercises, and Readiness Assurance.

9.2.2.2 Response Elements

(a) **Emergency Response Organization.** The primary influence of plutonium's hazardous properties on the Emergency Response Organization (ERO) is in the staffing of the consequence assessment component. As will be discussed in e) below, staff should be assigned to the ERO who are knowledgeable of and able to quantitatively evaluate the radiological aspects of the hazard.

(b) **Offsite Response Interfaces.** The specific properties of the hazardous material do not significantly affect the content of this program element.

(c) **Operational Emergency Event Classes.** As with all hazardous materials, classification of emergencies for plutonium facilities should be based on the predicted consequences at specific receptor locations, as compared with numerical criteria for taking protective action (TED). The classification of the postulated event or condition should be determined during the EPHA process and the observable features and indications identified as Emergency Action Levels (EALs) for that event/condition.

(d) **Notification.** The specific properties of the hazardous material do not significantly affect the content of this program element.

(e) **Consequence Assessment.** As discussed in section 9.2.1.2 d, models and calculation methods used for consequence assessment should be appropriate to the physical, chemical, and radiological properties of the hazards. Models used to calculate and project the radiological consequences of a release of plutonium should be the same ones used in the EPHA process. If the same models are not used, the differences between outputs should be characterized and documented to avoid the potential for confusion and indecision during response to an actual emergency.

Environmental monitoring capability for assessing consequences of a plutonium release should conform to several general principles.

- Procedures for measurement of airborne plutonium should provide for timely analysis and reporting of results in units that correspond to decision criteria. Decision points based on initial alpha screening measurements with field instruments should account for the expected levels of radon progeny collected on the air sample media. Alternatively, portable survey instruments capable of performing alpha spectroscopy measurements can be used to provide rapid isotopic analysis of plutonium collected on sample media. Precautions should be taken when using radon stripping instrumentation. One site found that it would not work with mixed alpha and beta emitters such as Uranium and Thorium. Another site found the same result using Strontium and Plutonium.

- Measurement of plutonium deposition should be planned and proceduralized to yield results that correspond to those needed by the predictive models used for emergency response. The correlation between direct or indirect radioactivity measurements (in units of activity) and measurement methods that give mass or concentration of plutonium in a sample should be established for standard sample sizes, collection efficiencies, and the expected isotopic mixture(s) of material that might be released. Information on expected isotopic mixture should be available for converting the results of measurements made with photon-sensitive instruments, such as the Fiddler and Violinist, into plutonium activity per unit area.

- If the potential exists for release of plutonium in conjunction with materials of high chemical toxicity, it is generally not practical to plan on use of survey teams to quantify concentrations in a plume. The high risk to survey personnel, the protective equipment necessary to minimize that risk, the time needed to prepare and deploy a team for such a survey and the limited value of the information that could be gained all weigh against this approach to assessing the consequence of a highly toxic release.

- Continuous environmental air samples are taken around the perimeter of some plutonium facilities for environmental reporting purposes.

  Consequence assessment procedures should provide for the rapid retrieval and analysis of sample media from any fixed samplers that may be operating in an area affected by a plutonium release. The procedures should specify the type of measurements to be done on those sample media, including any instrument settings, conversion factors, or adjustments needed to produce useful results in the shortest time possible.

(f) **Protective Actions.** The Protective Action Guides (PAGs) published by the U.S. Environmental Protection Agency (EPA, 1991) have been adopted by DOE as its basic protective action criteria for planning and response. The terms "PAG" and "EPA Protective Action Guides" used in the Order should be interpreted as follows:

- A projected dose equivalent of 10 mSv (1 rem) total effective dose equivalent to reference man, where the projected total effective dose equivalent is the sum of the effective dose equivalent from exposure to external sources and the committed effective dose equivalent from inhalation during the early phase; or
- A projected committed dose equivalent to the adult thyroid of 50 mSv (5 rem); or
- A projected committed dose equivalent to the skin of 500 mSv (50 rem).

Facilities having substantive and persuasive arguments for using other protective action threshold values may propose values that are specific to their radioactive material holdings and operations. Any alternative proposals should be supported by an analysis that addresses the four principles that form the basis for the selection of the EPA PAG values and the other considerations utilized in the selection process, as discussed in Appendix C of the EPA 400-R-92-001.

(g) **Medical Support.** If the potential exists for large intakes of plutonium, the emergency management program should include specific planning for the quantification of exposure, diagnosis of health effects, and treatment. Medical facilities providing emergency medical support should be provided with references relating to plutonium toxicity and treatment protocols. Criteria for implementing treatments such as surgical excision of contaminated tissue, lung lavage, or use of chelating agents should be discussed with the medical staff and sources of real-time advice and assistance should be identified.

(h) **Recovery and Reentry.** The specific properties of the hazardous material do not significantly affect the content of this program element.

(i) **Public Information.** The specific properties of the hazardous material do not significantly affect the content of this program element.

(j) **Emergency Facilities and Equipment.** Except for instruments and analysis methods used in consequence assessment, little by way of specialized facilities and equipment will be required to meet the emergency management program needs of plutonium facilities. Equipment and analytical techniques for detection and measurement of plutonium in environmental sample media should have sufficient sensitivity to measure levels at or below those corresponding to decision criteria. Whereas larger sample sizes, chemical processing, or longer counting times may be used to reduce the limit of detection for routine environmental surveillance, time constraints may dictate that more sensitive techniques be available to meet the information needs of emergency response.

10.0 DECONTAMINATION AND DECOMMISSIONING

At the end of the useful life of a facility, activities are undertaken to restore the facility to non-contaminated status and permit its unrestricted use. These activities are typically termed decontamination and decommissioning (D&D).

Although plutonium facilities are no longer useful and operational activities are no longer conducted, measures shall be continued to control the residual radioactivity. The decision may be made to undertake a D&D program to minimize or eliminate long-term institutional control. This may be done in a variety of ways, most of which may be termed D&D. The exception is converting the facility to some other nuclear use. With the elimination of the DOE weapons production mission, more plutonium-contaminated facilities will require D&D in the near future.

This chapter provides guidance on establishing and implementing an effective D&D program. Major topic areas include regulations and standards, design features, D&D program, D&D techniques, and D&D experience. This chapter concentrates on the radiation-protection aspects of D&D at plutonium-contaminated DOE facilities.

10.1 REGULATIONS AND STANDARDS

The standards that apply to the decommissioning of a plutonium-contaminated facility include virtually all of those that were applicable during facility operations, (e.g., 10 CFR 835, DOE P 450.1 and 10 CFR 851) plus some additional ones such as 10 CFR 835.1002(d). The occupational safety and radiation dose limits, safety management requirements, radioactive and hazardous chemical disposal regulations, and transportation requirements are unaffected by the activity to which they apply.

No single DOE regulation covers all D&D requirements due to the wide variety of issues encompassed by D&D. These issues include project management, environmental surveillance, health and safety of workers and the public, engineering design, characterization survey techniques, D&D techniques, waste management, and waste transport. The primary DOE Orders pertaining to D&D activities are DOE Order 430.1B, Ch.2, Real Property Asset Management (DOE, 2008f); DOE Order 458.1, Ch. 2, Radiation Protection of the Public and Environment (DOE, 2011c); DOE O 231.1B, Environment Safety and Health Reporting (DOE, 2009g); DOE Order 420.1C, Facility Safety (DOE, 2012b). The DOE operations offices may have implementation procedures corresponding to these Orders that which contractors will also need to comply.

DOE Order 430.1B, Ch.2, Real Property Asset Management (DOE, 2008f), provides the requirements to ensure a disciplined, systematic, and coordinated approach to project management. All projects, including D&D projects, should have clearly defined goals and objectives that support program requirements. Specific objectives include (1) promoting project execution that meets technical, schedule, and cost objectives, (2) meeting all applicable environmental, health and safety, and quality assurance requirements, and (3) avoiding a commitment of major resources before project definition. Good program management techniques should consider D&D costs as part of the lifecycle cost and select a tentative D&D method during the facility design phase.

DOE Order 458.1, Ch.2, Radiation Protection of the Public and Environment (DOE, 2011c), provides radiological protection requirements and guidelines for cleanup of residual radioactive material and management of the resulting wastes and residues and release of property. This DOE Order establishes a basic public dose limit for exposure to residual radioactive material (in addition to naturally occurring "background" exposures) of a 100-mrem (1-mSv) effective dose equivalent in a year. A more detailed discussion is presented below in Section 10.1.3.

DOE O 420.1C, Facility Safety (DOE, 2012b), establishes facility safety requirements related to: nuclear safety design, criticality safety, fire protection and natural phenomena hazards mitigation.

DOE O 231.1B Environment, Safety and Health Reporting (DOE, 2009g), ensures collection and reporting of information on environment, safety and health that is required by law or regulation to be collected, or that is essential for evaluating DOE operations and identifying opportunities for improvement needed for planning purposes within the DOE.

10.1.1 Other Regulations

The D&D of most plutonium-contaminated facilities will involve cleanup of a combination of radioactive wastes, hazardous wastes, and mixed wastes. Some other Federal regulations not already discussed that are applicable to the cleanup and disposal of these wastes are summarized in this section along with the DOE guidance on implementation. This is not an all inclusive list. It is the facility responsibility to identify applicable requirements and ensure compliance.

-- National Environmental Policy Act (NEPA) (USC, 1970) and 40 CFR 1500 (CEQ, 1992)
- This act established a national policy to ensure that environmental factors are considered in any Federal agency's planning and decision making. DOE O 451.1B, National Environmental Policy Act Compliance Program (DOE, 2010c), defines DOE responsibilities and procedures to implement NEPA. The decommissioning of a DOE plutonium facility will require a determination of whether or not the action is a "major or significant government action adversely affecting the environment" in accordance with NEPA. If it qualifies as such an action, an environmental assessment (EA) or environmental impact statement (EIS) will be required. An EA or EIS will need to discuss the amount of material that will remain onsite and its effect, in addition to addressing the alternatives. The alternatives will include retaining radioactive material onsite under DOE control, cleaning the site to a level that would be acceptable for unrestricted release, and the null or no-action alternative of "walking away" from the site. If the action does not require an EA or EIS, either because the possible adverse effects are insignificant or because decommissioning was adequately addressed in a preoperational or other EA or EIS, then the decommissioning can proceed in accordance with the information contained in other applicable regulations.

- Resource Conservation and Recovery Act (RCRA) (USC, 1976a) - This act authorizes the EPA and the States to regulate hazardous and solid wastes.

- Comprehensive Environmental Response, Compensation, and Liability Act (USC, 1980) and 40 CFR 300 (EPA, 1992c) - This act requires the identification and cleanup of inactive hazardous waste sites by responsible parties, and imposes certain response and reporting requirements for releases of hazardous substances.

- Superfund Amendments and Reauthorization Act (USC, 1986) and 40 CFR 300 (EPA, 1992c).

Interagency agreements can also exist between DOE, EPA, state, and local agencies (Daugherty, 1993). Any special arrangement agreed to as part of an interagency agreement will need to be honored during the D&D activities.

10.1.2 Residual Radioactivity Levels

A primary concern in the D&D of any nuclear facility is the level of residual radioactivity that may be permitted for unrestricted use. However, the emphasis of this document is on occupational radiological protection. See Section 4.2.4 for guidance on contamination monitoring in the workplace. Additional information on acceptable residual levels may be found in the following sources. This list is not inclusive and facilities shall determine the applicable requirements. For clearance of DOE property for use by the public, DOE O 458.1 requirements are applicable. The U.S. Nuclear Regulatory Commission (NRC) in Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors (AEC, 1974), provide definitive values for acceptable surface contamination levels for termination of operating licenses for nuclear reactors and for materials, equipment, and facilities and ANSI/HPS N13.12 (ANSI, 1999b) and IAEA Safety Guide RS-G-1.7, *Application of the Concepts of Exclusion, April 2005, Exemption and Clearance,* provide values for materials and equipment.

DOE Order 458.1, Ch. 2, Radiation Protection of the Public and Environment (DOE, 2011c), provides the following DOE guidelines for cleanup of residual radioactive material, management of the resulting wastes, and release of property. The basic public dose limits for exposure to residual radioactive material in addition to natural background exposures is a 100-mrem (1-mSv) effective dose equivalent in a year from all sources and pathways. The effective dose equivalent in a year is the sum of the effective dose equivalent from exposures to radiation sources external to the body during the year plus the CED from radionuclides taken into the body during the year. Because the limit applies to all sources and pathways DOE recommends use of a 25 mrem in a year dose constraint to ensure that exposures from this source (residual radioactive material) does not combine with other non-background sources to cause doses in excess of 100 mrem in a year.

DOE Order 458.1, Ch. 2 (DOE, 2011c) also provides the following guidelines for (1) residual concentrations of radionuclides in soil, (2) concentrations of airborne radon decay products, (3) external gamma radiation, (4) surface contamination, and (5) radionuclide concentrations in air or water:

-- **Residual radionuclides in soil** - Generic guidelines for thorium and radium (^{226}Ra, ^{228}Ra, ^{230}Th, and ^{232}Th) are 5 pCi/g averaged over the first 15 cm of soil below the surface and 15 pCi/g averaged over 15-cm-thick layers of soil more than 15 cm below the surface. For other radionuclides in soil (e.g., plutonium), specific guidelines should be derived to be as low as reasonably achievable below the basic dose limit and consistent with DOE dose constraints. It should be supported by means of an environmental pathway analysis using specific property data where available. Residual concentrations of radioactive material in soil are defined as those in excess of background concentrations averaged over an area of 100 m^2.

-- **Airborne radon decay products** - Applicable generic guidelines are found in 40 CFR 192 (EPA, 1992b). In any occupied or habitable building, the objective of remedial action should be, and a reasonable effort should be made to achieve, an annual average (or equivalent) radon decay product concentration (including background) not to exceed 0.02 WL. Remedial actions by DOE are not required to comply with this guideline when there is reasonable assurance that residual radioactive material is not the source of the radon concentration.

-- **External gamma radiation** - The average level of gamma radiation inside a building or habitable structure on a site to be released without restrictions should not exceed the background level by more than 20 μR/h.

-- **Residual Surface Activity** - The DOE guidelines on transuranic surface contamination levels are consistent with NRC Regulatory Guide 1.86 and are discussed in a DOE memorandum dated November 17, 1995, "Application of DOE 5400.5 requirements for release and control of property containing residual radioactive materials," the guideline values are as follows:

<u>Guidelines</u>

Removable Contamination	20 dpm/100 cm^2
Total (Fixed plus Removable Contamination)	100 dpm/100 cm^2
Maximum	300 dpm/100 cm2

The order also permits alternative surface activity guidelines that ALARA-based and derived using pathway dose analysis.

-- **Residual radionuclides in air and water** - Residual concentrations of radionuclides in air shall not cause members of public to receive an effective dose equivalent greater than 10 mrem (0.1 mSv) in one year [DOE Order 458.1 (DOE, 2011c)]. In 40 CFR 141, National Primary Drinking Water Regulations (EPA, 1992a), the EPA provides a limit of 4 mrem/y annual dose equivalent to the whole body or any internal organ of any member of the public from manmade radionuclides in drinking water.

The NRC has updated their decommissioning regulations and criteria, see 10 CFR Part 20 subpart E. NRC established a 25 mrem in a year plus ALARA requirement that is to be demonstrated by use of pathway dose analyses to derive criteria and surveys to demonstrate properties meet the derived criteria. Screening levels are also provided in the associated NRC guidance documents that may be used instead of the derived criteria. Both DOE and NRC allow use of RESRAD or RESRAD-build for deriving criteria. NRC screening levels are not applicable to DOE operations and may be used only with DOE pre-approval.

The derivation of criteria requires calculation of dose to members of the general population. The scenarios for exposure will have to include all exposure pathways that are credible under the proposed disposition. If the site is part of a closely guarded government reservation, certain pathways may be eliminated, such as the use of well water directly from the site and ingestion of significant quantities of fruits and vegetables grown on the site. However, if the site will be released for unrestricted use, such scenarios should be considered. The computer codes used for calculation of dose to the public from decommissioned facilities will include the currently accepted exposure models and site-specific or maximum credible parameters for exposure pathways.

A multi-agency effort has developed measurement and decision criteria applicable to D&D projects. The Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) has been published (DOE, 2000). It provides detailed survey techniques applicable to the D&D of DOE facilities. A related document for sampling and analysis of environmental samples has also been approved for interagency use: *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (MARLAP) EPA 402-B-04-001A, July 2004.

10.2 DESIGN FEATURES

Design of the facility should allow easy D&D of equipment and materials. Details on designing facilities for ease of decommissioning are discussed in the following sections. 10 CFR 835.1002 and Appendix C of this document provide additional guidance on facility design.

10.2.1 Building Materials

In general, the design features that aid in contamination control during operation also facilitate decommissioning. The inclusion of all the building materials suggested in this section may be cost-prohibitive, but they should be considered if the budget allows. The maintenance procedures that are used during operation are also important in controlling the spread of contamination to clean areas and, therefore, they facilitate decommissioning, too.

Less permeable building materials are more easily decontaminated. Any concrete with uncoated surfaces that comes in contact with plutonium solutions or plutonium contaminated air will require surface removal and disposal as radioactive waste at the end of its life. If there are cracks through which contaminated solutions have penetrated, the entire structure may need to be disposed of as radioactive waste.

Metal surfaces may also require decontamination. In general, the more highly polished the surface, the easier it will be to decontaminate. If feasible, all stainless steel that will come into contact with plutonium should be electropolished before being placed into service. If high-efficiency particulate air (HEPA) filtration has failed at any time during facility operation, roofs may require decontamination. Metal roofs are easiest to decontaminate, but even these may contribute to the volume of radioactive waste unless unusual measures are taken to clean them. Built-up and composition roofs will be difficult to clean to unrestricted release levels.

Interior surfaces are most easily cleaned if they were completely primed and painted before the introduction of radioactive materials into the facility. If interior surfaces are repainted during operation, their disposal as clean waste is likely to require removal of the paint. However, if the paint has deteriorated, cleaning for unrestricted use may be as difficult as if the material had never been painted. Wood will almost certainly become contaminated, as will plasterboard and other such materials.

Floor surfaces are likely to be a problem. Concrete should be well sealed and covered with a protective surface. Single sheet, vinyl flooring with heat-sealed seams is preferable to asphalt or vinyl tile because it is more easily cleaned. If the floor needs resurfacing, it is preferable to overlay new flooring material rather than remove the old material and expose the underlying floor.

Carpets are not recommended because they are difficult to clean and survey and bulky to dispose of and they do not adequately protect the underlying surface. In some areas, such as control rooms, their use may be justified by noise control requirements; however, their contamination control limitations should be considered. If used, carpets should be surveyed frequently and disposed of as radioactive waste when they become contaminated.

10.2.2 Ventilation Systems

In addition to decommissioning considerations, the design of the ventilation system will depend on the operations that will be conducted in the facility. Adequate air flow for all operations and good design practices will help keep the facility clean during operations and will facilitate decommissioning. Fiberglass duct work may present a fire hazard and may be more difficult to decontaminate than stainless steel, especially stainless steel that has been electropolished. Welded joints are less likely to collect contamination than bolted ones; however, bolted joints are easier to remove and the most contaminated areas are readily accessible for cleaning.

Filters should be positioned in ventilation systems to minimize contamination of ductwork (e.g., filtration of glove-box exhaust air before it enters a duct leading to a plenum).

10.2.3 Piping Systems

Potentially contaminated piping systems that are imbedded in concrete are a common and relatively expensive decommissioning problem. Most often, they shall be sealed and removed last, after all other radioactive material has been removed and the building is being demolished by conventional methods. Often, they provide the major impetus for demolishing a building rather than converting it to some non-nuclear use. For this reason, it is best to run pipes in chases or tunnels that have been lined (usually with stainless steel) to prevent contamination from penetrating building surfaces. To minimize hand jackhammer work required during decommissioning, floor drains should not be enclosed in concrete.

10.2.4 Soil-Contamination Considerations

Depending on the activity levels found, locations where contaminated effluents have penetrated the ground may require excavation during decommissioning. The facility design should minimize such areas. Particular attention should be paid to storm runoff from roofs, storage areas, contaminated equipment storage, and liquid waste treatment impoundments (including sanitary sewage systems if they may receive some small amount of contamination during the life of the facility.)

10.2.5 Other Features

Installed decontamination and materials-handling equipment that facilitates operation and maintenance generally facilitates decommissioning in two ways. First, it can be used for its intended purposes of cleaning and moving equipment during the decommissioning phase. Even more important, it usually contributes to a cleaner, better maintained facility, where nonfunctional equipment is moved out when it is no longer needed and work surfaces are kept free of spreadable contamination.

Other features include the following:

-- Minimizing service piping, conduits, and ductwork;

-- caulking or sealing all cracks, crevices, and joints;

-- using modular, separable confinements for radioactive or other hazardous materials to preclude contamination of fixed portions of the structure;

-- using localized liquid transfer systems that avoid long runs of buried contaminated piping;

-- using equipment that precludes the accumulation of radioactive or other hazardous materials in relatively inaccessible areas, including curves and turns in piping and ductwork;

-- using designs that ease cut-up, dismantling, removal, and packaging of contaminated equipment from the facility;

-- using modular radiation shielding, in lieu of or in addition to monolithic shielding walls;

-- using lifting lugs on large tanks and equipment; and

-- using fully drainable piping systems that carry contaminated or potentially contaminated liquids.

10.3 DECONTAMINATION AND DECOMMISSIONING PROGRAM REQUIREMENTS

Planning for facility decommissioning should be initiated during the design phase for new facilities and before termination of operations for existing operational facilities. To assist in D&D activity planning the Office of Environmental Management distributed the "Decommissioning Resource Manual." Refer to that document for guidance.

Requirements relating to occupational radiological protection include (this is not an all inclusive list, facilities shall determine the applicable set of requirement):

DOE Order 430.1B, Ch.2, Real Property Asset Management (DOE, 2008f), contains the requirements by which all DOE projects shall be managed; It requires that a project management plan be developed for major system acquisitions and major projects and states that environment, safety, and health technical requirements for project design and implementation should be included in the work-plan section of the project management plan.

10.4 DECONTAMINATION AND DECOMMISSIONING TECHNIQUES

This section concentrates on decontamination techniques to be used in the final decommissioning of a plutonium-contaminated facility for unrestricted release. Some of these techniques are similar to those used during routine operations (e.g., some equipment and building surface decontamination). Contamination detection methods are similar for routine and D&D operations and are discussed in Chapter 4.

10.4.1 Equipment and Surface Decontamination

Decontamination of surface areas may be as simple as hosing off the floors with water, washing surfaces with detergent and water, or wiping with household dust cloths. Waste material generated from decontamination activities (e.g., water and wipe material) shall be contained and disposed of as radioactive waste. For some locations, vacuuming the surfaces may be appropriate. If vacuuming is used, HEPA-filtered vacuum systems are required to keep airborne radioactive material out of the vacuum exhaust.

For some operations, periodic surface flushing with water may be adequate to maintain acceptable contamination levels. Precautions should ensure control and collection of run-off water so that material may be recovered and waste water analyzed before discharge. Depending upon which isotope of plutonium is involved, geometrically safe containers may be required for collecting and holding the liquid.

Depending upon the physical and chemical form of the plutonium and the type of surface, plutonium may become imbedded in the surface. Removal of embedded material may require physical abrasion, such as scabbling, grinding, sand blasting, or chipping, or it may be accomplished using chemical etching techniques. If the surface is porous, complete replacement could be necessary. The use of high-pressure water (hydroblasting) has been quite successful for metal and concrete surfaces.

Ultrasonic cleaning techniques (electropolishing) or chemical baths may be useful for decontamination of high-cost items if the chemicals used are compatible with the material to be cleaned.

A description of different decontamination techniques is found in DOE/EV/10128-1, DOE Decommissioning Handbook (DOE, 1980), and publications by Allen (1985) and the Electric Power Research Institute (EPRI, 1989). The DOE Decommissioning Handbook also includes guidance on decontamination techniques, assessment of environmental impacts, disposition of wastes, and preparation of decommissioning cost estimates.

10.5 DECONTAMINATION AND DECOMMISSIONING EXPERIENCE

Considerable experience has been gained in D&D of commercial plutonium facilities, as discussed in Hoovler et al. (1986), Denero et al. (1984), and Adams et al. (1982). Hoovler et al. (1986) discuss the decommissioning programs carried out at two Babcock and Wilcox buildings in Lynchburg, Virginia, which housed plutonium/uranium fuel development laboratories. They include information on decommissioning and quality assurance plans, conducting D&D work, performing radiological surveys before and after D&D work, and disposing of the waste. Denero et al. (1984) discuss the D&D of the Westinghouse Nuclear Fuel Facility at Cheswick, Pennsylvania. They describe the facility and its operations, nondestructive assay techniques, equipment required for dismantling and packaging the waste, and management of the TRU waste. Adams et al. (1982) discuss the complete D&D of the Westinghouse Advanced Reactors Division Fuel Laboratories at the Cheswick, Pennsylvania, site. The report describes the D&D plans, the EA written for the operation, the quality assurance plan, and the health physics, fire control, and site emergency manuals written for the operation.

Discussions of D&D activities at several DOE plutonium facilities are provided by Adkisson (1987), Bond et al. (1987), and King (1980), as well as by Shoemaker and Graves (1980), Garner and Davis (1975), Wynveen et al. (1982), Hunt et al. (1990), Freas and Madia (1982), and Garde et al. (1982a, 1982b). They describe D&D activities that took place in several types of plutonium facilities, including fabrication facilities, research and development laboratories, and a storage facility. Plutonium-contaminated glove boxes, hoods, ventilation ductwork, laboratory equipment, structural components (i.e., walls and floors), and filter banks were decontaminated. Typically, decontamination methods included wiping with a damp cloth or mop, using strippable coatings, mechanical spalling of concrete floor surfaces, and fixating contamination on a piece of equipment (e.g., a hood), followed by disassembling the item inside a contamination control enclosure.

Some lessons learned from past studies include the following:

- Waste management planning should begin early in the D&D planning stages and consider the following:
  - The possibility exists that there may be more stringent regulations for shipping hazardous or radioactive wastes than disposing of it and
  - Compliance with all applicable waste management requirements may be difficult (e.g., WIPP has unique limits on Beryllium content, typically <1%, for criticality safety due to its disposal array being so large).
- It is difficult to decontaminate some items with inaccessible surfaces to less than the TRU limit (100 nCi/g) so that they can be disposed of as LLW. In some situations, it may be possible to decontaminate to <100 nCi/g of TRU, but the decontamination process may generate a large volume of liquid waste or be time-consuming enough to prohibit its use.
- Temporary enclosures, with appropriate venting, are effective in controlling contamination when reducing the size of large equipment such as glove boxes. Any loose contamination on the equipment should be fixed before placing it in the enclosure.
- Criticality safety issues regarding the geometry of any waste material containing fissile material need to be considered.

Adkisson (1987) reported on the decommissioning of a plutonium fuel fabrication plant at the Sequoyah Fuels Corporation's Cimarron Facility, located in north-central Oklahoma. Process equipment, glove boxes, tanks, piping, and ventilation ducts required decontamination. Controlling personnel exposures, maintaining containment of radioactive material during the dismantling of contaminated items, and reducing the volume of TRU material were the primary considerations during the decommissioning activities. A large modified glove box provided containment for dismantling and cutting up the various equipment using a plasma-arc unit. A passive, gamma-ray nondestructive assay technique (heavily shielded NaI detector with collimator) was used to measure the plutonium content of cut-up pieces. Finally, the loaded waste drums were measured using a waste drum counter to ensure that plutonium levels were less than 100 nCi/g.

A number of plutonium-contaminated facilities have been decommissioned at Mound Laboratory (Bond et al., 1987). Interdepartment management teams, including representatives from program management, operations, project engineering, maintenance, technical support, and environmental, safety, and health were established for the D&D projects. The team met monthly to discuss program status and they met quarterly with DOE staff. A graded D&D approach was used. First, standard cleaning (e.g., wiping with a damp cloth) and flushing techniques were used to remove loose contamination. Then, more aggressive decontamination methods were performed inside temporary enclosures. Finally, glove boxes and equipment that could not be decontaminated to unrestricted release levels were cut into sections using a plasma-cutting method and then packaged as waste. The plasma-cutting method generated less smoke, thus reducing the particulate accumulation on the HEPA filters.

During cleanup of a plutonium-contaminated storage facility, strippable fixatives were used as a contamination control and a decontamination method (King, 1980). Fixatives in combination with cheese cloth were used to clean smooth vertical surfaces and difficult-to-reach areas. The cheese cloth was placed on the area to be cleaned and then sprayed with a fixative. The cheese cloth and fixative were then stripped from the surface, removing contamination in the process. Accidental criticalities can be a concern when disposing of this material that contains fissile material contamination, as discussed in Section 8.0, and criticality safety specialists should be consulted. Facility personnel also need to determine if the fixative is classified as a hazardous material and dispose of it accordingly.

Addition sources of relevant information can be found at:

- http://rockyflats.apps.em.doe.gov/references/Closure_Legacy_Document.pdf
- https://www.dndkm.org
- http://webdev.arc.fiu.edu/wp-content/uploads/downloads/2010/09/ARC-Project-Factsheet-DD-Toolbox-Fixative-sprayer-demo-rev-1a.pdf
- http://www5.hanford.gov/pdw/fsd/AR/FSD0001/FSD0012/DA451399/DA451399_40061_22.pdf
- http://www.efcog.org/wg/esh_rp/events/RPSG_Spring_12_Meeting/Eric%20McKamey%20-%20PFP%20D&D%20Rad%20Con.pdf
- http://ndreport.com/resources/ (This is commercial information and may be helpful to individuals working in oversight of D&D activities.)

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APPENDIX A

GLOSSARY

Terms used consistent with their regulatory definitions.

abnormal situation: Unplanned event or condition that adversely affects, potentially affects or indicates degradation in the safety, security, environmental or health protection performance or operation of a facility. **(RCS)**

activity median aerodynamic diameter: The diameter of a sphere having a density of 1 g cm^{-3} with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol. **(Internal Dosimetry Chapter of the IG)**

air sampling: A form of air monitoring in which an air sample is collected and analyzed at a later time, sometimes referred to as retrospective air monitoring.

air monitoring: Actions to detect and quantify airborne radiological conditions by the collection of an air sample and the subsequent analysis either in real-time or off line laboratory analysis of the amount and type of radioactive material present in the workplace atmosphere. **(Internal Dosimetry Chapter of the IG)**

airborne radioactive material: Radioactive material in any chemical or physical form that is dissolved, mixed, suspended, or otherwise entrained in air.

alarm set point: The count rate at which a continuous air monitor will alarm, usually set to correspond to a specific airborne radioactive material concentration by calculating the sample medium buildup rate.

ambient air: The general air in the area of interest (e.g., the general room atmosphere) as distinct from a specific stream or volume of air that may have different properties.

breathing zone air monitoring: Actions conducted to detect and quantify the radiological conditions of air from the general volume of air breathed by the worker, usually at a height of 1 to 2 meters. See *personal air monitoring*. **(Workplace Air Monitoring Chapter of the IG)**

continuous air monitor (CAM): An instrument that continuously samples and measures the levels of airborne radioactive materials on a “real-time” basis and has alarm capabilities at preset levels.

decision level (*DL, L*c): The amount of a count or a count rate or the final instrument measurement of a quantity of analyte at or above which a decision is made that the analyte is definitely present. **(ANSI, 2011b)**

decontamination: The process of removing radioactive contamination and materials from personnel, equipment or areas. **(RCS)**

detector: A device or component that produces a measurable response to ionizing radiation. **(Portable Instrument Calibration Chapter of the IG)**

DOELAP: The Department of Energy Laboratory Accreditation Program for personnel dosimetry. **(RCS)**

dose: The amount of energy deposited in body tissue due to radiation exposure. **(RCS)**

exposure: The general condition of being subjected to ionizing radiation, such as by exposure to ionizing radiation from external sources or to ionizing radiation sources inside the body. In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air. **(Internal Dosimetry Chapter of the IG)**

fissionable materials: A nuclide capable of sustaining a neutron - induced fission chain reaction (e.g., uranium-233, uranium-235, plutonium-238, plutonium 239, plutonium -241, neptunium-237, americium- 241 and curium-244) **(10 CFR 830)**.

fixed contamination: Any area with detectable removable contamination less than the removable contamination values of Appendix D of 10 CFR 835 and fixed contamination at levels that exceed the total contamination values of Appendix D of 10 CFR 835. **(Posting and Labeling Chapter of the IG)**

fixed-location sampler: An air sampler located at a fixed location in the workplace.

grab sampling: A single sample removed from the workplace air over a short time interval, typically less than one hour.

hazardous waste: Because of its quantity, concentration, and physical, chemical, or infectious characteristics, hazardous waste may cause or significantly contribute to an increase in mortality, or an increase in serious irreversible or incapacitating reversible illness; it may pose a potential hazard to human health or the environment when improperly treated, stored, transported, disposed of, or otherwise managed. **(DOE/S-0101)**

high-efficiency particulate air (HEPA) filter: Throwaway extended pleated medium dry-type filter with 1) a rigid casing enclosing the full depth of the pleats, 2) a minimum particle removal efficiency of 99.97% for thermally generated monodisperse di-octyl phlalate smoke particles with a diameter of 0.3 µm, and 3) a maximum pressure drop of 1.0 in. w.g. when clean and operated at its rated airflow capacity. **(RCS)**

HLW: High-level waste (HLW) is the material that remains following the reprocessing of spent nuclear fuel and irradiated targets from reactors. The HLW is highly radioactive and generates heat on its own. Some of its elements will remain radioactive for thousands of years. Because of this, HLW shall be managed very carefully and all handling shall be performed from behind heavy protective shielding. **(DOE/S-0101)**

intake: The amount of radionuclide taken into the body by inhalation, absorption through intact skin, injection, ingestion or through wounds. Depending on the radionuclide involved, intakes may be reported in mass (e.g., µg, mg) or activity (e.g., µCi, Bq) units. **(Internal Dosimetry Chapter of the IG)**

LLW: Low-level waste (LLW) is any radioactive waste that is not HLW, spent nuclear fuel, TRU waste, or uranium mill tailings. The LLW is typically contaminated with small amounts of radioactivity dispensed in large amounts of material. The LLW is generated in every process involving radioactive materials in the DOE including decontamination and decommissioning projects. **(DOE/S-0101)**

minimum detectable amount/activity (MDA): The smallest amount (activity or mass) of an analyte in a sample that will be detected with a probability β of non-detection (Type II error) while accepting a probability α of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error). **(ANSI N13.30-2011)**

MW: Mixed waste (MW) is waste that contains both radioactive and hazardous wastes. Any of the types of radioactive waste described can be a mixed waste if it contains any hazardous wastes. In fact, all of DOE's HLW is mixed waste because of the chemicals used to reprocess the fuel that resulted in the generation of the material or because it is suspected to contain hazardous materials. **(DOE/S-0101)**

personal air monitoring: The monitoring of air for radioactive particles in the immediate vicinity of an individual radiation worker's nose and mouth, usually by a portable sampling pump and collection tube (such as a lapel sampler) worn on the body. Personal air monitoring is a special case of breathing zone air monitoring. **(Workplace Air Monitoring Chapter of the IG)**

portable air sampler: An air sampler designed to be moved from area to area.

radiation-generating device (RDG): The collective term for devices which produce ionizing radiation, sealed sources which emit ionizing radiation, small particle accelerators used for single purpose applications which produce ionizing radiation (e.g., radiography), and electron-generating devices that produce x-rays incidentally. **(Radiation-Generating Devices Chapter of the IG)**

radioactive material: For the purposes of the standard, radioactive material includes any material, equipment or system component determined to be contaminated or suspected of being contaminated. Radioactive material also includes activated material, sealed and unsealed sources, and material that emits radiation. **(RCS)**

radiological work permit (RWP): The permit that identifies radiological conditions, establishes worker protection and monitoring requirements, and contains specific approvals for radiological work activities. The Radiological Work Permit serves as an administrative process for planning and controlling radiological work and informing the worker of the radiological conditions. **(RCS)**

radiological protection organization: A contractor organization responsible for radiation protection activities within contractor facilities. This organization is independent of the line organizational element responsible for production, operation, or research activities and should report to the contractor senior site executive. **(Sealed Source Chapter of the IG)**

real-time air monitoring: Collection and real-time analysis of the workplace atmosphere using continuous air monitors (CAMs).

refresher training: The training scheduled on the alternate year when full retraining is not completed for Radiological Worker I and Radiological Worker II personnel. **(RCS)**

removable contamination: Radioactive material that can be removed from surfaces by nondestructive means, such as casual contact, wiping, brushing or washing. **(RCS)**

representative air sampling: The sampling of airborne radioactive material in a manner such that the sample collected closely approximates both the amount of activity and the physical and chemical properties (e.g., particle size and solubility) of the aerosol to which the workers may be exposed.

sanitary waste: Sanitary waste is waste that is neither hazardous nor radioactive. **(DOE/S-0101)**

source-specific air sampling: Collection of an air sample near an actual or likely release point in a work area using fixed-location samplers or portable air samplers.

survey: An evaluation of the radiological conditions and potential hazards incident to the production, use, transfer, release, disposal, or presence of radioactive material or other sources of radiation. When appropriate, such an evaluation includes a physical survey of the location of radioactive material and measurements or calculations of levels of radiation, or concentrations or quantities of radioactive material present.

TRU: Transuranic (TRU) waste refers to waste materials containing elements with atomic numbers greater than 92. These elements are generally alpha-emitting radionuclides that decay slowly. The TRU waste contains a concentration of these elements greater than 100 nCi/g. The TRU waste is not as intensely radioactive as HLW. The TRU waste also decays slowly, requiring long-term isolation. **(DOE/S-0101)**

workplace monitoring: The measurement of radioactive material and/or direct radiation levels in areas that could be routinely occupied by workers.

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CONCLUDING MATERIAL

Review Activities:

DOE
Ops Offices

NNSA
AL
HS
CH
EM
ID
SC
NV
SA
OR
LM
RL
NE
SR
NS
PR

Area/Site Offices
Ames
Argonne
Berkeley
Brookhaven
Carlsbad
Fermi
Kansas City
Los Alamos
Oak Ridge Site Office
Princeton
SLAC
Thomas Jefferson
West Valley
Y-12

National Laboratories
Ames
ANL
BNL
FNAL
LBNL
ORNL
PPPL
SLAC
TJNAL

Preparing Activity:

DOE HS-11