Report to NEAC Fuel Cycle Subcommittee Meeting of October 22, 2015

Washington, DC December 7, 2015

Al Sattelberger (Chair), Carol Burns, Margaret Chu, Raymond Juzaitis, Chris Kouts, Sekazi Mtingwa, Ronald Omberg, Joy Rempe, Dominique Warin

I. Introduction

The agenda for the October 22, 2015 Fuel Cycle Subcommittee meeting is given below. The meeting provided members an overview of several research efforts funded by the DOE Office of Nuclear Energy's Fuel Cycle Technologies (FCT) program and related research that is coordinated with the FCT program. As usual, the meeting started with a budget overview. All members of the Subcommittee, with the exception of Ray Juzaitis, were present.

Agenda

Chair: Dr. Alfred P. Sattelberger Location: Argonne National Lab Offices, L'Enfant Plaza	
9:00 am	Executive Session
9:15 am	Fuel Cycle Research and Development FY 2016 Budget Update
9:30 am	Aqueous Separation Research within the Material Recovery and Waste Form Development Campaign
10:30 am	Material Recovery Q&A
10:45 am	Break
11:00 am	MELCOR Overview and Applicability to ATF Response
11:45 am	The Fuels Product Line: Update on BISON & MARMOT Development
12:30 am	Working Lunch
1:15 pm	Advanced Reactor Program
2:15 pm	Nuclear Fuel Storage & Transportation Program Update and FY16 Planning
3:15 pm	Joint EM-NE-SC-International Study of Glass Behavior over Geologic Time Scales
4:15 pm	Closed Session
5:30 pm	Adjourn

Our report is organized more or less along the lines of the agenda.

II. Material Recovery – Aqueous Update

The Subcommittee received an overview of aqueous separations research in the Material Recovery and Waste Form Development Campaign. The presentation provided a good overview of the history of separations research within DOE-NE in the context of the timeline of initiatives within the Department, and their specific goals and objectives. The Subcommittee enjoyed the discussion of "lessons learned" from these past efforts, and we encourage DOE-NE to capture these thoughts more formally. In addition, the Subcommittee would like to learn more about cost estimates of the various material recovery schemes. The Subcommittee acknowledged the positive integration of the Separations and Waste Form Campaign activities, since there should be an obvious link for optimization between the separated nuclear materials and the tailored final waste forms.

Advanced separations research has been part of most of these programs. Important developments (and demonstrations) have resulted from these efforts, although the development of successive generations of separations processes founded on UREX were acknowledged to add perhaps unnecessary complexity, cost, and risk for engineering-scale implementation. Efforts within the FCT program have now been "rebalanced" to reflect development of the scientific basis for advanced separations and the further development and integration of separations processes into flowsheets.

The identification of preferred fuel cycle options (in association with the fuel cycle options study) continues to support the need for research in this area, aimed at enhancing fuel utilization and reducing waste. Primary areas of R&D include enhancing TRU recycle options, management of U/Pu co-extraction, and management of neptunium and technetium in the fuel cycle; additional needs remain in understanding solvent degradation, improving monitoring of processes, etc.

A goal of the program is a closed fuel cycle by mid-century. Of course, there remains significant uncertainty in the path forward, which translates into lack of technology down-select and plant-scale design. There was thoughtful discussion of comparisons between aqueous and electrochemical reprocessing approaches, dispelling some myths about the applicability of these technologies. Similarly, the presentation emphasized that both approaches are amenable to (and subject to) appropriate safeguards. We agree that work in both areas merits continuation within the portfolio.

We appreciate that the separations program has been undergoing a transition to include work on the development of flowsheets. Even in the face of continued uncertainty, there is still some opportunity to present the metrics driving investment, building on the fuel cycle options study. For the reference technologies, important questions that research should address include: how to reduce risk in scale-up of separations in the preferred fuel cycle options, and are there additional enabling science areas that will remain topics for long-term study? Program researchers indicated that work is prioritized using a set of metrics; the Subcommittee would like to discuss these metrics in a future meeting.

Positive international collaborations were mentioned. However, it was unclear if these are opportunistic collaborations or integrated efforts important to achieving program goals.

International collaboration in R&D has the potential to serve an even more integral role in forwarding the objectives of the program. The Subcommittee was previously briefed about the Joint Fuel Cycle Studies which is an international effort to assess the technical and economic feasibility and non-proliferation acceptability of electrochemical recycling. Within this program, an Integrated Recycling Test is forecast for 2017-2018. This raises the question about the interest and objectives of a parallel and similar aqueous recycling test process, such as the coextraction (COEX[™]) process at the 1 kg engineering scale, which should bring essential elements of comparison between aqueous and electrochemical recycling. As the Campaign is now transitioning back into process (flowsheet) development, the Subcommittee also requested a presentation of the objectives, timeframe and conditions for such an aqueous recycling test at a future meeting.

Emphasis on the value of the educational pipeline through engagement with universities was featured in this presentation. Several NEUP efforts were highlighted. The number and breadth of projects in separations are not as extensive as we might have envisioned; this merits some consideration. In future meetings, the Subcommittee would like to hear about the process used to develop university proposal calls, the efforts used to generate interest from the university research community to respond to the call, and the adoption of university research results in the Material Recovery Campaign.

III. MELCOR Overview and Applicability to ATF Response

The Subcommittee continues to monitor the progress of the Accident Tolerant Fuel (ATF) program, which has been tasked by Congress to pursue the development and qualification of accident tolerant nuclear fuels that would enhance the safety of present and future generation Light Water Reactors (LWRs). The current ATF program is oriented around a ten-year timeline with a fuel prioritization to be made in 2016 and a Lead Fuel Assembly (LFA) or Lead Fuel Rods (LFR) ready for reactor insertion in 2022. In FY2015, the program was allocated \$60 M. House and Senate marks predict similar funding for FY2016. The program is currently exploring multiple ATF concepts. In 2016, the concepts are to be prioritized and selected for the next development and qualification phase. During this meeting, our Subcommittee heard presentations and reviewed available references associated with analysis efforts to evaluate ATF concepts---MELCOR calculations to evaluate plant response during beyond-design-basis accidents (discussed in this section) and Nuclear Energy Advanced Modeling and Simulation (NEAMS) toolkit calculations to evaluate fuel behavior during normal and off-normal events (discussed in Section IV). Results from both types of evaluations are required to characterize ATF concept performance.

The ATF program initially focused on the ability of new fuels to extend the time before initiation of the exothermic oxidation reaction associated with hydrogen generation from zircaloy-based

cladding in current LWR fuel. In prior full NEAC and NEAC AFC Subcommittee meetings, members expressed concerns about the ability of the program to support the 2016 ATF downselection and ultimate insertion of a LFA without fuel performance analyses for normal and offnormal events and during severe accidents. Clearly, new data to characterize material properties for proposed cladding materials are required to complete these analyses. The ATF program is developing models in the NEAMS toolset to evaluate fuel performance during normal and design basis events. However, NEAC members recommended that state-of-the-art plant systems analyses codes, such as the NRC-sponsored MELCOR code or the EPRI-sponsored MAAP code, be applied to assess fuel performance during severe accident events. In addition, our NEAC Subcommittee members emphasized that it is important to improve the accident tolerance of the entire plant (rather than just the fuel). During a 'severe' accident, the performance of other core components may be equally important. One example would be lower temperature relocation of control rod materials that could result in a loss of reactivity control, while another would be oxidation of BWR channel boxes and other steel structures that could result in production of combustible gas (both hydrogen and carbon monoxide).

In response to our request, the ATF program initiated several severe accident evaluations with the MELCOR code. MELCOR is a fully integrated, engineering-scale systems analysis code whose development was led by Sandia National Laboratory (SNL) and funded by the U.S. Nuclear Regulatory Commission (NRC). The MELCOR code has been applied to a wide range of designs, including Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs). Models within the code cover a wide range of accident phenomena, from core heat-up, degradation and relocation, core-concrete interactions, hydrogen production, and fission product release and transport.

ATF MELCOR calculations consider the TMI-2 accident, which is essentially a small loss of coolant accident (LOCA) scenario in a B&W PWR [1] and several mitigated and unmitigated station blackout accidents for the Peach Bottom BWR, which is a General Electric BWR/4 in a Mark I containment.[2] Recent ATF MELCOR evaluations have implemented modifications that allow users to redefine cladding material properties and oxidation kinetics. Analysts are now able to simulate new materials under consideration in ATF fuel concepts, such as silicon carbide (SiC)-based materials and FeCrAl, and are able to model 'over-coats' of these cladding materials as well as entire replacement of the fuel cladding with these new materials. However, BWR MELCOR analyses are still limited in that calculations apply the new cladding material properties to all core components that originally contained zircaloy (e.g., analyses still over-estimate the benefits of new cladding materials because other components, such as channel boxes, are also assumed to be fabricated from these new cladding materials).

Properties for these new cladding materials (e.g., SiC and FeCrAl) were selected based on available data in the literature. There are limited data for estimating the properties of oxides that may form when SiC or FeCrAl is exposed to high temperature steam. For example, the steam oxidation rates of FeCrAl alloys are of concern. The basis for these reaction rates is essentially an empirical fit of experimental data to Ae^{-B/RT}, which is a function of temperature, T, the gas constant, R, and two coefficients, A and B, that are dependent on experimental data. There are also limited data for evaluating the melting temperature of FeCrAl oxide. Data reflecting the degradation of these cladding materials with irradiation are even more limited, especially data that would help characterize the survivability of new cladding materials during the quench process of an accident and its coolability in a degraded geometry. In addition, models do not consider the potential eutectic formation of FeCrAl with B₄C, Inconel, and UO₂. The program recognizes this limitation, and efforts are underway to obtain the additional data.

Most ATF MELCOR calculation results indicate that peak cladding temperatures (PCTs) and hydrogen generation are lower than predicted for fuels with zircaloy-based cladding materials. This effect is attributed to the lower oxidation energy associated with proposed ATF cladding materials and other factors, such as the 50% reduction of cladding thickness proposed for FeCrAl cladding. This 50% reduction in FeCrAl cladding thickness comes from reactor physics concerns related to the need to reduce parasitic neutron capture, and potential difficulties associated with large-scale fabrication have not yet been addressed. Furthermore, the program recognizes there are other metrics that must be considered in evaluating the enhanced safety associated with proposed cladding concepts, such as the timing of ignition events due to the overall production of combustible gas (CO and H₂ from cladding and other core components), the timing of fission gas release, and the timing of vessel and containment failure. Although there is still uncertainty in some severe accident phenomena, MELCOR evaluations can provide insights on these metrics.

Although analysts have implemented ingenious methods to complete ATF MELCOR evaluations, these modifications were not implemented by MELCOR code developers into SNL 'production versions' of this code. Available results from MELCOR calculations raised several questions. As noted above, these modifications still require that the BWR analysis be completed assuming that the same material is assigned to fuel cladding and channel boxes. In addition, available results do not provide confidence in the benefits of some cladding materials. For example, it is unclear if the reduction in hydrogen production predicted in the BWR analysis is due to the assumed properties of the new cladding (and channel box) material or due to the reduced cladding thickness required for FeCrAl-clad fuels. Furthermore, PWR evaluations assuming FeCrAl cladding indicate that PCT predictions are very close to assumed failure temperatures for this material (and data are needed to support this failure criterion). Finally, available results do not address what changes must be implemented to address re-criticality concerns due to

relocation of control materials that may become molten during a severe accident. Our Subcommittee recommends that the MELCOR evaluation effort be expanded. Future calculations should use a production version of MELCOR that allows users to only apply the new cladding materials to the fuel cladding. Furthermore, additional risk-important accident scenarios for PWRs and BWRs should be analyzed to provide a more complete perspective related to the plant safety benefits of proposed cladding concepts. In addition, the Subcommittee recommends that the enhanced MELCOR evaluations include all available high temperature material property data for proposed cladding materials, including data from irradiated samples if available. Finally, the Subcommittee recommends that a detailed technical peer-review be performed on the MELCOR analysis effort to increase confidence in inputs used for the down-selection process of this \$60 M/year program.

IV. The Fuels Product Line: Update on BISON & MARMOT Development

As discussed in Section III, this topic was the second presented to our Subcommittee on analysis efforts supporting the ATF program and the planned FY2016 down-selection effort. This presentation focused on NEAMS toolkit calculations being performed to evaluate fuel behavior during normal and off-normal events.

Fuel performance models, either those that have been developed by fuel vendors for regulatory evaluations or those developed by the regulator, are semi-empirical. They are validated against separate effects and integral data at the engineering scale. As such, they are limited to the range and domain over which the validation data exist. In contrast, the models being developed by the NEAMS program consider atomistic, meso, and engineering scales with the goal of being capable for use outside the limited range of available engineering scale data. Achievement of this goal requires that such models first be developed using meso-scale data and then validated against engineering-scale data. However, it is not practical (from a cost viewpoint) to obtain all of the data required for the models. Hence, the NEAMS program is pursuing a path that relies on first principals models that are developed based on limited meso-scale and engineering-scale data.

The NEAMS Toolkit strives to obtain a "Pellet-to-Plant" simulation capability useful for predicting performance and safety for a broad range of nuclear reactor power systems. The Toolkit is modular in design with components organized under a Fuel Products Line (FPL) and a Reactor Products Line (RPL). Individual components represent key physical phenomena (e.g., neutronics, structural, thermal, and fluid mechanics; and materials science). The FPL toolkit development focuses on delivering an integrated set of mechanistic-based computational tools for fuel performance analysis and design. It uses the Multi-physics, Object-Oriented Simulation Environment (MOOSE) computational framework. Fuel performance simulations with the engineering-scale BISON code are informed by material property and irradiation performance

models developed from meso-scale MARMOT code simulations of microstructure evolution under irradiation. BISON simulations are informed by inputs from fundamental materials parameters obtained from atomistic scale simulations using stand-alone codes. MOOSE is able to run both BISON and MARMOT simultaneously to create a three-dimensional MOOSE-BISON-MARMOT (MBM) simulation that displays microscopic radiation effects evolving into fuel or cladding failures at the macroscopic scale.

The combined physics capabilities of these new tools (e.g., coupled structural deformation, thermal response, and fission gas release) are not possible with existing tools. There are also important benefits associated with the enhanced numerical capabilities and geometrical representations possible with NEAMS tools. However, during our October 2015 Subcommittee meeting, members continued to voice concerns similar to those expressed during previous Subcommittee meetings and by the NEAMS Subcommittee;[3] namely, there is a need to validate new FPL tools. The NEAMS development team acknowledged that funding limitations will preclude them from obtaining all of the data required to validate such models. Hence, the program is focused on validating tools using available engineering scale data. Fundamentally, this is the same process used by existing empirical fuel performance codes, such as FRAPCON, but the NEAMS development team maintains that their process relies on more mechanistic models, allowing the MBM codes to better match available engineering-scale data.

In light of existing funding constraints, the Subcommittee concurs that the proposed approach for assessing the FPL tools is reasonable, but that its limitations must be recognized. The principal limitation is the lack of data for justifying the extrapolation of NEAMS models beyond the range of data over which it is validated. Applications to assist in the down-selection of ATF concepts must recognize that these new tools have limitations similar to those associated with current fuel performance codes that rely on empirical fits to available engineering-scale data. Furthermore, at this time, it does not appear that results from the MBM tools are interfaced with any RPL analysis codes. Hence, at this time, it is not possible to apply the MBM methods to estimate the plant 'safety' benefit of proposed ATF concepts during either design basis or severe accidents.

References

- B. J. Merrill, S. M. Bragg-Sitton, and P. W. Humrickhouse, "Status Report on Advanced Cladding Modeling Work to Assess Cladding Performance under Accident Conditions," INL/EXT-13-20206, August 2015.
- K. R. Robb, "Severe Accident Scoping Simulations of Accident –Tolerant Fuel Concepts for BWRs," 16th International Topical Meeting on Nuclear Reactor Thermal Hydraulics (NURETH-16), Chicago, IL, USA, August 30-September 4, 2015.

3. Juzaitis, Ray, Chair, "NEAC Review: NEAMS, Summary of Subcommittee Report" (December 6, 2012).

V. Advanced Reactor Program

The Advanced Reactor Program is a broad-based research and development program with a top level goal of developing a safe and economical advanced reactor. The current fleet of Light Water Reactors (LWRs) is nearing retirement and the Advanced Reactor program is timed to provide the research and development needed for the replacements for these reactors. Lower level goals supporting this are: (a) to reduce technical, financial, and regulatory risk associated with an advanced reactor, (b) to examine the need for a new test/demonstration reactor, and (c) to work with industry to further advanced reactor development.

An advanced reactor has not been designed and built in the U.S. since the Fast Flux Test Facility (FFTF), which was designed in the 1970s, operated in the 1980s, and was shut down in 1992. Given this, the goals of the Advanced Reactor Program include two important elements. First, to improve on the technology that was used on FFTF, and second, to maintain and preserve the knowledge that was developed for an actual reactor design/build/operate program.

The Subcommittee was informed of a number of subprograms conducted within the Advanced Reactor Program with these objectives. These subprograms are broad sweeping and appropriately span the advanced reactor technology space. Examples are: (a) the Mechanisms Engineering Test Loop with the capability to test advanced liquid metal reactor components in sodium; (b) the Advanced Fuel Handling System with the capability to reduce refueling outage times; (c) the testing of Under-Sodium Viewing equipment which provides the capability to visually examine components under an opaque sodium surface; (d) the FFTF Data Preservation Program which preserves the knowledge gained during the operation of this reactor; (e) the development of advanced alloys such as Alloy 709 with the possibility of allowing higher operating temperatures and hence, higher thermal efficiencies; and (f) the development of the compact Supercritical CO₂ Heat Exchanger Technology which has the possibility of eliminating the potential for sodium-water reactions associated with conventional liquid metal heat exchangers, which in the past has resulted in expensive heat exchanger designs. In addition, the overall program engages in international collaborations with the Chinese, the Japanese, the French, and the Koreans to avoid duplication and to ensure that developments within these programs are not overlooked.

Although it was evident to the Subcommittee that the Advanced Reactor Program's efforts to develop a fast reactor are comprehensive and broad-based, notably absent is a key component, i.e., the existence of a fast flux test reactor in the United States. While the FY-15 Omnibus Spending Bill did include language and funding to examine the need for a test/demo reactor,

the Subcommittee notes that amount of funding (\$7M) is only sufficient for this task and is insufficient to complete a preliminary conceptual design of a test/demo reactor.

The Subcommittee wishes to emphasize the need for a test/demo reactor capable of supplying fast neutrons with a high flux. Such a reactor would be capable of irradiating fuels to a high burnup and materials to high displacements per atom (dpa). A fast reactor with these characteristics could provide capability for all future testing and data evaluation because fast neutrons can be moderated to lower energies when needed, as was demonstrated in FFTF. This will ensure that testing and data evaluation can be conducted for all other applications with an energy spectrum tailored to the application.

VI. Nuclear Fuel Storage & Transportation Program Update and FY16 Planning

The Department continues to pursue a variety of activities related to the implementation of spent fuel interim storage, along with related transportation activities. These activities are being undertaken in anticipation of new authorizing legislation. As discussed at the briefing, the potential for the enactment of such legislation in the near term is still unclear. This area of discussion and the overall presentation were somewhat limited due to the sensitive nature of the subject and the intense public interest in this topic.

<u>Interim Storage Facility Design Development</u> - The Department indicated it is continuing to pursue a canister-based storage facility concept and will give priority to the receipt of canistered spent fuel from shut down reactors. A Topical Safety Analysis Report (TSAR) is being prepared which can facilitate future licensing, assuming that the site parameters of a selected site fall within the boundary conditions of the generic design within the TSAR. No date or schedule was given regarding submission of the TSAR to the NRC, although discussions with NRC staff are apparently ongoing.

The Subcommittee continues to note that two industry-led efforts to develop Interim Storage facilities are also underway. It is still unclear to the Subcommittee how the Departmental-funded design effort is coordinated with these two industry efforts. In addition, the legal responsibility/liability of the Department versus private industry needs to be clarified.

<u>Transportation Activities</u> - The Subcommittee notes that there appear to be underlying assumptions driving the Department's transportation planning, but those assumptions were not discussed and would need to be reflected in any enabling authorizing legislation for the interim storage facility as well as for transportation activities. The Department has repeatedly referenced in this and previous briefings on Nuclear Waste Policy Act (NWPA) Section 180(c) activities, yet that section would only apply to shipments to Yucca Mountain or to an NWPA Monitored Retrievable Storage Facility closely linked to Yucca Mountain Repository development and operations. The Subcommittee strongly recommends that the Department not reference NWPA sections in its presentations, since they are not legally applicable to its ongoing activities.

In addition, the Subcommittee recommends that the Department develop a list of assumptions under which it is developing its transportation activities. Such assumptions could include whether the Department will be self-certifying its transportation cask designs or whether it will seek NRC certification, whether the Department will be following DOE guidelines for physical protection and pre-notification of shipments or whether the Department intends on following NRC regulations in that area, as well as the extent and use of private industry in its planned activities, etc. Such an assumptions list would be helpful as a baseline in planning and schedule development activities, in addition to orienting external audiences regarding the Department's transportation planning activities.

Similarly, the Subcommittee recommends the Department compile a list of assumptions related to the development of an interim storage facility. Issues such as NRC licensing, NEPA compliance, and use of private industry could be addressed in the assumptions list and would also serve as a baseline document for planning purposes, schedule development and public outreach efforts.

VII. Joint EM-NE-SC-International Study of Glass Behavior over Geologic Time Scales

A second presentation from the Material Recovery and Waste Form Development area described an international study on long-term glass corrosion mechanisms. The fuel cycle options study identified several of the most promising options, all of which involve a closed fuel cycle. Both aqueous and electrochemical reprocessing methods result in the generation of small quantities of high level waste, and there is an acknowledged need within the program to support cost effective approaches to the generation of high performance waste forms. Given the significance of vitrification in the current plans for HLW immobilization, it is a long-standing technical grand challenge to demonstrate a technical basis for projections of the long-term behavior of glass (and the potential for radionuclide release of this waste form). There are potential cost-saving implications, both in the selection of the waste form (maximizing loading and decreasing volume) and in creating a defensible basis for licensing. This presentation described an international study, that is based on an evaluation of an "ancient glass" sample in which a set of multiple methods were employed to study composition, structure, and kinetics associated with glass corrosion to develop an improved understanding of the long-term behavior for glass waste forms. The characterization approach was then extended to evaluating glass ceramics as alternative waste forms with improved waste loading tolerance. R&D in this area is already leading to an improved mechanistic understanding. It is important to note that this is a challenging problem, and the conditions studied to date are limited. It defines a successful research approach, however, that can be extended to look at additional

families of waste forms, radiation loadings, and a wider variety of potential repository conditions. Beyond the understanding and the knowledge of the performances of glass and glass ceramics, additional research should also be pursued to connect their corrosion properties with the near field geological characteristics, since both waste form and repository conditions have to be considered in a synergetic way for optimization of long term disposal.

The study made effective use of resources from multiple partners (both international and within DOE), and had an important goal of helping to arrive at international scientific consensus. This topic is well suited to the collaboration described, and the areas of research also benefit by interacting with a number of NEUP projects (engaging universities and students in the work). A FOA has been released for a joint IRP (IRP-FC-EM-1). The level of cooperation and integration in this area is to be commended.