DOE Nuclear Separation Technologies – 15148

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ABSTRACT

Realizing that challenges are facing US actinide science, separations, and waste form research and development activities including meeting future manpower needs, the U.S Department of Energy (DOE), Office of Environmental Management (EM), Office of Nuclear Energy (NE), and National Nuclear Security Administration (NNSA) jointly co-sponsored a workshop entitled "Separations Technologies: "Going from Where We Are to Where We Want to Be" on nuclear separations technologies in Bethesda, Maryland (July 2011) to identify common needs and potential requirements in separations technologies and opportunities for program partnerships. Recognizing the different mission requirements of the sponsoring programs, participants were able to identify areas with common uses and related applications.
The two follow-on technical workshops held during the Fall 2012 addressed both the chemical aspects and the physical aspects of nuclear separations. Each workshop consisted of technical discussions regarding future work relevant to DOE goals. The large number of subject matter experts from the national laboratories, academia, and headquarters staff developed a long list of existing and new technology developments in the areas of aqueous separations, pyrochemical processing, waste forms, fuel technology etc….Each of these workshops identified the current state of knowledge or technology, the resources needed, technology development opportunities, and the objectives to reach the desired end-state(s). A consolidated report with its eight appendices will be issued at the end of December 2014 summarizing the outcomes of these workshops. Some examples of these findings will be presented in our contribution. It is clear that, in the current environment of decreasing budgets it is illogical for NE, EM and NNSA to develop its own homegrown technology. Therefore, there is a strong case to be made for leveraging limited resources in order to accomplish all objectives while significantly reducing development costs.

INTRODUCTION

Solution-based separations are integral to the missions of three offices within the U.S. Department of Energy (DOE), Office of Environmental Management (EM), Office of Nuclear Energy (NE), and the National Nuclear Security Administration (NNSA). Nuclear separations technologies cut across these DOE organizations despite the differences in mission. Even though the specific needs of these three offices vary widely, as do the timelines upon which they are operating, the missions of NE, EM and NNSA depend on research and development (R&D) of new technologies that will safely and securely accelerate schedules and reduce lifecycle costs. The scales at which aqueous separations are applied also vary significantly, from analytical separations at the microliter scale required for some NNSA applications, to hundreds of metric tons per year processing of commercial fuel ultimately required by DOE-NE, to the 3.4×10^5 m^3 of radioactive wastes stored in tanks at the Hanford site and Savannah River Site (SRS) that DOE-EM must process and immobilize. Similarly, the concentrations involved range from ultra-trace level detection of radionuclides to hundreds of grams per liter, and the complexity of the solution matrices can vary from simple dilute solutions (groundwater) to highly concentrated electrolyte mixtures (salts, acids, bases, complexants, and elements across the periodic table). Despite this disparity, common themes exist that tie together the missions of DOE-EM, DOE-NE, and NNSA.
On July 26- 28, 2011 DOE hosted an initial Workshop [1] for subject matter experts from the national laboratories, academia and industry to evaluate the state of the art in current nuclear separations

technologies and determine the a path forward for where DOE would like be and what they would like to attain in the next three to five years. The objectives of the workshop were to (a) identify common needs and potential requirements in nuclear separations technology and opportunities for program partnerships, and (b) evaluate the need for a DOE nuclear separations center of knowledge to meet the requirements of NE, EM, and NNSA. SC, which establishes basic research needs and funds fundamental science programs in relevant areas of actinide separations. The July 2011 Workshop report concluded that NE, EM and NNSA had established the foundation for cooperation and communication in nuclear separations technologies and what the next steps would be to follow-up with concrete actions to integrate these technologies across Offices. This would lead to optimizing resources and ultimately, the generation of a DOE "Center of Knowledge" for nuclear separations technologies. Based on the foregoing NE, EM and NNSA organized two follow-up workshops to develop a roadmap for nuclear separations technologies.

The second workshop held in September 18-20, 2012 in Gaithersburg, MD focused on chemical separations with discussions framed around aqueous separations, gas-phase chemistry, and high-temperature processes. The third workshop held in December 4-6, 2012 in Germantown, MD focused on physical separations and covered physical characterization of flow sheet starting points, interim- and end-state products, and process engineering.

To capitalize on opportunities for program partnerships, common needs as well as potential requirements in nuclear separations technologies were identified. Participants were able to identify areas with common uses and related applications, as discussed below.

NUCLEAR SEPARATIONS TECHNOLOGIES WORKSHOP (JULY 2011)

A number of common need and potential requirement in nuclear separations were identified during this workshop. A few examples are listed below.

Chemistry and Speciation of Actinides and Key Fission Products (Cs, Tc, I)

Understanding the underlying physical and chemical properties of given materials is the basis for all separations. In addition, the area of nuclear separations involves the need to understand how these properties change in the presence of a radioactive environment. Understanding the chemistry of actinides is a high priority because the actinides are uniquely important to DOE, and have complex chemistries that are less well explored and more difficult to model than most other elements. A better understanding of actinide and fission products chemistry will impact a variety of challenges facing DOE, such as monitoring actinides during processing or minimizing technetium volatilization in waste treatment. To help predict process performance and behavior, understanding the fundamental effects of radiolysis on solvents, ligands and other separations molecules will enable development of advanced models of radiation resistance, decomposition, and formation of radicals. In addition, advanced tools are needed to predict the molecular structure of actinides and fission products in extreme systems, ranging from ultra-low-concentration contaminants to complex environments at high temperatures.

Design of Molecules with Selective Nuclear Separations Capability

The development of nuclear separation technologies has historically involved the empirical screening of chemical compounds for their ability to perform separations. The last decade has seen a maturity in the technology to synthesize 'designer' molecules and materials for a specific function. Improved predictive science that couples experimentation with modeling and theory allows researchers to design molecules and materials that precisely target particular nuclear separation requirements. Breakthroughs of this type require a full-cycle, systematic approach by teams with diverse capabilities from molecular design to synthesis to chemical engineering. Advancing this design approach supports multiple DOE separations needs

Interface Synergies between Nuclear Separations and Waste Management

Whether separating the contents of a high-level waste (HLW) tank for stabilization and disposition or separating used nuclear fuel (UNF) for fuel material recycling, the interface between the different processes and the waste forms that they produce is a critical consideration when reducing costs and optimizing the overall operation. Efficiencies in one area may lead to complications in other areas. A systems analysis approach is therefore essential to guide R&D. Tools that capture functions and requirements of a process must be developed to improve synergies between separation processes and follow-on processes. For example, developing a separation technology commonly involves chemicals that function as complexing agents, such as iron. If subsequent steps in this process entail using borosilicate glass for waste disposal, the presence of iron will limit the ability to maximize waste loading, reducing efficiency and ultimately increasing the overall costs. Understanding the impact that each step has on the overall process and capturing interface synergies will result in improved overall performance and reduced costs.

NUCLEAR SEPARATIONS TECHNOLOGIES WORKSHOP (OCTOBER AND DECEMBER 2012)

The purpose of these two Separations Workshops was to outline current nuclear processes and practices for NE, EM and NNSA, and identify technology and other gaps necessary to move to the next step. The participants defined the objectives and challenges within their respective area(s) of expertise. The groups followed the same format of break-out sessions with chairs and co-chairs having developed a list of topical questions to generate a formative discussion between their group members and identify relevant separations processes practiced at a site or national laboratory. The six break-out sessions for the two workshops conducted had substantive discussions which were held in each of the break-out sessions with detailed reports prepared summarizing the results of their discussions; these reports describe the current state of technology and the desired path forward for the timelines discussed and are included as Appendices A-H accompanying the consolidated report [2]. The Appendices (see Figure 1) which will be accessible on line provide a list of new technologies that will assist in the future the three offices with their respective missions and strategies.

<table>
<tr><th>Processing</th><th>Material States</th><th rowspan="5">Process Engineering [Appendix F]</th></tr>
<tr><td>In Aqueous Solutions [Appendix A]</td><td rowspan="2">Starting States [Appendix D] = Reactants</td></tr>
<tr><td rowspan="2">In Gas Phase [Appendix B]</td></tr>
<tr><td rowspan="2">Interim and End States [Appendix E] = Products and Wastes</td></tr>
<tr><td>At High Temperatures [Appendix C]</td></tr>
</table>

Cross cut **Modeling and Simulation [Appendix G]**
Cross cut **Characterization via Sensors, Instrumentation, and Analytical Methods [Appendix H]**

Figure 1. Organization of the Science and Technology as outlined in the consolidated report [2]

The first three appendices (Appendices A, B, and C) pertain to processing methods, and are divided into three categories:

A. Processing in aqueous solutions, which are based on nitric acid

B. Processing in the gas phase

C. Processing at high temperature, e.g. electrochemical salts

Aqueous processing options have been the most well studied in the US and elsewhere, for decades. Nonetheless, there are still key issues as pertain technologies and science to support US objectives. Processing in the gas phase is the least studied. Although gas phase processing occurs as high temperatures, it is separated from the third category because of the innovation and exploration involved.

The next two appendices (D and E) pertain to material states, the starting points or reactants, interim states, and final states. Interim states include materials intended for long-term storage. Final states can be useful products or waste destined for disposal.

The sixth appendix F involves overall process engineering. This is followed by two cross-cutting appendices G and H – essentially software and hardware. Thus, Appendix G addresses modeling and simulation. Appendix H addresses characterization of material states and processing via sensors, instrumentation, and analytical Methods. Table 1 lists all 8 appendices with specific questions that must be answered to advance the aqueous separation challenges faced by primarily DOE-NE and DOE-EM. There is some overlap between the NE-related questions and those faced by NNSA; but it should be kept in mind that this paper as well as the workshops did not comprehensively address NNSA research needs.

Table 1. Questions that must be answered to advance the aqueous separations challenges faced by primarily DOE-NE and DOE-EM

	Processing in Aqueous Solutions (Appendix A)	Processing in the Gas Phase (Appendix B)	Processing in High Temperature Conditions (Appendix C)	Physical Characterization of Starting States, Reactants" (Appendix D)	Process Interim and End State Products (Appendix E)	Process Engineering (Appendix F)	Modeling and Simulation (Appendix G)	Characterization via Sensors, Instrumentation, and Analytical Methods (Appendix H)
What is it	Using solutions of water-based nitric acid to dissolve used nuclear fuel and separate it into different products and wastes	Using mixtures of reactive gases change the chemistry of used nuclear fuel and separate it into different products and wastes.	High-temperature processes are critical components to enabling treatment of used nuclear fuel and handling the resulting nuclear waste from fuel treatment. Examples of those are: Electrochemical Processing, Vitrification Advanced Waste Forms and Waste Form Processes	Existing nuclear materials must be well characterized regarding their physical and chemical properties.	Interim (or intermediate) products and end state (or final state) products are the results of processing used nuclear fuel and nuclear wastes. The performance, packaging, disposal, and monitoring of these products must be understood.	Process engineering is making the process science and technology fit together in a practical engineered system	Modeling and simulation can model very complex chemical processes that help understanding complicated phenomena	Suite of sensors, instrumentation, and analytical chemical and radiochemical techniques complement the understanding gained from modeling and simulation.

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Why is it needed	Specific needs of EM, NE and NNSA offices vary widely. For example, understanding radionuclides' transport in the environment and their subsequent recovery is normally considered part of the EM mission; however, NNSA needs similar chemistry to detect nuclear weapons activities, as does NE to model transport of radioactivity from disposal sites.	Gas phase processing offers the potential to perform valuable separations without generating liquid wastes	Aqueous separation is well understood, but with known unresolvable limitations. Gas phase processing has potential, but is highly innovative. Advanced waste-forms processes are needed to treat any waste streams coming from advanced recycling technologies.	The better one characterizes the reactants to be processed, the most effectively, economically, and safely one can process the reactants	Interim and end state products are an important consideration to enabling treatment of used nuclear fuel and handling the resulting nuclear waste from fuel treatment.	Bench top studies are necessary to study new processes but ultimately engineering scale studies will support the expansion of recycling technologies	There is a need to share M&S at the molecular scale, microscale, systems scale, and unit operations scale.	Tremendous potential exists for improvements in sensors and instrumentation—along with the characterization and analytical methods needed to interpret and utilize the resulting information—with potential impact across a wide range of diverse activities

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Objective	Virtual separations capability to respond to specific nuclear separations challenges faced by DOE as they arise.	Exploration of gas phase separation science and technology to determine its potential to address separation needs.	Electrochemical processing and vitrification are of highest priority for successful development of an advanced fuel cycle	Characterization of reactants will bring an order of magnitude improvement in separations technologies	The science and technology must be adequate to meet operational, safety, security, and non-proliferation requirements	Investment in process engineering capabilities is required to successfully fulfill DOE missions. Specifically, DOE must maintain core capabilities in Liquid and slurry retrieval, transfer, and mixing (particle and rheology characterization), 2-Dissolution processes, 3-Physical and chemical separation processes	Improvement of modeling and simulation sufficient to improve guidance of experimental program planning, to help down-select options, to improve the safety of experimental operation.	To exploit the clear potential that will result in increased efficiency, throughput, and materials accountability with the main goal of significant footprint and cost reductions. An additional main driver is to maintain and enhance the highest level of safety and risk reduction

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challenges	Understanding and controlling molecular chemistry of actinides and fission products in media of inherent complexity. Understanding and mitigating the effects of radiation chemistry in separations processes. Improving and validating molecular modeling and simulation and other prediction approaches. Understanding the properties of interfacial systems to address interfacial problems that deter scale up.	The basic challenge is to quickly, efficiently, and effectively explore the potential of different categories of processing. Reaction of a gas with a solid to achieve a key separation of interest resulting in one or more gaseous products and solid products Reactions of a gas with a solid to remove species from the gas phase Reaction of a gas with a liquid to remove species of interest into the gas phase or to make a species remain in the liquid Reaction of a gas with a liquid to remove species from the gas stream Reacting of a gas with a gas, and possible roles of catalysis	Which constituents are most likely to dictate waste form selection and loading? What advances in high temperature processes can be made to reduce these limitations?	Three main areas, each with their characteristic challengesare: Tank Waste Slurry Sampling Nuclear Materials and Used Nuclear Fuel Soil and Groundwater Sampling and Monitoring	The science and technology must be adequate to meet operational, safety, security, and non-proliferation requirements.	An over-arching challenge is the level of knowledge of most of the processing options. Aqueous processing is the most well understood and therefore it is the focus of most of the current process engineering analysis.	The objective is to improve modeling and simulation sufficient to improve guidance of planning of the experimental program, to help down-select options, to improve the safety of experimental operation.	Key opportunities and challenges for nuclear separations will take advantage of existing technology in the areas of sensors and analytical tools, incrementally translating them into the nuclear energy domain. At the same time, identifying research areas ripe for investment will result in entirely new systems that support safe, efficient, and improved nuclear energy utilization of nuclear energy and waste management implementation. The areas of sensors, instrumentation, characterization, and analytical methods are fundamental to nearly all science and technology areas that underpin nuclear separations now and in the future.

CONCLUSION

The Nuclear Separations Technologies Workshops have established a foundation of cooperation in this important area between EM, NE, NNSA and SC. The final consolidated report with its 8 appendices will be released in December 2014. Despite differences in the missions of EM, NE, and NNSA, there are common themes that tie together the missions of these three offices but more discussions and broad participation will be needed to advance the concepts and ideas introduced during the workshops. It is clear that DOE must maintain core capabilities in different separation-related areas. In addition, certain core facilities must be available to support R&D work. These include both radiological and nuclear facilities, including hot cells and alpha-active gloveboxes, and engineering-scale facilities (radiological and non-radiological). Finally, continuity of funding to support these efforts is critical to successfully meeting the challenges of restoring legacy production sites, developing sustainable nuclear fuel cycles, and developing effective international safeguards technologies and systems that enhance detection of diversion or misuse of nuclear materials. The next step is to establish the plans and strategies necessary to develop the cooperation initiated during these workshops into a sustainable collaboration between the sponsoring programs and to refine the concept of a nuclear separations center of knowledge that responds to DOE needs, improves program partnerships, and supports the effective use of available funding and resources.

REFERENCES

[1] "Nuclear Separations Technologies Report" July 27-28, 2011: Information and Documents from the initial July 27-28, 2011 Nuclear Separations Technologies Workshop can be found at: http://events.energetics.com/NuclearSeparationsTechnologyWorkshop/documents.html

[2] Consolidated Report of the Follow-On Workshops to the 2011 Nuclear Separations Technology Workshop, to be published, anticipated date December 2014.