# Radionuclides Related to Historical Operations at the Santa Susana Field Laboratory Area IV

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# ACRONYMS

Advanced Epithermal Thorium Reactor
boiling water reactor
California Department of Public Health
U.S. Department of Energy
Energy Technology Engineering Center
International Atomic Energy Agency
Kinetics Experiment Water Boiler
kilowatts
Nuclear Materials Development Facility
Preliminary Remediation Goal
pressurized water reactor
Sodium Reactor Experiment
Santa Susana Field Laboratory
U.S. Environmental Protection Agency

## GLOSSARY

activation product – A nuclide, usually radioactive, formed by the bombardment and adsorption in material with neutrons, protons, or other nuclear particles.

AEC – U.S. Atomic Energy Commission

corrosion product - An activation product formed by the activation and oxidation of metals

criticality – The condition in which a system is capable of sustaining a nuclear chain reaction.

DOE – U.S. Department of Energy

EIS - Environmental Impact Statement -

Environmental Assessment – A written environmental analysis which is prepared pursuant to the National Environmental Policy Act to determine whether a federal action would significantly affect the environment and thus require the preparation of a more detailed environmental impact statement.

EPA – U. S. Environmental Protection Agency

ETEC – Energy Technology Engineering Center

fertile material – Substance capable of becoming fissile, by capturing neutrons, possibly followed by radioactive decay; e.g., Th-232, U-238, Pu-240.

fission product – A nuclide resulting either from the fission of heavy elements such as uranium. Usually radioactive.

fission yield – The fraction of fissions resulting in a specified fission product.

fuel assembly – Structured collection of fuel rods or elements, the unit of fuel in a reactor.

fuel cladding – The outer metal jacket of a nuclear fuel element or target. It prevents fuel corrosion and retains fission products during reactor operation and subsequent storage, as well as providing structural support. Zirconium alloys, stainless steel, and aluminum are common cladding materials.

fuel element – Arrangement of a number of fuel rods into which the nuclear fuel is inserted in the reactor.

half-life – The time in which one-half of the atoms of a particular radioactive isotope disintegrate to another nuclear form. Half-lives vary from millionths of a second to billions of years.

high enriched uranium – Uranium enriched to at least 20% U-235.

kW – Kilowatt – A unit of electrical power equal to a thousand watts (joules per second)

low enriched uranium – Uranium enriched to less than 20% U-235. (That in power reactors is usually 3.5 - 5.0% U-235.)

MW – Megawatt – A unit of electrical power equal to a million watts (joules per second).

noble gas - Any of the chemically inert gaseous elements of the helium group in the periodic table

nuclear activation (neutron) cross section – Cross section: a measure of the probability of an interaction between a particle and a target nucleus, expressed in barns (1 barn =  $10^{-24}$  cm<sup>2</sup>).

nuclear reactor – A device in which a fission chain reaction can be initiated, maintained, and controlled. It's essential components are fissionable fuel, moderator, shielding, control rods, and coolant.

nuclide – Elemental matter made up of atoms with identical nuclei, therefore with the same atomic number and the same mass number (equal to the sum of the number of protons and neutrons). Also referred to as an isotope of an element.

Particle Accelerator – A scientific instrument that increases the kinetic energy of charged particles

PRG – Preliminary remediation goal

radioactive decay – The change of one radioactive nuclide into a different nuclide by the spontaneous emission of alpha, beta, or gamma rays, or by electron capture. The end product is a less energetic, more stable nucleus. Each decay process has a definite half-life.

radionuclide – An unstable nuclide that undergoes spontaneous transformation, emitting radiation.

Risk – The probability of a detrimental effect of exposure to a hazard.

risk assessment – The science of studying the amount of risk associated with doing something.

site characterization – A general term applied to the investigation activities at a specific location that examine natural phenomena and human-induced conditions

spent fuel – Used fuel assemblies removed from a reactor after several years use and treated as waste.

spontaneously fissionable (fissile) material – Substance in which fission occurs spontaneously, not induced by an incident particle.

SSFL - Santa Susana Field Laboratory

transuranic –Refers to any element whose atomic number is higher than that of uranium (atomic number 92), including neptunium, plutonium, americium, and curium. All transuranic elements are produced artificially and are radioactive.

unirradiated – Not having been exposed to radiation

Van de Graaff Generator – Electrical device that produces a high voltage by building up a charge of static electricity

# **EXECUTIVE SUMMARY**

This paper summarizes the history of nuclear operations at Area IV of the Santa Susana Field Laboratory (SSFL) and identifies the radionuclides that would result from documented processes. It also presents the results of calculations estimating which radionuclides could still be present in 2009 in significant concentrations. These calculations are based on the initial production amounts of each radionuclide and the radioactive decay that has occurred since last production. The calculations also evaluate the proportion each radionuclide would contribute to the overall potential dose and associated risk in SSFL Area IV.

The U.S. Department of Energy (DOE) commissioned this research because the California Department of Public Health (CDPH), other regulators, and stakeholders asked, in various ways based on a 2008 analysis of existing data, the question: Based on existing knowledge of SSFL Area IV nuclear operations and documented radioactive decay rates, what radionuclides might still be present in significant quantities today? In response, this paper was prepared using the best available references describing the history of radiological operations, processes, and uses. As the research progressed, DOE recognized that this paper could be used as a reference point not only to respond to the various questions CDPH and others had asked, but also to meet two additional objectives: 1) to ensure that all significant contributors to risk are included in the planned Risk Assessment, and 2) to help prioritize analytical requirements for new samples to be collected for pending studies.

This paper does not evaluate the actual concentrations remaining in the environment in Area IV of the SSFL or the actual dose. The actual concentrations present in Area IV of SSFL and resulting risk to the public depend on the quantities of radionuclides that were released to the environment and the residual persistence in the environs after previous remediation efforts. These questions will be the focus of the radiological survey and sampling efforts currently in planning by the U.S. Environmental Protection Agency (USEPA). Rather, this paper documents the potential that each identified radionuclide could be present in significant quantities today, and in what proportion, relative to the other radionuclides identified based on the initial production rates and the rate at which each radionuclide decays. In other words, this paper provides a list of "suspects" – radionuclides that both were in the area and are known to have relatively long half-lives. DOE offers this list so the teams of scientists investigating contamination can prioritize their sampling and analyses toward these radionuclides. Following completion of the EPA's radiological survey and a chemical survey being conducted by the California Department of Toxic Substances Control, DOE will prepare human health and ecological risk assessments to support of the Environmental Impact Statement currently being developed.

The majority of process related radionuclides at the SSFL Area IV resulted from the following activities:

- operation of ten nuclear reactors,
- operation of seven criticality test facilities,
- manufacture of reactor fuel assemblies,
- disassembly and inspection of reactors and used reactor fuel assemblies,
- preparation of radioactive material for disposal, and
- on-site storage of nuclear materia1.

In addition to those operations, smaller quantities of radionuclides were associated with small-scale laboratory work that included the following activities:

• fabrication, use, and storage of radioactive sources;

- research focused on reprocessing spent nuclear fuel;
- operation of particle accelerators;
- research using radioisotopes; and
- miscellaneous operations and commercial items that used radioactive materials.

In compiling the list of radionuclides that would have been produced as a result of the documented reactor processes, radionuclides with half-lives of less than one year were not considered. The rationale for this is that only 0.0000002 percent or less of the original radioactivity would remain after the 29 years (the time that has elapsed since the last reactor was shut down on the SSFL Area IV, in 1979 and the production of radionuclides ceased). Standard references were used to calculate the relative quantities of fission products, activation products, and transuranic radionuclides in spent reactor fuel that were produced in reactors. These quantities were then adjusted to account for radioactive decay since cessation of reactor operations. The identity and amounts of accelerator and research related radionuclides were also qualitatively evaluated.

This evaluation has resulted in a list of radionuclides that are expected to have the potential to contribute more than 1 percent of the current activity. They are:

### **Reactor fuel-related radionuclides:**

- Two isotopes of thorium Th-228, Th-232;
- Three isotopes of uranium U-234, U-235, U-238;
- Four isotopes of plutonium Pu-238, Pu-239, Pu-240, Pu-241;

## **Reactor fission products:**

- tritium (H-3, an isotope of hydrogen,),
- strontium-90 (Sr-90),
- cesium-137 (Cs-137),
- promethium-147 (Pm-147), and
- europium-155 (Eu-155);

## **Reactor activation products:**

- beryllium-10 (Be-10),
- cadmium-113m (Cd-113m),
- barium-133
- two isotopes of europium Eu-152 and Eu-154, and
- uranium-233 (U-233);

## **Reactor-produced transuranics:**

- Four isotopes of plutonium Pu-238, Pu-239, Pu-240, Pu-241;
- Americium-241 Am-241; and
- Curium-244 Cm-244;

## Accelerator activation product:

• tritium (H-3); and

### **Research-related radionuclides:**

- Three isotopes of uranium U-234, U-235, U-238;
- Neptunium-237 (Np-237);
- Four isotopes of plutonium Pu-238, Pu-239, Pu-240, Pu-241;
- Cobalt-60 (Co-60);
- Cesium-137 (Cs-137); and
- Promethium-147 (Pm-147).

This paper also presents the results of calculations to evaluate the risk these radionuclides could present. The radionuclides that contribute greater than 1 percent of the risk potential are the same as those listed above, except that reactor produced fission products Pm-147 and Eu-155, reactor produced activation product Co-60, and all reactor produced transuranics are not included in the list that produce 99% of the risk. However, some of these are still included as fuel or research related risk contributors. A combined priority list based on the potential to contribute to greater than 1% of the risk include: Th-228, Th-232, U-234, U-235, U-238, Pu-239, Pu-240, Pu-241, H-3, Sr-90, Ba-133, Cs-137; Be-10, Cd-113m, Eu-152, Eu-154, U-233, Np-237, Co-60, Cs-137, and Pm-147.

The calculation of relative activity, dose and risk percentage could be quantitatively performed only for radionuclides in spent reactor fuel, based on the available references for relative production amounts. However, radionuclides associated with fuel elements, used in the manufacture of fuel assemblies, are assumed to have the potential to contribute more than 1 percent of the current activity, dose, and risk. Based on this review, radionuclides in spent nuclear fuel and fuel elements are expected to be responsible for the majority of SSFL Area IV contamination. Information about specific concentrations, quantities, and processes for radionuclides from research was not available. Thus, no relative activity information is available and therefore the relative potential for contamination is unknown. As a conservative precaution, these radionuclides were included in the list above, for further investigation.

## 1.0 BACKGROUND AND PURPOSE

The U.S. Department of Energy (DOE) commissioned this paper to summarize the history of nuclear operations at Area IV of the Santa Susana Field Laboratory (SSFL) and identify the radionuclides that would result from those operations. DOE asked that the author also present the results of calculations to estimate which radionuclides could still be present in 2009 in significant concentrations. This paper provides these calculations based on the initial production amounts of each radionuclide relative to all others and the radioactive decay that would have occurred since production. The calculations also evaluate how much each radionuclide would contribute to the overall potential dose and associated risk that might exist at SSFL Area IV.

DOE initiated this paper after receiving comments on the *Draft Gap Analysis Report*, submitted for regulatory and public review on June 1, 2008. That analysis was conducted to determine whether existing data for Area IV of the SSFL are adequate for the purpose of developing and evaluating risk-based cleanup alternatives in an Environmental Impact Statement. Section 3.2.5.3 and Table 3-11 of the report included a list of radionuclide contaminants of interest developed from the radionuclide lists contained in the *Historical Site Assessment*<sup>1</sup> and the *Environmental Assessment*<sup>2</sup>, supplemented by additional radionuclides based on process knowledge from other reactor and fuel separation facilities, and filtered on the basis of half-life considerations.

In his review comments, Jerry Hensley of the California Department of Public Health-requested that a new listing of all radionuclides generated during reactor operations be provided and pared down using industry acceptable methods (i.e., radiological half-life). A number of other reviewers from both regulatory agencies and the public also requested more information about radionuclides related to all processes and their current significance, while taking into consideration half-life and health affects. This information is needed not only to make sure that all significant contributors to risk are included in the Risk Assessment, but also to aid in prioritization of target analytes for new sampling and analysis efforts. This paper has been drafted to respond to these comments and requests and to meet these needs.

## 2.0 SCOPE AND APPROACH

This paper provides a review and an evaluation of the best available references describing the historical radiological operations related to nuclear reactor research conducted by DOE and its predecessor, the Atomic Energy Commission. This review also includes other nuclear research operations conducted in Area IV of the SSFL. While it is understood that the process descriptions in these references may not be exhaustive, it is believed that they represent the types of radiological processes that were performed in Area IV of the SSFL and include all significant radionuclides that were used or produced. DOE plans to compile further records and information related to all activities at the Energy Technology Engineering Center (ETEC). As part of that effort, former workers and all stakeholders will be asked to provide any

<sup>&</sup>lt;sup>1</sup> Historical Site Assessment of Area IV Santa Susana Field Laboratory, Sapere Consulting, Inc. and The Boeing Company for the Department of Energy Under Contract DE-AC03-99SF21530 (May 2005).

http://etec.energy.gov/Cleanup/Historical-Site-Assessment.html

<sup>&</sup>lt;sup>2</sup> Environmental Assessment for Cleanup and Closure of the Energy Technology and Engineering Center, Final. U.S. Department of Energy, NNSA Service Center, Oakland, CA (March 2003). http://etec.energy.gov/Regulation/RegDocs/ETECEA.pdf

information that they may have on the history of the site. If additional information is collected that changes the conclusions of this paper, it will be revised accordingly.

This paper provides a summary of the historical nuclear operations and a list of the associated processrelated radionuclides produced at Area IV of the SSFL. It also includes an evaluation of the probability for each radionuclide to be present at the current time in significant concentrations. The potential for these radionuclides to be present today has been evaluated based on the initial relative production amounts and radioactive decay since their production. The significance of each radionuclide's contribution to the potential dose and associated risk is also evaluated.

This paper does not evaluate the actual concentrations remaining in the environment in Area IV of the SSFL or the actual dose, but only the potential for the current likely presence of each radionuclide relative to all others. The actual concentrations present in Area IV of SSFL and resulting risk to the public depend on the quantities of radionuclides that were released to the environment and the residual persistence in the environs after previous remediation efforts. That subject will be the focus of the radiological survey and sampling efforts currently in planning by the U.S. Environmental Protection Agency (USEPA) and the dose/risk assessment aspect of the EIS that DOE will complete following the USEPA's radiological survey and a chemical survey being conducted by the California Department of Toxic Substances Control.

# 3.0 SUMMARY OF NUCLEAR OPERATIONS AT SSFL AREA IV

Process-related radionuclides at the SSFL Area IV were primarily the result of the following activities:

- Operation of ten nuclear reactors
- Operation of seven criticality test facilities
- Manufacture of reactor fuel assemblies
- Disassembly and inspection of reactors and used reactor fuel assemblies
- Preparation of radioactive material for disposal
- On-site storage of nuclear material.

Small-scale laboratory work, including the activities below, may have involved smaller amounts of radionuclides:

- Fabrication, use, and storage of radioactive sources
- Research on reprocessing used reactor fuel
- Operation of particle accelerators
- Research using radioisotopes
- Miscellaneous operations
- Commercial items which use radioactive materials.

# 4.0 SSFL AREA IV NUCLEAR OPERATIONAL HISTORY

## 4.1 REACTOR OPERATIONAL HISTORY<sup>3</sup>

The majority of man-made radioactivity at the SSFL Area IV is the result of the operation of ten nuclear reactors. The amount of radioactivity generated by a nuclear reactor depends largely on the amount of heat it generates, called its "power level" and the period of operation. The reactors operated at the SSFL Area IV all had very low power levels: six had power levels of less than 100 kilowatts (kW), three had power levels of 600 to 1,000 kW, and one (the Sodium Reactor Experiment, or SRE) was a 20-MW test reactor. By comparison, reactors used for commercial electric power generation have thermal power levels of 3,000 MW or more. The reactors were operated in seven different facilities. Table 1 lists the name, facility number, facility name, nominal power level, and operating period for each reactor.

Operation	Bld.		<b>Power Level</b>	
Name	No.	Facility Name	( <b>kW</b> )	<b>Operating Period</b>
KEWB	4073	Kinetics Experiment Water Boiler	1	7/56 - 11/66
L-85/AE-6	4093	L-85 Nuclear Experiment Reactor	3	11/56 - 2/80
SRE	4143	Sodium Reactor Experiment	20,000	4/57 - 2/64
SER	4010	S8ER Test Facility	50	9/59 - 12/60
S2DR	4024	SNAP Environmental Test Facility	65	4/61 - 12/62
STR	4028	Shield Test Irradiation Facility	50	12/61 - 7/64
S8ER	4010	S8ER Test Facility	600	5/63 - 4/65
STIR	4028	Shield Test Irradiation Facility	1,000	8/64 - 6/73
SI0FS3	4024	SNAP Environmental Test Facility	37	1/65 - 3/66
S8DR	4059	SNAP Ground Prototype Test Facility	619	5/68 - 12/69

 Table 1. Reactor Operations at the SSFL Area IV

A nuclear reactor contains nuclear fuel, usually in the form of fuel assemblies, composed of spontaneously fissionable (fissile) radioactive material (e.g., uranium-235 or plutonium-239) plus other materials that may be added for various purposes (e.g., thorium as a fertile material for breeding fissile uranium-233), contained within a cladding material (usually steel, aluminum, or zirconium). The fuel assemblies are arranged in a "core", and surrounded by reflectors, shields, and containment vessels. Two reactors that operated at the SSFL Area IV, the Kinetics Experiment Water Boiler (KEWB) and the L-85 Nuclear Experimentation Reactor, had fuel in the form of liquid solutions of uranyl sulfate.

Several incidents occurred during the operating history of the SRE reactor that resulted or may have resulted in the releases of radionuclides to the environment.<sup>4</sup> On June 4, 1959, an explosion resulting from an unexpected hydrogen-oxygen reaction blew a fuel element (which was undergoing sodium cleaning) out of the wash cell. On July 12, 1959, depletion in coolant flow due to blockage resulted in overheating and damage to 13 of 43 fuel elements in the reactor core. During this event, the damage to these assemblies caused failure of cladding on all seven fuel rod elements, and some iron uranium

<sup>3</sup> Largely abstracted from *Nuclear Operations at Rockwell's Santa Susana Field Laboratory – A Factual Perspective,* Rockwell International, Report N001E1R000017, October 2, 1990, Revision B. http://www.etec.energy.gov/library/Reading-Room/N001ER000017 Nuclear Operations SSFL

Factual Perspective.pdf.

<sup>4</sup> Historical Site Assessment of Area IV Santa Susana Field Laboratory, Sapere Consulting, Inc. and The Boeing Company, for the U.S. Department of Energy, Ventura County, CA, Contract DE-AC03-99SF21530, May 2005.

eutectic was molten for a short period of time in the reactor. Between 5,000 and 10,000 curies of mixed fission product were released and contained in the primary sodium cooling system. It was calculated that approximately 28 curies of Kr-85 were released to the environment. The Kr-85, a non-reactive noble gas, quickly dispersed in the atmosphere. On March 12, 1960, a contaminated sodium fire broke out in the sodium service vault. Personnel were unable to extinguish the fire with standard suppression equipment, so the vault was sealed and purged with argon gas. Accidents such as these and others likely resulted in accidental release of radionuclides to the environment likely contributed to the overall amount of contamination that may be present. However, they in general do not impact the identity or relative ratios of radionuclides to each other that may currently be present in the environment, which is the subject of this paper.

## 4.2 CRITICALITY TEST FACILITY OPERATIONAL HISTORY<sup>5</sup>

A controlled nuclear chain reaction can be sustained only when neutrons generated by fission of reactor fuel balance the neutrons used up and lost. When the reactor is adjusted so that this balance is achieved, it is said to be "critical". Criticality can be achieved in several ways: for example, by bringing parts of a core of fissionable material together (to reduce the number of neutrons that escape); by removing control rods (to reduce the number of neutrons captured in the control rods); or by increasing the number of neutrons reflected back into the core. Criticality Test Facilities were built to conduct tests to aid in developing new types of reactors by determining exactly which reactor configurations are critical, and how criticality is affected by changes in reactor design parameters. The criticality tests were operated at a very low power level (up to a few hundred watts), and neutron levels were correspondingly very low. Thus, a large number of criticality tests could be performed in the same test facility without generating much by-product radioactivity.

There were dozens of criticality tests performed at the SSFL Area IV, in seven different test facilities. Table 2 lists these facilities, their facility number, and operating periods.

	Bldg.	Operating	
Facility Name	No.	Period	Notes
SNAP Critical Test	4373	1957-63	First SNAP-2 Criticality Tests
Organic Moderated Reactor	4009	1958-67	Basic Tests of Reactor Concept
Sodium Graphite Reactor	4009	1958-67	Basic Tests of Reactor Concept
SNAP Critical Equipment Lab.	4012	1961-71	Later SNAP Criticality Tests
Fast Critical Experiment Lab.	4100	1961-74	Started as AETR
SNAP Flight System	4019	1962	SNAP Flight System Criticality
SNAP Transient Test	4024	1967-69	SNAP Transient Response Tests

 Table 2. Criticality Test Facilities at the SSFL Area IV

#### 4.3 MANUFACTURE OF REACTOR FUEL ASSEMBLY OPERATIONAL HISTORY<sup>6</sup>

As part of the nuclear reactor development work performed for the government, three different reactor fuel manufacturing operations were performed at the SSFL Area IV. The first operation was the assembly of fuel elements for the SRE. The second operation was manufacture of plutonium fuel, and the third was

<sup>6</sup> Ibid.

<sup>&</sup>lt;sup>5</sup> Nuclear Operations at Rockwell's Santa Susana Field Laboratory - A Factual Perspective.

manufacture of uranium carbide fuel. There was also a Fuel Storage Facility, which stored the special nuclear materials (enriched uranium and plutonium) used to make reactor fuel.

The SRE fuel elements were assembled in the Engineering Test Building (Building 4003). Uranium and thorium metal slugs were brought into the SSFL Area IV for this purpose. In Building 4003, the slugs were loaded into metal tubes, the interstices were filled with sodium metal, and the tubes sealed. Although fuel elements for three cores for the SRE were prepared, only two cores were used. The third core was eventually shipped off-site. The first core did not contain thorium.

The plutonium fuel manufacturing facility, named the Nuclear Materials Development Facility (NMDF; Building 4055) was built specifically for development work involving plutonium. It was completed in 1967 and operated until 1979. Its operating history is summarized in Table 3.

<b>Operating Period</b>	Operation
1967 – 1968	Development of Analysis Technologies for uranium-plutonium oxide fuels
4/68 - 6/69	Recycle of scrap uranium-plutonium fuel
7/68 - 6/70	Development of technologies to mix tungsten into uranium-plutonium carbide fuel
4/70 - 9/70	Preparation of samples for uranium-plutonium oxide irradiation studies
9/70 - 3/74	Idle
1974 – 1975	Bench scale tests-recovery of plutonium from simulated waste
1975 – 5/77	Mixed uranium-plutonium carbide fuel fabrication
5/77 – 11/78	Partial decontamination and clean-up
11/78 – 11/79	Fabrication of depleted uranium carbide fuel
11/79 - 10/82	Idle
10/82 - 10/86	Decontamination and decommissioning
7/87	Released for unrestricted use

 Table 3. Operations at the Nuclear Materials Development Facility

The uranium carbide fuel manufacturing pilot plant was located in Building 4005. It was a small scale production facility built to study the operations associated with manufacturing reactor fuel assemblies from uranium carbide. In the pilot plant, uranium oxide was reacted with graphite to convert it to uranium carbide, which was then cast into pellets, machined to the proper dimensions, and assembled into cladding tubes to make fuel assemblies. Initial operations were performed using depleted uranium to test the equipment, and then enriched uranium was used to make fuel assemblies for a critical assembly to be built at another AEC facility. Operations were completed in about nine months in 1967 and production was small.

The plutonium and uranium carbide materials were stored in the Fuel Storage Facility (Building 4064), a vault built to provide secure storage for fissile fuel material (enriched uranium and plutonium) used to make reactor fuel. The building was constructed above ground using concrete and concrete blocks, to comply with the AEC criteria for vaults for storage of fissionable materials.

## 4.4 DISASSEMBLY AND EXAMINATION OF REACTORS AND USED REACTOR FUEL ASSEMBLIES OPERATIONAL HISTORY<sup>7</sup>

A number of operations were performed remotely in the heavily-shielded Hot Laboratory (Building 4020, the "Hot Lab"), built in Area IV specifically to provide protection during processes that involved highly radioactive materials. These operations included:

- **Fuel assembly performance evaluation.** During reactor test operations, it was often necessary to examine reactor fuel assemblies and other test specimens to evaluate their performance, which involved the handling and examination of highly radioactive items.
- **Reactor disassembly.** When each reactor operation was completed and the reactor was no longer needed, it was removed from its operating location, disassembled, its fuel removed, its radioactive structure cut into pieces suitable for shipment. The radioactive material was shipped off-site for appropriate processing and disposal. Given the highly radioactive materials involved, the disassembly, fuel removal, and size reduction operations were also usually performed using the Hot Lab.
- Used fuel disassembly. The Hot Lab was also the location for work involving disassembly of used irradiated reactor fuel generated outside the SSFL. The fuel elements were shipped into the Hot Lab, disassembled or separated from their cladding material, and the separated materials then packaged and shipped back to other DOE facilities.
- **Manufacture of sealed sources.** The Hot Lab was also used to manufacture sealed radioactive sources (see below), for performing leak testing of sources, and for cutting and machining operations involving radioactive cobalt-60.

Construction of the Hot Lab facility was completed in 1959, and it was in use until 1989 when it underwent decontamination and decommissioning. The Hot Lab was used to examine fuel and/or components from the SRE, SER, S2DR, S8ER, S8DR, and S10FS3 reactors operated at the SSFL Area IV, the OMR and SGR criticality test facilities at SSFL, and the Piqua, Ohio, reactor. It was also used to declad fuel from the, EBR-I, EBR-11, Hallam, Fermi, and SEFOR reactors from other DOE sites.

# 4.5 FABRICATION, USE, AND STORAGE OF RADIOACTIVE SOURCES OPERATIONAL HISTORY<sup>8</sup>

A variety of radiation sources were used in the Hot Lab, including sources essential for the calibration of the many instruments required to detect and measure radioactivity at SSFL Area IV. These instruments were calibrated periodically, using known quantities and types of radioactivity. The calibration "sources" produced in the Hot Lab consisted of sealed containers that contained small measured quantities of radioisotopes. Other sources produced in the Hot Lab included those for radiography, irradiation testing, and other applications. The sources manufactured in the Hot Lab were used in various facilities at the SSFL Area IV and elsewhere. Only a small number of commercially-produced calibration sources are currently in use at SSFL today. Although approximately 140,000 curies of radioactive material (primarily promethium-147) were fabricated into sources at the Hot Lab, only a small fraction of this activity would remain today due to radioactive decay of the sources produced, especially for the promethium sources.

<sup>&</sup>lt;sup>7</sup> Ibid.

<sup>&</sup>lt;sup>8</sup> Ibid.

## 4.6 RESEARCH ON REPROCESSING USED REACTOR FUEL OPERATIONAL HISTORY9

The used fuel assemblies from nuclear reactors contain unused fuel materials, and fission and activation products. Rockwell International developed a process to effect a partial separation of used fuel, removing part of the fission products so that the material could be used again as reactor fuel. Tests were performed in a well-shielded "Hot Cave" located in Building 4003, the Engineering Test Building. These experiments used up to one-kilogram quantities of un-irradiated uranium and thorium, and up to 100-gram quantities of highly irradiated materials.

## 4.7 PARTICLE ACCELERATORS OPERATIONAL HISTORY<sup>10</sup>

Another way to generate artificial (induced) radioactivity is to bombard a target material with atomic particles that have been accelerated to high speeds by means of a particle accelerator. A common form of particle accelerator is a "Van de Graaff generator", which uses a high-voltage electrostatic field to accelerate atomic particles to high speeds (high energy levels). Collisions of these particles with a target material (such as aluminum or tritium) can generate small amounts of radioactivity. Rockwell International operated a Van de Graaff generator in Building 4030, which bombarded tritium targets with deuterons to produce neutrons. The neutrons produced could then make other materials radioactive. A second Van de Graaff generator was operated at the SRE facility, generating neutrons for neutron activation analyses of materials. It was removed before the SRE facility was decontaminated and decommissioned.

## 4.8 RESEARCH USING RADIOISOTOPES OPERATIONAL HISTORY<sup>11</sup>

Some of the research done at the SSFL Area IV required the use of special radioisotopes. For these tests, small quantities of specially-prepared radioisotopes were brought to the SSFL Area IV, used in laboratories under controlled conditions, and then either transported off the facility or stored under controlled conditions if reuse was required.

One research program that required the use of radioisotopes was the TRUMP-S program. Although the original plan was to perform the TRUMP-S tests in the Hot Lab at the SSFL Area IV, the test program was transferred to the University of Missouri. Seventy-five grams of depleted uranium, five grams of plutonium, and four grams of neptunium, were received at SSFL Area IV and stored in Building 4064 before being shipped to the University of Missouri sometime after 1990.

Another research program that used a radioisotope was a corrosion test program carried out in the Corrosion Testing Laboratory (Building 4023). A pumped sodium corrosion test loop was built there, and used to study the deposition behavior of activation products (Mn-54 and Co-60) in flowing sodium so as to develop more effective traps for these isotopes. An activated piece of fuel cladding containing these isotopes was used in these tests.

<sup>&</sup>lt;sup>9</sup> Ibid.

<sup>&</sup>lt;sup>10</sup> Ibid.

<sup>&</sup>lt;sup>11</sup> Ibid.

# 5.0 RADIONUCLIDES ASSOCIATED WITH IDENTIFIED OPERATIONS AT SSFL AREA IV

## 5.1 RADIONUCLIDES ASSOCIATED WITH REACTOR OPERATIONS, CRITICALITY TEST OPERATIONS, MANUFACTURE OF NEW AND DISASSEMBLY OF USED FUEL ASSEMBLIES, AND WASTE OPERATIONS

Operation of a nuclear reactor not only involves the use of nuclear fuel but also creates three types of byproduct radionuclides: fission products, transuranics, and activation products. When part of the fissionable material in the fuel element is used up, or when a reactor is decommissioned, the fuel elements are removed from the reactor. These "spent" fuel elements contain the fission products and transuranics generated by operation of the reactor, and the activation products in the cladding. The same radionuclides associated with reactor operations are also associated with criticality test operations, disassembly of used fuel assemblies, and waste operations since they all are related to nuclear reactor operations either as new fuel or fuel with the operational by-products.

## 5.1.1 Reactor Fuel Radionuclides

The radioactive material placed in a reactor includes the nuclear fuel made up of fissile radioactive material (e.g., uranium, plutonium) and other fuel elements (thorium). Although plutonium fuel was manufactured at SSFL Area IV, according to operational descriptions, none of the reactors or criticality experiments operated at SSFL Area IV used plutonium as a fuel. Many of the reactors, including the SRE, which produced the vast majority of the power generated, used a low-enriched uranium fuel (3-5 percent U-235 content by mass). However, according to operational descriptions, all reactors and criticality experiments associated with the SNAP program used high-enriched uranium fuel (~90 percent U-235 content by mass).<sup>12</sup>

The majority of the radioactivity in unirradiated low-enriched uranium fuel is from the isotope U-234 (75-85 percent) with lesser amounts from U-238 (10-20 percent) and only a few percent from U-235 (~5 percent).<sup>13</sup> In high-enriched unirradiated uranium fuel, even more of the radioactivity is from U-234 (~95 percent) while very little is from U-238 (less than 1 percent).<sup>14</sup> Sometimes thorium was added as a fertile material for breeding fissile U-233. According to operational descriptions, only two reactors, the Sodium Reactor Experiment (SRE) and the Advanced Epithermal Thorium Reactor (AETR), used fuel that contained thorium as a fertile material.<sup>15</sup> The thorium isotopes present in the fuel would have consisted mainly of Th-232 and its decay progeny Th-228 in partial equilibrium.

The manufactured plutonium fuel was actually a mixture of plutonium and uranium oxide pellets. The plutonium would have been comprised primarily of the isotope Pu-239, with lesser amounts of Pu-238, Pu-240, and Pu-241. There is no evidence that the plutonium fuel or plutonium-uranium mixed fuel was ever used as fuel for any onsite reactors.<sup>16</sup>

<sup>&</sup>lt;sup>12</sup> Ibid.

<sup>&</sup>lt;sup>13</sup> Health Physics Manual of Good Practices for Uranium Facilities, Idaho National Engineering Laboratory, EGG-2530 UC-41, June 1988, pp 2-10 and Guide of Good Practices for Occupational Radiological Protection In Uranium Facilities, U.S Department of Energy, DOE-STD-1136-2000, August 2000, pp 2-10.

<sup>&</sup>lt;sup>14</sup> Ibid.

<sup>&</sup>lt;sup>15</sup> Nuclear Operations at Rockwell's Santa Susana Field Laboratory - A Factual Perspective.

<sup>&</sup>lt;sup>16</sup> Ibid.

Table 4 provides a summary of fuel element radionuclides that were used or produced in SSFL Area IV and thus could be potentially present today.

Radionuclide	Half-life
Th-228	1.91 y
Th-232	14.0 by
U-234	246 ky
U-235	704 my
U-238	4.47 by
Pu-238	87.7 y
Pu-239	24.1 ky
Pu-240	6.56 ky
Pu-241	14.4 y

#### Table 4. Summary of Fuel Element Radionuclides

y = years; k = thousand; m = million; b = billion

## 5.1.2 Fission Product Radionuclides

When a reactor is operated, atoms of the fissionable material split, releasing neutrons and heat, and leaving behind fragments of the atom called fission products, which are made up of various isotopes of newly-formed elements. Some of the neutrons that are released are captured by other atoms of fissionable material, and these capture reactions cause some of these atoms to split, releasing more neutrons and heat and creating more fission products in a controlled "chain reaction."

Most of the fission products from a nuclear reactor are radioactive, emitting beta and gamma radiation. The fission products and fission yields can be identified by review of a Chart of the Nuclides.<sup>17</sup> Fission yields vary by radionuclide in a predictable way as demonstrated by Figure 1. The fission yields vary depending on the fuel used and are identified separately in the chart for U-235 fission products and U-233 fission products. All of the fissile fuel utilized at SSFL Area IV was U-235; however, since thorium was utilized as a fertile fuel element in some reactor experiments, some fission products from U-233 fission may have also been produced. Tables 5 and 6 list the fission product radionuclides with half-lives longer than one year along with their fission yields for U-235 and U-233 fuel, respectively.

A one-year cut-off was used since the decay of radionuclides is exponentially proportional to their halflife. After 10 half-lives, the fraction of original radioactivity remaining is reduced by decay to 0.1 percent. After 20 half-lives it is reduced to 0.0001 percent of the original amount and after 30 half-lives, it is reduced to 0.0000001 percent of the original amount. After 10 half-lives of any radionuclide, the radioactivity is usually considered to be insignificant. However, to be conservative, any radionuclide with a half-life greater than one year was included in the potential list. Any radionuclide with a half-life of less than one year will only have 0.0000002 percent or less of the original activity remaining after 29 years that have elapsed since the last reactor was shut down on the SSFL Area IV and the production of fission products ceased.

<sup>&</sup>lt;sup>17</sup> Nuclides and Isotopes, Fourteenth Edition, General Electric Company, 1989.



Figure 1. Fission yields vary by radionuclide in a predictable way

Radionuclida	onuclide tra (years) Vield <sup>a</sup> (%)		Relative	Activity Vield (%)	Relative Activity	Activity %
So 70	$t_{1/2}$ (years)	1100(70)	Activity Tielu	1 leiu (76)	2 77E 05	
Se-79	0.30E+04	0.044	4.09E-07	0.0000	3.//E-03	0.0005
Kr-85	10.73	1.317	8.51E-02	6.8337	0.00E+00	0.0000
Sr-90	29.1	5.8	1.38E-01	11.0970	6.88E+00	47.9597
Zr-93	1.50E+06	6.37	2.94E-06	0.0002	2.36E-04	0.0016
Tc-99	2.13E+05	6.1	1.99E-05	0.0016	1.59E-03	0.0111
Ru-106	1.02E+00	0.401	2.73E-01	21.8884	2.56E-05	0.0002
Pd-107	6.50E+06	0.145	1.55E-08	0.0000	1.24E-06	0.0000
Cd-113	9.00E+15	0.015	1.16E-18	0.0000	9.28E-17	0.0000
Sb-125	2.76E+00	0.031	7.79E-03	0.6258	4.01E-03	0.0280
Sn-126	1.00E+05	0.059	4.09E-07	0.0000	3.28E-05	0.0002
I-129	1.57E+07	0.75	3.31E-08	0.0000	2.66E-06	0.0000
Cs-134	2.07E+00	7.66E-06	2.57E-06	0.0002	2.43E-07	0.0000
Cs-135	2.30E+06	6.54	1.97E-06	0.0002	1.58E-04	0.0011
Cs-137	3.02E+01	6.19	1.42E-01	11.4231	7.20E+00	50.2146
Sm-146	1.03E+08	3	2.02E-08	0.0000	1.62E-06	0.0000
Pm-147	2.62E+00	2.25	5.94E-01	47.7515	2.36E-01	1.6461
Eu-152	1.35E+01	1.79E-10	9.20E-12	0.0000	2.63E-10	0.0000
Eu-154	8.59E+00	1.91E-07	1.54E-08	0.0000	2.45E-07	0.0000
Eu-155	4.71E+00	0.032	4.71E-03	0.3783	1.96E-02	0.1371
		High	nlighted	<u>9</u> 9		100

Table 5. U-235 Fission Products with Half-Lives Greater Than One Year

<sup>*a*</sup>All fission yields taken from *Nuclides and Isotopes*, Fourteenth Edition, General Electric Company, 1989, except for Cs-134, Eu-152, and Eu-154 which came from *Fission Product Yields*, WIMS Library Update Project, NAPC Nuclear Data Section, IAEA, since these isotopes are shielded from other fission products in the same mass fission decay chain.

 $t_{1/2} =$  half-life

Radionuclide	$t_{1/2}$ (years)	Fission Yield <sup>a</sup> (%)	Relative Activity Yield	Activity Yield (%)	Relative Activity at 29 Years	Activity % at 29 Years
Se-79	6.50E+04	0.14	1.49E-06	0.0001	1.32E-04	0.0007
Kr-85	10.73	2.25	1.45E-01	12.8995	0.00E+00	0.0000
Sr-90	29.1	6.8	1.62E-01	14.3750	8.91E+00	49.7785
Zr-93	1.50E+06	6.37	2.94E-06	0.0003	2.61E-04	0.0015
Tc-99	2.13E+05	4.9	1.59E-05	0.0014	1.42E-03	0.0079
Ru-106	1.02E+00	0.25	1.70E-01	15.0775	1.77E-05	0.0001
Pd-107	6.50E+06	0.114	1.22E-08	0.0000	1.08E-06	0.0000
Cd-113	9.00E+15	0.019	1.46E-18	0.0000	1.30E-16	0.0000
Sb-125	2.76E+00	0.12	3.02E-02	2.6766	1.71E-02	0.0958
Sn-126	1.00E+05	0.22	1.52E-06	0.0001	1.35E-04	0.0008
I-129	1.57E+07	1.6	7.06E-08	0.0000	6.27E-06	0.0000
Cs-134	2.07E+00	2.69E-04	9.02E-05	0.0080	9.41E-06	0.0001
Cs-135	2.30E+06	6.3	1.90E-06	0.0002	1.69E-04	0.0009
Cs-137	3.02E+01	6.81	1.56E-01	13.8855	8.75E+00	48.9070
Sm-146	1.03E+08	2.56	1.72E-08	0.0000	1.53E-06	0.0000
Pm-147	2.62E+00	1.74	4.60E-01	40.8014	2.02E-01	1.1270
Eu-152	1.35E+01	7.70E-09	3.96E-10	0.0000	1.25E-08	0.0000
Eu-154	8.59E+00	2.10E-06	1.69E-07	0.0000	2.97E-06	0.0000
Eu-155	4.71E+00	0.021	3.09E-03	0.2743	1.42E-02	0.0796
		Hig	hlighted	100		100

Table 6. U-233 Fission Products with Half-Lives Greater Than One Year

<sup>*a*</sup>All fission yields taken from *Nuclides and Isotopes*, Fourteenth Edition, General Electric Company, 1989, except for Cs-134, Eu-152, and Eu-154 which came from *Fission Product Yields*, WIMS Library Update Project, NAPC Nuclear Data Section, IAEA, since these isotopes are shielded from other fission products in the same mass fission decay chain.  $t_{1/2} = half-life$ 

The fission yield is the fraction of fissions resulting in the specified isotope and is shown in the table in percentages. Since the radioactive decay of an isotope is proportional to  $\ln(2)/t_{\nu_2}$  (where  $t_{\nu_2}$  is the radionuclide half-life), the relative activity of each of the fission products produced in a reactor is the product of the fission yield and that factor. This product is shown in Tables 5 and 6 as the "Relative Activity Yield" which is the relative amount of each fission product isotope produced. The percentage of total fission product activity is also shown in Tables 5 and 6. These figures were decay-corrected for the 29 years that have elapsed since the last reactor was shut down on the SSFL Area IV, resulting in the activity percentage at 29 years shown in the tables. It should be noted that it is assumed that all Kr-85 has dissipated into the atmosphere and is no longer onsite since it is a noble (non-reactive) gas. It can be seen from Tables 5 and 6 that greater than 98 percent of the remaining fission product activity is composed of Sr-90 and Cs-137, with most of the remaining percentage being from Pm-147 for both uranium fuels.

Till and Meyer<sup>18</sup> quantified the potentially significant fission products found in spent reactor fuels. Table 7 contains this list of fission products also decay-corrected for the 29 years that have elapsed since the last reactor was shut down on the SSFL Area IV. It can be observed from the Table 7 that greater than 98-percent of the remaining fission product activity is composed of the same radionuclides as identified above, Sr-90, Cs-137, and Pm-147, with most of the rest being from Eu-155.

<sup>&</sup>lt;sup>18</sup> J. E. Till and H. R. Meyer, *Radiological Assessment, A Textbook on Environmental Dose Analysis*, U.S. Nuclear Regulatory Commission, NUREG/CR-3332, 1983, pp.1-49.

			Activity	<b>Relative Activity</b>	Activity %	
Radionuclide	t <sub>1/2</sub> (years)	Ci/tonne <sup>a</sup>	(%)	at 29 Years	at 29 Years	
H-3	12.3	800	0.0592	1.91E-02	0.2397	
Kr-85	10.73	1.05E+04	0.7776	0.00E+00	0.0000	
Sr-90	29.1	6.00E+04	4.4434	2.75E+00	34.5752	
Zr-93	1.50E+06	2.00E+00	0.0001	1.48E-04	0.0019	
Tc-99	2.13E+05	1.50E+01	0.0011	1.11E-03	0.0139	
Ru-106	1.02E+00	8.20E+05	60.7264	7.12E-05	0.0009	
Sb-125	2.76E+00	1.30E+04	0.9627	6.17E-03	0.0774	
I-129	1.57E+07	4.00E-02	0.0000	2.96E-06	0.0000	
Cs-134	2.07E+00	1.00E+05	7.4057	8.71E-03	0.1094	
Cs-135	2.30E+06	1.20E+00	0.0001	8.89E-05	0.0011	
Cs-137	3.02E+01	1.06E+05	7.8500	4.95E+00	62.1287	
Pm-147	2.62E+00	2.00E+05	14.8113	7.32E-02	0.9193	
Eu-155	4.71E+00	4.00E+04	2.9623	1.54E-01	1.9325	
	Highl	ighted	9 <mark>9</mark>		100	

Table 7. Representative Quantities of Potentially SignificantFission Products in Spent Reactor Fuels

<sup>*a*</sup>Ci/tonne = curies per metric ton;  $t_{1/2}$  = half-life

To identify and quantify the major dose contributors from the identified fission products, the calculated activity percentage at 29 years has been multiplied by the USEPA inhalation,<sup>19</sup> ingestion,<sup>20</sup> and external<sup>21</sup> dose conversion factors. The resulting percentages of the total effective dose equivalents are shown in Table 8. It can be seen from Table 8 that Sr-90 and Cs-137 comprise greater than 99 percent of the dose potential from fission products. Likewise, to identify and quantify the major risk contributors from the identified fission products, the calculated activity percentage at 29 years has been divided by the USEPA Preliminary Remediation Goals (PRGs) for both Residential and Agricultural land use scenarios. The resulting percentages of the relative risk are shown in Table 9. It can be seen from Table 9 that Sr-90 and Cs-137 comprise greater than 99 percent of the risk from fission products.

#### 5.1.3 Activation Product Radionuclides

Some of the neutrons that are released by fuel fissions are captured by the fuel cladding or the other materials in the reactor, and others escape from the reactor and are captured in the shielding around the reactor; far fewer escape the shielding and are captured in the reactor building or the ground. When a neutron is captured by an atom other than the fuel, such as in the fuel cladding or the reactor structure or shield, it creates a new isotope called an "activation product". The term activation product is reserved for products of neutron capture by materials other than the fuel, such as structural components of the nuclear reactor, the reactor coolant, control rods or other neutron poisons, or materials in the environment of the reactor. Most of these activation products are also radioactive, emitting beta and gamma radiation.

<sup>&</sup>lt;sup>19</sup> Federal Guidance Report No.11, *Limiting Values of Radionuclide Intake And Air Concentration and Dose Conversion Factors For Inhalation, Submersion, And Ingestion*; Environmental Protection Agency, EPA-520/1-88-020, September 1988.

<sup>&</sup>lt;sup>20</sup> *Ibid*.

<sup>&</sup>lt;sup>21</sup> Federal Guidance Report No.12, *External Exposure To Radionuclides In Air, Water, And Soil; Environmental* Protection Agency, EPA-520/1-88-020, September 1988.

			Relative			Relative			Relative	External
	Activity		Inhalation	Inhalation		Ingestion	Ingestion		External	Dose %
	% at 29	Inhalation	Dose at 29	Dose % at	Ingestion	Dose at 29	Dose % at	External	Dose at 29	at 29
Radionuclide	Years	DCF	Years	29 Years	DCF	Years	29 Years	$\mathbf{DCF}^{a}$	Years	Years
H-3	0.2397	1.73E-11	4.15E-12	0.0000	1.73E-11	4.15E-12	0.0002	0	0.00E+00	0.0000
Kr-85	0.0000	0	0.00E+00	0.0000	0	0.00E+00	0.0000	4.58E-20	0.00E+00	0.0000
Sr-90	34.5752	3.51E-07	1.21E-05	95.5177	3.85E-08	1.33E-06	61.2531	2.95E-21	1.02E-19	0.0149
Zr-93	0.0019	8.67E-08	1.61E-10	0.0013	4.48E-10	8.33E-13	0.0000	0.00E+00	0.00E+00	0.0000
Tc-99	0.0139	2.25E-09	3.14E-11	0.0002	3.95E-10	5.51E-12	0.0003	5.74E-22	8.01E-24	0.0000
Ru-106	0.0009	1.29E-07	1.15E-10	0.0009	7.40E-09	6.61E-12	0.0003	3.93E-18	3.51E-21	0.0005
Sb-125	0.0774	3.30E-09	2.56E-10	0.0020	7.59E-10	5.88E-11	0.0027	7.66E-18	5.93E-19	0.0869
I-129	0.0000	4.69E-08	1.74E-12	0.0000	7.46E-08	2.78E-12	0.0001	6.92E-20	2.57E-24	0.0000
Cs-134	0.1094	1.25E-08	1.37E-09	0.0108	1.98E-08	2.17E-09	0.0996	2.83E-17	3.09E-18	0.4534
Cs-135	0.0011	1.23E-09	1.37E-12	0.0000	1.91E-09	2.13E-12	0.0001	1.85E-22	2.06E-25	0.0000
Cs-137	62.1287	8.63E-09	5.36E-07	4.2200	1.35E-08	8.39E-07	38.5948	1.09E-17	6.77E-16	99.2194
Pm-147	0.9193	1.06E-08	9.74E-09	0.0767	2.83E-10	2.60E-10	0.0120	2.29E-22	2.11E-22	0.0000
Eu-155	1.9325	1.12E-08	2.16E-08	0.1703	4.13E-10	7.98E-10	0.0367	7.94E-19	1.53E-18	0.2248
Highlighted	100			100			100			100

Table 8. Potential Dose from Representative Quantities of Potentially Significant Fission Products in Spent Reactor Fuels

<sup>*a*</sup>Based on 5 cm contamination depth. DCF = Dose Conversion Factor

			Relative			Relative	
	Activity		Residential	Residential		Agricultural	Agricultural
	% at 29	Residential	Risk at 29	Risk % at	Agricultural	Risk at 29	Risk % at
Nuclide	Years	PRG	Years	29 Years	PRG	Years	29 Years
H-3	0.2397	2.28E+00	1.05E-01	0.0088	1.60E-01	1.50E+00	0.0020
Kr-85	0.0000	2.41E+01	0.00E+00	0.0000	2.23E+01	0.00E+00	0.0000
Sr-90	34.5752	2.31E-01	1.50E+02	12.5578	1.39E-03	2.49E+04	32.4436
Zr-93	0.0019	3.38E+02	5.50E-06	0.0000	2.00E+02	9.30E-06	0.0000
Tc-99	0.0139	2.50E-01	5.58E-02	0.0047	5.57E-03	2.50E+00	0.0033
Ru-106	0.0009	2.25E+00	3.97E-04	0.0000	1.72E-01	5.20E-03	0.0000
Sb-125	0.0774	4.62E-01	1.68E-01	0.0141	4.60E-01	1.68E-01	0.0002
I-129	0.0000	5.96E-01	6.24E-05	0.0000	2.76E-05	1.35E+00	0.0018
Cs-134	0.1094	1.57E-01	6.97E-01	0.0584	7.47E-03	1.46E+01	0.0191
Cs-135	0.0011	1.78E+01	6.27E-05	0.0000	5.09E-03	2.19E-01	0.0003
Cs-137	62.1287	5.97E-02	1.04E+03	87.3134	1.20E-03	5.18E+04	67.5291
Pm-147	0.9193	1.03E+03	8.93E-04	0.0001	6.69E+02	1.37E-03	0.0000
Eu-155	1.9325	3.80E+00	5.09E-01	0.0427	3.74E+00	5.17E-01	0.0007
Highlighted	100			100			100

Table 9. Potential Risk from Representative Quantities of Potentially Significant Fission Products in Spent Reactor Fuels

The probability of an activation product being produced in a reactor is the product of a number of factors, including the concentration of the element being activated in the environment of the reactor, the abundance of the isotope in the element being activated, and the ability of a given material to capture neutrons (i.e., neutron cross section) for the isotope being activated. The International Atomic Energy Agency (IAEA) Handbook on Nuclear Activation Cross Sections<sup>22</sup> provides a list of elements and their isotopes with their abundance and their thermal neutron cross sections. A list of some example long-lived activation product radionuclides and the materials they may be produced from are shown in Table 10.23 Since not only beryllium and cadmium but also europium are common neutron absorbers used in reactor control rods, Eu-152, and Eu-154 were added to the list and will be considered to be potentially present for purposes of developing a process related radionuclide list. In addition, since Th-232 was used as a fertile material in some reactor fuel, U-233 as an activation product will also be considered as potentially present for purposes of developing a process related radionuclide list. However, activation products will not be present unless the parent material was present in the environment of the reactor. Therefore, some of these activation products are not likely present at SSFL Area IV given that the parent material was not likely present in significant concentration in the environment of the reactor. These low probability activation products include Cl-36, Ar-39, Mo-93, Nb-93m, Nb-94, Tc-99 (except as a fission product), Ag-108m, Sn-121m, Pb-205, andPo-210. Table 11 presents a list of activation products that are typically present in nuclear power reactor coolants along with their relative concentrations for both boiling water reactor (BWR) and pressurized water reactor (PWR) coolants.<sup>24</sup> Although many of the reactors at SSFL Area IV were sodium-cooled reactors, this list provides an indication of additional activation products that may have been produced onsite. Many of the activation products listed in both Tables 9 and 10 have short half-lives relative to the 29 years time elapsed since any reactor was operational at SSFL Area IV and could have possibly produced them. Table 12 provides a summary list of long lived ( $t_{1/2} > 1$  year) activation products potentially present at SSFL Area IV.

Till and Meyer<sup>25</sup> quantified the corrosion products (a major subset of activation products) present in spent reactor fuels. Table 13 contains this list of corrosion products also decay corrected for the 29 years that have elapsed since the last reactor was shut down on the SSFL Area IV. It can be seen from the Table 13 that greater than 99 percent of the remaining corrosion product activity remaining is composed of Fe-55 and Co-60.

## 5.1.4 Transuranic Radionuclides

Some of the neutrons that are released by fuel fissions are captured by fuel element atoms that do not split, but instead form new isotopes, called "transuranics." Although these radionuclides are created by "activation" in the reactor, they are generally considered to be a separate category than activation products. Transuranics are elements heavier than uranium, all of which are essentially man-made and do not naturally occur in significant quantities in nature. All of the transuranic radionuclides are radioactive, emitting alpha, beta, and/or gamma radiation.

<sup>&</sup>lt;sup>22</sup> Handbook on Nuclear Activation Cross Sections, Technical Reports Series No.156, International Atomic Energy Agency, Vienna, 1974.

<sup>&</sup>lt;sup>23</sup> This table is licensed under the GNU Free Documentation License. It uses material from the Wikipedia article "Activation Products".

<sup>&</sup>lt;sup>24</sup> Chien C. Lin, *Radiochemistry in Nuclear Power Reactors*, Nuclear Science Series, NAS-NS-3119, National Research Council, National Academy Press, Washington, D.C. 1996.

<sup>&</sup>lt;sup>25</sup> J. E. Till and H. R. Meyer, pp.1-50.

Radionuclide	Half-life	Activation Parent
Tritium	12.3 y	Lithium-6,7
		Boron-10
Beryllium-10	1.53 my	Boron-10
Carbon-14	5,730 y	Nitrogen-14
		Carbon-13
Sodium-24	15.0 h	Sodium-23
Chlorine-36	301 ky	Chlorine-35
Argon-39	269 y	Argon-38
Iron-55	2.73 y	Iron-54
Nickel-59	76 ky	Nickel-58
Cobalt-60	5.27 y	Cobalt-59
Nickel-63	100 y	Nickel-62
Molybdenum-93	3.5 ky	Molybdenum-92
Niobium-93m	16.1 y	Niobium-93
Niobium-94	20.3 ky	Niobium-93
Technetium-99	211 ky	Molybdenum-98
Silver-108m	108 y	Silver-107
Cadmium-113m	12.2 y	Cadmium-112
Tin-121m	56 y	Tin-120
Ba-133	10.5 y	Ba-132
Europium-152	13.5 y	Europium-151
Europium-154	8.59 y	Europium-153
Lead-205	15.3 my	Lead-204
Polonium-210	138 d	Bismuth-209
Uranium-233	159 ky	Thorium-232

## Table 10. Example Activation Products

h= hours; d = days; y = years; k = thousand; m = million; b = billion

Radionuclide	Half-Life	<b>BWR</b> <sup><i>a</i></sup>	PWR <sup>b</sup>
H-3	12.3 y	10	1,000
N-16	7.1s	60,000	60,000
F-18	1.8h	1.0	
Na-24	15h	10	47
P-32	14d	0.2	
Cr-51	27d	6.0	3.1
Mn-54	312d	0.07	1.6
Mn-56	2.6h	50	_
Fe-55	2.7y	1.0	1.2
Fe-59	45d	0.03	0.3
Co-58	71d	0.2	4.6
Co-60	5.3y	0.4	0.53
Ni-63	100y	0.001	_
Cu-64	12.7h	30	_
Zn-65	244d	0.2	0.51
Ag-110m	250d	0.001	1.3
W-187	24h	0.3	2.5

Table 11. Selected ANS Standard Radionuclide Concentrations in Reactor **Coolants Activation Products (mCi/kg)** 

<sup>*a*</sup> A reference BWR is a 3,400 MW BWR/5. <sup>*b*</sup> A reference PWR is a 3,400 MW PWR with U-tube steam generators. mCi/kg = millicuries per kilogram s = seconds; h = hours; d = days; y = years

k = thousand; m = million; b = billion

Radionuclide	Half-life
Tritium	12.3 y
Beryllium-10	1.53 my
Carbon-14	5,730 y
Iron-55	2.73 у
Nickel-59	76 ky
Cobalt-60	5.27 y
Nickel-63	100 y
Cadmium-113m	13 y
Ba-133	10.5 y
Europium-152	13.5 y
Europium-154	8.59 y
Uranium-233	159 kv

#### Table 12. Summary of Potential Long-Lived Activation Products

y = years; k = thousand; m = million; b = billion

Radionuclide	t <sub>1/2</sub> (years)	Activity Ci/tonne <sup>a</sup>	Activity (%)	Relative Activity at 29 Years	Activity % at 29 Years
Mn-54	0.86	30,000	36.3636	3.36E-06	0.0011
Fe-55	2.7	20,000	24.2424	1.39E-01	44.8079
Fe-59	0.12	500	0.6061	2.33E-51	0.0000
Co-58	0.2	30,000	36.3636	2.05E-29	0.0000
Co-60	5.26	2000	2.4242	1.72E-01	55.1911
	Highl	ighted	99		100

 Table 13. Representative Quantities of Corrosion Products in Spent Reactor Fuels

<sup>*a*</sup>Ci/tonne = curies per metric ton;  $t_{1/2}$  = half-life

To identify and quantify the major dose and related potential risk contributors from the identified corrosion products, the calculated activity percentage at 29 years has been multiplied by the USEPA inhalation,<sup>26</sup> ingestion,<sup>27</sup> and external<sup>28</sup> dose conversion factors. The resulting dose percentages are shown in Table 14.

It can be seen from Table 14 that Fe-55 and Co-60 comprise greater than 99 percent of the dose potential from corrosion products. Likewise, to identify and quantify the major risk contributors from the identified corrosion products, the calculated activity percentage at 29 years has been divided by the USEPA PRGs for both Residential and Agricultural land use scenarios. The resulting percentages of the relative risk are shown in Table 15. It can be seen from Table 15 that Co-60 contributes greater than 99 percent of the risk from corrosion products. The other potentially significant activation products not reflected in Tables 14 and 15 would include H-3, Be-10, C-14, Cd-113m, Ba-133, Eu-152, Eu-154, and U-233.

Till and Meyer<sup>29</sup> quantified the transuranics present in spent reactor fuels. Table 16 contains this list of transuranics also decay corrected for the 29 years that have elapsed since the last reactor was shut down on the SSFL Area IV. It can be seen from the Table 16 that greater than 99 percent of the remaining transuranics activity remaining is composed of Pu-238, Pu-239, Pu-240, Pu-241, Am-241, and Cm-244.

To identify and quantify the major dose and related potential risk contributors from the identified transuranics, the calculated activity percentage at 29 years has been multiplied by the USEPA inhalation,<sup>30</sup> ingestion,<sup>31</sup> and external<sup>32</sup> dose conversion factors. The resulting dose percentages are shown in Table 17. It can be seen from Table 17 that all of the listed transuranics except Pu-242 and Cm-242 combine to contribute greater than 99 percent of the dose potential from transuranics for at least one of the exposure pathways.

<sup>&</sup>lt;sup>26</sup> Federal Guidance Report No.11.

<sup>&</sup>lt;sup>27</sup> *Ibid*.

<sup>&</sup>lt;sup>28</sup> Federal Guidance Report No.12.

<sup>&</sup>lt;sup>29</sup> J. E. Till and H. R. Meyer, pp.1-50.

<sup>&</sup>lt;sup>30</sup> Federal Guidance Report No.11.

<sup>&</sup>lt;sup>31</sup> *Ibid*.

<sup>&</sup>lt;sup>32</sup> Federal Guidance Report No.12.

Radionuclide	Activity % at 29 Years	Inhalation DCF	Relative Inhalation Dose at 29 Years	Inhalation Dose % at 29 Years	Ingestion DCF	Relative Ingestion Dose at 29 Years	Ingestion Dose % at 29 Years	External DCF <sup>a</sup>	Relative External Dose at 29 Years	External Dose % at 29 Years
Mn-54	0.0011	1.81E-09	1.95E-12	0.0001	7.48E-10	8.08E-13	0.0002	1.51E-17	1.63E-20	0.0007
Fe-55	44.8079	7.26E-10	3.25E-08	0.9875	1.64E-10	7.35E-09	1.7961	0	0.00E+00	0.0000
Fe-59	0.0000	4.00E-09	3.00E-57	0.0000	1.81E-09	1.36E-57	0.0000	2.12E-17	1.59E-65	0.0000
Co-58	0.0000	2.94E-09	1.94E-35	0.0000	9.68E-10	6.38E-36	0.0000	1.77E-17	1.17E-43	0.0000
Co-60	55.1911	5.91E-08	3.26E-06	99.0125	7.28E-09	4.02E-07	98.2037	4.45E-17	2.46E-15	99.9993
Highlighted	100			100			100			100

Table 14. Potential Dose from Representative Quantities of Corrosion Products in Spent Reactor Fuels

<sup>*a*</sup>Based on 5 cm contamination depth. DCF = Dose Conversion Factor

Cable 15. Potential Risk from Representative	e Quantities of Corrosion	<b>Products in Spent Reactor Fuels</b>
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Radionuclide	Activity % at 29 Years	Residential PRG	Relative Residential Risk at 29 Years	Residential Risk % at 29 Years	Agricultural PRG	Relative Agricultural Risk at 29 Years	Agricultural Risk % at 29 Years
Mn-54	0.0011	6.92E-01	1.56E-03	0.0001	3.69E-01	2.93E-03	0.0000
Fe-55	44.8079	2.69E+03	1.67E-02	0.0011	8.21E-01	5.46E+01	0.0890
Fe-59	0.0000	3.26E+00	2.30E-49	0.0000	1.20E+00	6.25E-49	0.0000
Co-58	0.0000	2.66E+00	2.48E-27	0.0000	1.27E-01	5.19E-26	0.0000
Co-60	55.1911	3.61E-02	1.53E+03	99.9988	9.01E-04	6.13E+04	99.9110
Highlighted	100			100			100

Radionuclide	t <sub>1/2</sub> (vears)	Activity Ci/tonne <sup>a</sup>	Activity (%)	Relative Activity at 29 Years	Activity % at 29 Years
Np-237	2.14E+06	1	0.0005	5.18E-04	0.0016
Pu-238	87.7	4,000	2.0734	1.77E+00	5.3954
Pu-239	2.41E+04	500	0.2592	2.59E-01	0.7901
Pu-240	6.56E+03	650	0.3369	3.36E-01	1.0255
Pu-241	1.44E+01	150 <b>,</b> 000	77.7512	2.96E+01	90.1398
Pu-242	3.75E+05	2	0.0010	1.04E-03	0.0032
Am-241	4.33E+02	750	0.3888	3.76E-01	1.1482
Am-243	7.37E+03	20	0.0104	1.03E-02	0.0316
Cm-242	4.50E-01	35,000	18.1420	6.53E-13	0.0000
Cm-244	1.81E+01	2,000	1.0367	4.80E-01	1.4647
	Highli	ighted	99		100

Table 16. Representative Quantities of Transuranics in Spent Reactor Fuels

<sup>*a*</sup>Ci/tonne = curies per metric ton;  $t_{1/2}$  = half-life

Likewise, to identify and quantify the major risk contributors from the identified transuranics, the calculated activity percentage at 29 years has been divided by the USEPA PRGs for both Residential and Agricultural land use scenarios. The resulting percentages of the relative risk are shown in Table 18. It can be seen from Table 18 that Pu-238, Pu-239, Pu-240, Pu-241, Am-241, and Cm-244 comprise greater than 99 percent of the risk from transuranics.

## 5.1.5 Combined Activity Dose and Risk

To identify and quantify the major dose and related potential risk contributors from all radionuclides produced by a reactor (excluding those associated with fuel) based on the quantities present in spent reactor fuels provided by Till and Meyer, the calculated activity percentage at 29 years has been multiplied by the USEPA inhalation,<sup>33</sup> ingestion,<sup>34</sup> and external<sup>35</sup> dose conversion factors for the combined sum of the fission products, activation products and transuranics. Both the radionuclides with the potential to contribute more than one percent of the total activity and the radionuclides with the potential to contribute more than one percent of the total dose and associated risk are reflected in Table 19. It can be seen from Table 19 that Sr-90, Cs-137, Co-60, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, and Cm-244 comprise greater than 99 percent of the pathway dose potential and that Sr-90 and Cs-137 comprise greater than 99 percent of the risk potential from reactor produced contaminants excluding H-3, C-14. The other potentially significant radionuclides associated with reactors would include the fuel element radionuclides Th-228, Th-232, U-234, U-235, and U-238. Another radionuclide comprising at least 0.5 % of the radioactivity potentially present onsite but that contributes less than 1 percent of the dose and risk potential is Eu-155.

<sup>&</sup>lt;sup>33</sup> Federal Guidance Report No.11.

<sup>&</sup>lt;sup>34</sup> Ibid.

<sup>&</sup>lt;sup>35</sup> Federal Guidance Report No.12.

	Activity		Relative Inhalation	Inhalation		Relative Ingestion	Ingestion		Relative External	External Dose %
	% at 29	Inhalation	Dose at 29	Dose % at	Ingestion	Dose at 29	Dose % at	External	Dose at	at 29
Radionuclide	Years	DCF	Years	29 Years	DCF	Years	29 Years	DCF <sup>a</sup>	29 Years	Years
Np-237	0.0016	1.46E-04	2.31E-07	0.0189	1.20E-06	1.90E-09	0.0189	3.51E-18	5.55E-21	1.9429
Pu-238	5.3954	1.06E-04	5.72E-04	46.7331	8.65E-07	4.67E-06	46.5119	7.60E-22	4.10E-21	1.4356
Pu-239	0.7901	1.16E-04	9.16E-05	7.4889	9.56E-07	7.55E-07	7.5274	1.15E-21	9.09E-22	0.3181
Pu-240	1.0255	1.16E-04	1.19E-04	9.7205	9.56E-07	9.80E-07	9.7706	7.44E-22	7.63E-22	0.2671
Pu-241	90.1398	2.23E-06	2.01E-04	16.4253	1.85E-08	1.67E-06	16.6192	2.44E-23	2.20E-21	0.7700
Pu-242	0.0032	1.11E-04	3.51E-07	0.0287	9.08E-07	2.87E-09	0.0286	6.43E-22	2.03E-24	0.0007
Am-241	1.1482	1.20E-04	1.38E-04	11.2591	9.84E-07	1.13E-06	11.2602	2.18E-19	2.50E-19	87.6378
Am-243	0.0316	1.19E-04	3.76E-06	0.3069	9.79E-07	3.09E-08	0.3079	6.59E-19	2.08E-20	7.2820
Cm-242	0.0000	4.67E-06	9.30E-18	0.0000	3.10E-08	6.17E-20	0.0000	8.60E-22	1.71E-33	0.0000
Cm-244	1.4647	6.70E-05	9.81E-05	8.0187	5.45E-07	7.98E-07	7.9553	6.74E-22	9.87E-22	0.3456
Highlighted	100			100			100			<u>9</u> 9

Table 17. Potential Dose from Representative Quantities of Transuranics in Spent Reactor Fuels

<sup>*a*</sup>Based on 5 cm contamination depth.

DCF = Dose Conversion Factor

## Table 18. Potential Risk from Representative Quantities of Transuranics in Spent Reactor Fuels

	Activity % at 29	Residential	Relative Residential Risk at 29	Residential Risk % at	Agricultural	Relative Agricultural Risk at 29	Agricultural Risk % at
Nuclide	Years	PRG	Years	29 Years	PRG	Years	29 Years
Np-237	0.0016	1.30E-01	1.22E-02	0.3222	4.48E-04	3.53E+00	0.2892
Pu-238	5.3954	2.97E+00	1.82E+00	48.1288	7.31E-03	7.38E+02	60.4765
Pu-239	0.7901	2.59E+00	3.05E-01	8.0817	6.09E-03	1.30E+02	10.6299
Pu-240	1.0255	2.60E+00	3.94E-01	10.4497	6.10E-03	1.68E+02	13.7749
Pu-241	90.1398	4.06E+02	2.22E-01	5.8820	1.05E+00	8.58E+01	7.0340
Pu-242	0.0032	2.73E+00	1.16E-03	0.0307	6.42E-03	4.93E-01	0.0404
Am-241	1.1482	1.87E+00	6.14E-01	16.2676	1.32E-02	8.70E+01	7.1274
Am-243	0.0316	1.66E-01	1.90E-01	5.0372	1.11E-02	2.84E+00	0.2330
Cm-242	0.0000	3.22E+02	6.18E-15	0.0000	1.89E+01	1.05E-13	0.0000
Cm-244	1.4647	6.69E+00	2.19E-01	5.8002	3.04E-01	4.82E+00	0.3948
Highlighted	100			100			99

	Activity % at	Inhalation Dose	Ingestion Dose	External Dose	Residential Risk	Agricultural Risk
Radionuclide	29 Years	% at 29 Years	% at 29 Years	% at 29 Years"	% at 29 Years	% at 29 Years
H-3	0.1507	0.0000	0.0001	0.0000	0.0088	0.0019
Kr-85	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Sr-90	21.7367	1.6565	1.6286	0.0148	12.4963	32.0820
Zr-93	0.0012	0.0000	0.0000	0.0000	0.0000	0.0000
Tc-99	0.0088	0.0000	0.0001	0.0000	0.0047	0.0032
Ru-106	0.0006	0.0000	0.0001	0.0005	0.0000	0.0000
Sb-125	0.0487	0.0000	0.0009	0.0861	0.0140	0.0002
I-129	0.0000	0.0000	0.0000	0.0000	0.0000	0.0017
Cs-134	0.0687	0.0002	0.0316	0.4494	0.0582	0.0189
Cs-135	0.0007	0.0000	0.0000	0.0000	0.0000	0.0003
Cs-137	39.0591	0.0732	12.2314	98.3511	86.8857	66.7764
Pm-147	0.5779	0.0013	0.0038	0.0000	0.0001	0.0000
Eu-155	1.2149	0.0030	0.0116	0.2228	0.0425	0.0007
Mn-54	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Fe-55	0.0672	0.0000	0.0003	0.0000	0.0000	0.0002
Fe-59	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Co-58	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Co-60	0.0828	0.0011	0.0140	0.8508	0.3044	0.1884
Np-237	0.0006	0.0185	0.0163	0.0005	0.0006	0.0027
Pu-238	1.9953	45.9221	40.0363	0.0004	0.0892	0.5600
Pu-239	0.2922	7.3589	6.4794	0.0001	0.0150	0.0984
Pu-240	0.3793	9.5519	8.4103	0.0001	0.0194	0.1276
Pu-241	33.3355	16.1402	14.3054	0.0002	0.0109	0.0651
Pu-242	0.0012	0.0282	0.0246	0.0000	0.0001	0.0004
Am-241	0.4246	11.0637	9.6925	0.0214	0.0302	0.0660
Am-243	0.0117	0.3016	0.2651	0.0018	0.0093	0.0022
Cm-242	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Cm-244	0.5417	7.8795	6.8477	0.0001	0.0108	0.0037
Highlighted	99	100	100	100	99	99

Table 19. Potential Dose and Risk from Representative Quantities of Produced Radionuclides in Spent Reactor Fuels

<sup>*a*</sup>Based on 5 cm contamination depth.

## 5.2 RADIONUCLIDES ASSOCIATED WITH ACCELERATOR OPERATIONS

The radionuclides produced by accelerator operations depend on the composition of the targets that were irradiated and the types of particles that were accelerated. It appears that the Van de Graaff generators were primarily used to bombard tritium targets with deuterons to produce neutrons. The activation products produced are likely to have been similar to the activation products produced in reactors. This similarity would be due to the construction materials for both reactors and accelerators (e.g., hydrogenous concrete and steal) and the long-lived activation products would be comprised of some of the radionuclides listed in Table 10. Estimation of quantities is not possible at this time but would likely be small compared to activation products produced by the reactors.

# 5.3 RADIONUCLIDES ASSOCIATED WITH RESEARCH AND FABRICATION, USE, AND STORAGE OF RADIOACTIVE SOURCES

The primary radionuclides produced as a result of research include isotopes of uranium, plutonium, and neptunium (the same isotopes as those listed under the reactor operations discussed above), stored onsite in conjunction with the TRUMP-S program. Radionuclides Mn-54 and Co-60 were produced as a result of operations associated with the Corrosion Testing Laboratory. It is likely that the contamination onsite from these operations was relatively small compared to that from reactor operations given that the sources were either sealed or in storage rather than in active operations. The primary radionuclide associated with fabrication and use of radioactive sources was Pm-147, but may have also included Co-60 and Cs-137.

#### 5.4 SUMMARY OF OPERATIONAL RELATED RADIONUCLIDES

Table 20 provides a summary of the SSFL Area IV historical operations-related radionuclides with halflives greater than one year. The table also shows which radionuclides are expected to contribute more than 1 percent of the current activity, potential dose, and potential risk. The actual calculation of relative activity, dose, and risk could only be performed for radionuclides in spent reactor fuel. Radionuclides associated with fuel elements are assumed to have the potential to contribute more than 1 percent of the current activity, dose, and risk. For some radionuclides from reactors and other processes, no relative activity information is available and therefore the potential for contamination is unknown. In the absence of specific concentration or process knowledge information, they were not eliminated from the list of radionuclides that could have the potential to contribute greater than 1 percent of the total activity, dose, and risk.

## 6.0 CONCLUSION

From this evaluation, those radionuclides that have the potential to contribute significantly to human or environmental dose and risk after 29 years since operations were suspended in Area IV of SSFL have been identified. The list of major contributors may be used to prioritize analytical requirements for new samples to be collected for site characterization and risk assessment. The actual concentrations present in Area IV of SSFL and resulting risk to the public depend on the quantities of radionuclides that were released to the environment and the residual persistence in the environs after 29 years of decay and prior remediation efforts. That will be the focus of the radiological survey and sampling efforts currently in planning by USEPA and the dose/risk assessment aspect of the Environmental Impact Statement currently being developed.

			Potential to	Potential to	Potential to
			Contribute > 1%	Contribute > 1%	Contribute > 1%
Radionuclide	t <sub>1/2</sub> (years)	Process Relationship	of Activity	of Pathway Dose	of Risk
Th-228	1.90E+00	Reactor Fuel Element	Th-228	Th-228	Th-228
Th-232	1.40E+10	Reactor Fuel Element	Th-232	Th-232	Th-232
U-234	2.46E+05	Reactor Fuel Element	U-234	U-234	U-234
U-235	7.04E+08	Reactor Fuel Element	U-235	U-235	U-235
U-238	4.50E+09	Reactor Fuel Element	U-238	U-238	U-238
Pu-238	8.77E+01	Reactor Fuel Element	Pu-238	Pu-238	Pu-238
Pu-239	2.40E+04	Reactor Fuel Element	Pu-239	Pu-239	Pu-239
Pu-240	6.60E+03	Reactor Fuel Element	Pu-240	Pu-240	Pu-240
Pu-241	1.44E+01	Reactor Fuel Element	Pu-241	Pu-241	Pu-241
H-3	1.23E+01	Reactor Fission Product	$H-3^a$	$H-3^a$	H-3 <sup><i>a</i></sup>
Se-79	6.50E+04	Reactor Fission Product			
Kr-85	10.73	Reactor Fission Product			
Sr-90	29.1	Reactor Fission Product	Sr-90	Sr-90	Sr-90
Zr-93	1.50E+06	Reactor Fission Product			
Tc-99	2.13E+05	Reactor Fission Product			
Ru-106	1.02E+00	Reactor Fission Product			
Pd-107	6.50E+06	Reactor Fission Product			
Cd-113	1.41E+01	Reactor Fission Product			
Sb-125	2.76E+00	Reactor Fission Product			
Sn-126	1.00E+05	Reactor Fission Product			
I-129	1.57E+07	Reactor Fission Product			
Cs-134	2.07E+00	Reactor Fission Product			
Cs-135	2.30E+06	Reactor Fission Product			
Cs-137	3.02E+01	Reactor Fission Product	Cs-137	Cs-137	Cs-137
Sm-146	1.03E+08	Reactor Fission Product			
Pm-147	2.62E+00	Reactor Fission Product	Pm-147		
Eu-152	1.35E+01	Reactor Fission Product			
Eu-154	8.59E+00	Reactor Fission Product			
Eu-155	4.71E+00	Reactor Fission Product	Eu-155		
H-3	1.23E+01	Reactor Activation Product			
Be-10	1.53E+06	Reactor Activation Product	Be-10 <sup><i>a</i></sup>	Be-10 <sup><i>a</i></sup>	Be-10 <sup><i>a</i></sup>
C-14	5.73E+03	Reactor Activation Product			
Fe-55	2.73E+00	Reactor Activation Product			
Ni-59	7.60E+03	Reactor Activation Product			
Co-60	5.27E+00	Reactor Activation Product		Co-60	
Ni-63	1.00E+02	Reactor Activation Product			
Cd-113m	1.41E+01	Reactor Activation Product	Cd-113m <sup>a</sup>	Cd-113m <sup><i>a</i></sup>	Cd-113m <sup><i>a</i></sup>
Ba-133	1.05 E+01	Reactor Activation Product	Ba-133	Ba-133	Ba-133
Eu-152	1.35E+01	Reactor Activation Product	Eu-152 <sup><i>a</i></sup>	Eu-152 <sup><i>a</i></sup>	Eu-152 <sup><i>a</i></sup>
Eu-154	8.59E+00	Reactor Activation Product	Eu-154 <sup>a</sup>	Eu-154 <sup><i>a</i></sup>	Eu-154 <sup><i>a</i></sup>
U-233	1.59E+05	Reactor Activation Product	U-233 <sup><i>a</i></sup>	U-233 <sup><i>a</i></sup>	U-233 <sup><i>a</i></sup>
Np-237	2.14E+06	Reactor Transuranic			
Pu-238	87.7	Reactor Transuranic	Pu-238	Pu-238	
Pu-239	2.41E+04	Reactor Transuranic	Pu-239	Pu-239	
Pu-240	6.56E+03	Reactor Transuranic	Pu-240	Pu-240	
Pu-241	1.44E+01	Reactor Transuranic	Pu-241	Pu-241	
Pu-242	3.75E+05	Reactor Transuranic			
Am-241	4.33E+02	Reactor Transuranic	Am-241	Am-241	

## Table 20. Summary of the SSFL Area IV Historical Operations Related Radionuclides with Half-Lives Greater Than One Year

			Potential to	Potential to	Potential to
			Contribute > 1%	Contribute > 1%	Contribute > 1%
Radionuclide	t <sub>1/2</sub> (years)	Process Relationship	of Activity	of Dose	of Risk
Am-243	7.37E+03	Reactor Transuranic			
Cm-242	4.50E-01	Reactor Transuranic			
Cm-244	1.81E+01	Reactor Transuranic	Cm-244	Cm-244	
H-3	1.23E+01	Accelerator Activation Product	H-3 <sup><i>a</i></sup>	H-3 <sup><i>a</i></sup>	H-3 <sup><i>a</i></sup>
Be-10	1.53E+06	Accelerator Activation Product			
C-14	5.60E+03	Accelerator Activation Product			
Cl-36	3.01E+05	Accelerator Activation Product			
Ar-39	2.69E+02	Accelerator Activation Product			
Fe-55	2.73E+00	Accelerator Activation Product			
Ni-59	7.60E+04	Accelerator Activation Product			
Co-60	5.27E+00	Accelerator Activation Product			
Ni-63	1.00E+02	Accelerator Activation Product			
Mo-93	4.00E+03	Accelerator Activation Product			
Nb-93m	1.60E+01	Accelerator Activation Product			
Nb-94	2.03E+04	Accelerator Activation Product			
Tc-99	2.13E+05	Accelerator Activation Product			
Ag-108m	1.08E+02	Accelerator Activation Product			
Cd-113m	1.30E+01	Accelerator Activation Product			
Sn-121m	5.60E+01	Accelerator Activation Product			
Pb-205	1.53E+07	Accelerator Activation Product			
U-234	2.46E+05	Research Radionuclide	U-234 <sup><i>a</i></sup>	U-234 <sup><i>a</i></sup>	U-234 <sup><i>a</i></sup>
U-235	7.04E+08	Research Radionuclide	U-235 <sup><i>a</i></sup>	U-235 <sup><i>a</i></sup>	U-235 <sup><i>a</i></sup>
U-238	4.50E+09	Research Radionuclide	U-238 <sup><i>a</i></sup>	U-238 <sup><i>a</i></sup>	U-238 <sup><i>a</i></sup>
Np-237	2.14E+06	Research Radionuclide	Np-237 <sup><i>a</i></sup>	Np-237 <sup><i>a</i></sup>	Np-237 <sup><i>a</i></sup>
Pu-238	87.7	Research Radionuclide	Pu-238 <sup><i>a</i></sup>	Pu-238 <sup><i>a</i></sup>	Pu-238 a
Pu-239	2.41E+04	Research Radionuclide	Pu-239 <sup><i>a</i></sup>	Pu-239 <sup><i>a</i></sup>	Pu-239 <sup><i>a</i></sup>
Pu-240	6.56E+03	Research Radionuclide	Pu-240 <sup><i>a</i></sup>	Pu-240 <sup><i>a</i></sup>	Pu-240 <sup><i>a</i></sup>
Pu-241	1.44E+01	Research Radionuclide	Pu-241 a	Pu-241 a	Pu-241 a
Co-60	5.27E+00	Research Radionuclide	Co-60 <sup><i>a</i></sup>	Co-60 <sup><i>a</i></sup>	Co-60 <sup><i>a</i></sup>
Cs-137	3.02E+01	Research Radionuclide	Cs-137 <sup><i>a</i></sup>	Cs-137 <sup><i>a</i></sup>	Cs-137 <sup><i>a</i></sup>
Pm-147	2.62E+00	Research Radionuclide	Pm-147 <sup><i>a</i></sup>	Pm-147 <sup><i>a</i></sup>	Pm-147 <sup><i>a</i></sup>

### Table 20. Summary of the SSFL Area IV Historical Operations Related Radionuclides with Half-Lives Greater Than One Year (continued)

<sup>*a*</sup>No relative activity information is available for these radionuclides from these processes and therefore the potential for contamination is unknown. In the absence of specific concentration information, they were not eliminated from the list to have the potential to contribute greater than 1 percent of the total activity, dose, and risk.  $t_{1/2}$  = half life

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# APPENDIX A ABOUT THE AUTHOR

# **ABOUT THE AUTHOR**

Dr. Thomas L. Rucker received his Ph.D. from The University of Tennessee in Analytical Chemistry with an emphasis in Radiochemistry and a minor in Health Physics. He is currently Manager of Radiological Assessment and Protection for Science Applications International Corporation (SAIC), where he leads a team of health physicists and radiochemists in providing radiological characterization, assessment, and protection services. Dr. Rucker specializes in radionuclide measurement and dose assessment for health and environment protection and also provides expertise in other areas of analytical chemistry, environmental chemistry, and health physics, including environmental monitoring, waste management, radiological site characterization, and risk assessment. He has extensive experience in analytical chemistry, radiochemistry, radiological monitoring, dose and risk assessment, radiological site characterization, and environmental and waste management. His experience in analytical and radiochemistry includes laboratory management; laboratory automation; research and development; quality assurance/quality control (QA/QC) program management; document/procedure preparation and review; personnel training; method development; data interpretation; information management, and statistical analysis. He has expertise in the sampling, separation, and measurement of both radiological and nonradiological analytes. He has extensive experience in alpha and gamma spectroscopy, liquid scintillation counting, gas chromatography/mass spectrometry, and nuclear nondestructive and field analysis techniques. He also has extensive experience in analytical data evaluation, validation, and management.

His radiological monitoring, dose and risk assessment, and environmental and waste management experience includes evaluation and preparation of recommendations for radiological effluent and environmental monitoring programs; the interpretation and implementation of radiation protection regulations in technical basis documents for internal dosimetry, air monitoring, radiological survey, and contamination control; establishment of internal dosimetry bioassay programs; evaluation of nuclear accident dosimetry programs; development of site and facility characterization plans; development of analytical data summary reports; development and verification of characterization databases and dose calculation software; development of waste characterization plans; evaluation of waste characterization data; and preparation of waste minimization reports. He has participated in the development or review of environmental assessments (EAs), environmental impact statements (EISs), decontamination and decommissioning (D&D) plans and reports and D&D Funding Plans for NRC license termination, ALARA assessments, air and water quality studies, data management and validation studies, waste characterization plans; waste certification assessments, sampling and analysis plans (SAPs), data quality objectives, field sampling plans, quality assurance project plans (QAPPs), health and safety plans, remedial investigation reports, pathway analyses, dose assessments, and human health risk assessments.

Dr. Rucker's project experience in site characterization and risk assessment includes support to the Department of Energy (DOE) under subcontract to CDM Federal for the development of an Environmental Impact Statement (EIS) for the decommissioning of the Energy Technology Engineering Center at the Santa Susanna Field Laboratory. The project has involved the evaluation of historical processes and data and the development of a Data Gap Analysis Report identifying additional survey and sampling required to perform risk assessments required to evaluate the alternatives evaluated in the Environmental Impact Statement. Dr. Rucker serves as the lead radiological risk assessor and a radionuclide site characterization expert for the project.

He also provides support to the Department of Energy for radiological data collection, evaluation, dose assessment, and human health risk assessment under contract to Bechtel Jacobs Company, LLC, for the East Tennessee Technology Park Reindustrialization Program in Oak Ridge, Tennessee, and under contract to Restoration Services Incorporated for the Portsmouth Gaseous Diffusion Plant in Ohio. This

support has included determination of extent and isotopic distribution of residual contamination, development of technical basis for radiological survey and sampling plans based on Multi-Agency Radiation Survey and Site Investigation guidance, development of survey and sampling plans and summary reports, and dose and risk assessment using RESRAD and RESRAD BUILD computer modeling codes as well as the use of EPA-approved modeling and slope factors.

As project manager he led a task to document the existing knowledge base for radiological characterization of the areas and facilities at DOE's Y-12 Weapons Complex and the East Tennessee Technology Park in Tennessee that are managed by Bechtel Jacobs LLC (BJC). He also supported for a Supplemental Site-Wide Environmental Impact Statement for the Los Alamos National Laboratory Site. He helped evaluate historical characterization data and exposure scenarios for a number of Material Disposal Areas (MDAs) containing legacy radiological waste from the site. This information was used to develop the chemical and radiological source term for air release to the public, transportation accidents, and worker accident exposures resulting from possible excavation of the waste from the MDAs for disposal at permanent repositories.

Dr. Rucker provided support for radiological site characterization studies for several Formerly Utilized Sites Remedial Action Program (FUSRAP) sites and DOE's Hanford site in Washington, and he served as project manager to review the existing characterization data found in UCN-2109 files associated with containers of Oak Ridge Reservation Legacy Low-Level Waste (LLLW) for WESKEM. The review provided quantitative evaluation of the uncertainties' impact on the the ability of each waste population to meet the waste acceptance criteria for the available disposal facility options.

Dr. Rucker has specialized training in Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) implementation, and the use of RESRAD, RESRAD-BUILD, and RESRAD-RECYCLE dose and risk modeling. Dr. Rucker's experience includes work with national laboratories, DOE defense facilities, DOE fuel cycle facilities, DOE waste storage facilities, EPA, NASA, Army Core of Engineers, commercial laboratories, and commercial instrumentation manufacturers.

Dr. Rucker is a member of the Health Physics Society and the American Chemical Society and is the author of numerous technical papers and publications, including several published in the Journal of Radioanalytical and Nuclear Chemistry. He has presented papers at national and international conferences, including the Annual Conference on Bioassay, Analytical and Environmental Radiochemistry, Third International Conference on Methods and Applications of Radioanalytical Chemistry, and the Southeastern Regional Meeting of the American Chemical Society.