

ENHANCING EFFECTIVENESS OF EMSP PROJECTS THROUGH STRONG CONNECTIONS TO SITE PROBLEMS

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ABSTRACT

The Environmental Management Science Program (EMSP) funds basic science research that will lead to reduced remediation cost, schedule, technical uncertainties, and risk for DOE's environmental clean up. The Tanks Focus Area (TFA) has partnered with EMSP to accomplish those same objectives for DOE's largest and most expensive remediation effort – to retrieve and immobilize the highly radioactive wastes that are our nation's chief nuclear defense program legacy. TFA has been tasked to facilitate success of the EMSP investment.

The key for EMSP projects to contribute to this remediation effort is communication. First, scientists need to understand much more about how their scientific results would be used than they could ever learn from the original EMSP solicitation or by reading the referenced DOE needs statements. Second, the scientists' results must be communicated to the site problem holders in a usable form and in a timely manner such that important information gaps can still be filled by the EMSP project. Research results can be used in a variety of ways besides deployment of new hardware or a new process. When results are USED the site problem holders become “users”. The important aspect that research results are to be used is captured in the TFA lexicon for their clients, the DOE sites--“users”.

The best method observed, so far, to accomplish the indispensable communication necessary for success is through direct contact between EMSP researchers and TFA/site problem holders, person to person. The observation that direct contact is the best medium for exchange of complex information may seem inately obvious. However, it is not the normal procedure in the more academic world of the fundamental scientists, where publishing of results in a peer-reviewed journal completes the transmittal of scientific results. Direct and ongoing communication between EMSP researchers and site users doesn't occur naturally. TFA actively bridges this gap between science and technology development and site users through its technology integration managers (TIMs). TIMs are experienced researchers, usually from DOE's national laboratories, with intimate knowledge of the needs of sites at the working level. They help focus the research projects to develop technical solutions to site submitted needs. They stand in a rather unique position with one foot in the research community and the other foot “in the tanks”.

This paper will show, through several examples, significant contributions EMSP scientists have made to solving DOE's high-level waste challenges through direct and enhanced communication with TFA and site users.

INTRODUCTION: EMSP and TFA Mission

The Environmental Management Science Program (EMSP) is a collaborative partnership between the DOE Office of Environmental Management (DOE-EM), Office of Science (DOE-SC), and the Idaho Operations Office (DOE-ID) to sponsor basic environmental and waste management related research. The mission of the EMSP is to develop and fund a targeted, long-term research program that will result in transformational or breakthrough approaches for solving the Department's environmental problems. The EMSP has harnessed the nation's scientific talent and focused it on critical EM problems in order to (1):

- Lead to significantly lower cleanup costs and reduced risks to workers, the public, and the environment over the long term.
- Bridge the gap between broad fundamental research that has wide-ranging applicability, such as that performed by the EM Office of Science and Technology, and needs-driven applied technology development conducted by the EM Office of Science and Technology (OST) Focus Areas.
- Serve as a stimulus for focusing the nation's science infrastructure on critical national environmental management problems.

DOE field sites have identified key science and technology needs that must be addressed in order to meet cleanup goals set forth in EM's plan to achieve closure at DOE sites, *Accelerating Cleanup: Paths to Closure* (2). With the assistance of the EM Focus Areas, the EMSP evaluates these needs and sets research priorities in seven major problem areas:

1. High-Level Waste
2. Transuranic and Mixed Waste
3. Subsurface Contamination
4. Deactivation and Decommissioning
5. Spent Nuclear Fuel
6. Nuclear Materials
7. Ecology/Health/Risk

In terms of life cycle cost and schedule, High Level Waste (HLW)¹ is the largest problem area targeted by EMSP. In 1998, life cycle costs were estimated to approach \$50 billion, more than twice any other problem area; and work was expected to continue for more than another 40 years, through 2046 (2). By 2001, the estimated costs and duration have risen to \$73 billion and 2070, respectively (3,4). The magnitude of the clean up cost and the duration to complete the clean up make HLW an important arena for EMSP in which to accomplish its primary goals-- reduce cost, schedule, and technical risk.

Partnering with EMSP and sharing the same primary goals to reduce cost, schedule, and technical risk is the Tanks Focus Area (TFA). TFA is one of five focus areas established by Environmental Management (EM) in 1994 to unify EM's distributed technology development activities under a single umbrella and to focus the investments on the major clean up areas identified by EM. The intent of the Focus-Area-centered approach is to make EM's science and technology (S&T) investments "solution oriented" and an integral part of clean up (2). The

Focus-Area-centered approach places full responsibility for all investments, science through deployment, under the management of the Focus Areas (5).

The DOE has approximately 340 million liters (90 million gallons) of waste in the form of solids, sludge, and liquid stored in 287 tanks (6). The current DOE cost estimate of converting these liquid and solid wastes into stable forms for shipment to a geological repository exceeds \$50 billion to be spent over the next several decades. TFA is responsible for managing, coordinating, and leveraging S&T development activities across the DOE complex to support remediation of its five major tank sites: the Hanford Site in Washington, the Idaho National Engineering and Environmental Laboratory in Idaho, the Oak Ridge Reservation in Tennessee, the Savannah River Site in South Carolina, and the West Valley Demonstration Project in New York.

As the tanks approach the end of their design life, the possibility of waste escaping to the environment increases. Sixty-eight tanks are known or are suspected to have leaked waste to the surrounding soils at the Hanford Site (67 tanks) and Savannah River Site (1 tank). Some of the tank contents have reacted to form flammable gases, introducing additional safety risks. To minimize the risk of waste migration and/or exposure to workers, the public, and the environment and to meet the regulations entered into by DOE, the waste in these tanks must be retrieved and the tanks closed.

TFA's technical scope covers the major functions that comprise a complete tank remediation system: waste storage, waste retrieval, waste pretreatment, waste immobilization, tank closure/waste disposal, with safety, characterization, and monitoring (of waste, tanks, and processes) integrated into all the functions. In addition to integrating with EMSP, TFA integrates program activities with a number of organizations that provide science and technology development, including DOE national laboratories, DOE cross-cut programs, universities, and international research organizations. The key integration for TFA, however, is with the users, the site problem holders. This key integration principle is captured in the TFA mission: *"The TFA Mission is to work with **users** to develop, implement, and deliver technical solutions through and integrated approach to safely and efficiently accomplish tank waste remediation at five major DOE tank sites."* (6)

The relationship of TFA to technology developers and the DOE sites is illustrated in Figure 1. The left-hand side of Figure 1 shows that sites own the primary responsibility for site clean up and working with all the parties that have a vested interest in site remediation. The sites bear the responsibility on behalf of DOE to establish clean up strategies acceptable to the multitude of interested parties and then to execute those strategies. To accomplish this huge task, the sites need science and technology. They need data and scientific understanding to define the wastes in the tanks, determine its impact on the environment and risks to populations for all the possible scenarios to manage the waste, and they need data, science, and technology to select, design, build, and operate facilities to remediate the site. TFA *supports* the sites to fulfill their science and technology needs.

To accomplish this, TFA plans and manages an S&T development program of approximately \$65 million per year to address needs that the sites have identified to accomplish their clean up missions. TFA interfaces with S&T providers across the country, indeed around the world, to bring S&T to the sites to use to solve their problems. *Use* is the key concept in the vision, and a fundamental aspiration embedded in the program vernacular --the sites are *users*.



Fig. 1. TFA Integrates Science and Technology Providers and DOE Problem Holders.

The real value of the S&T development program comes when sites *USE* the results to solve their problems. Research results are used in a variety of ways to contribute to HLW management:

- Technology (processes or hardware) for deployment
- Scientific and engineering data to support decision-making
- Technical expertise and assistance (7)

RESEARCH INTEGRATION PROCESS—Getting Science & Technology Results Used

In the center of Figure 1 at the interface between the sites (users) and the TFA program, are the Technology Integration Managers (TIMs). TIMs are experienced researchers, usually from DOE national laboratories, with an intimate knowledge of site needs at a working level. The TFA's technical work is organized into six functional areas, each with a TIM directing the work in that area: safe waste storage (safety), waste retrieval, waste pretreatment, waste immobilization, tank closure, and characterization of the waste at all stages of processing. These functional areas cover the complete tank waste remediation system.

Each year site users reassess their situation and update a set of science and technology needs. Each need is assigned to the appropriate functional area and the TIM of that area ensures that: 1) a response is prepared incorporating a sound technical approach to address the users' problems, 2) integrated technical solutions are available to meet the users' schedules, 3) technical solutions are useful to more than one site or more than one application wherever possible, and 4) users are integrally involved throughout the development of a technical solution.

The TFA bridges the gap between scientific research and site needs, which are applied by very nature, through the TIMs. They ensure that EMSP projects are closely linked with the Focus Area and EM clean up mission, and EMSP research results are transitioned in a timely manner into the Focus Area Science and Technology (S&T) program. They assist researchers by providing first hand knowledge of the issues and challenges that exist in the field. Collaboration between EMSP and other TFA researchers to share new knowledge, resources, and lessons learned is strongly encouraged.

Workshops are conducted as a forum for information exchange between EMSP investigators, TIMs, and site problem holders where EMSP scientists receive information on applying their research to tank remediation. By facilitating interaction between researchers and end users throughout the research process, avenues are created to more effectively apply science to the myriad of issues faced by site users to safely and efficiently remediate radioactive waste.

The objective of the EMSP integration process for TFA, like all S&T development activities is to get the results used. However, the EMSP integration process is different from the other S&T development activities in that the timeframe for their use may be longer term. Whereas other development activities directly address needs submitted by the sites, with answers needed typically within 1-3 years, EMSP activities are directed toward longer-term needs and broader issues. Nevertheless, value from the investment will be realized when the results are used.

EMSP RESEARCH SUCCESSES SUPPORTING EM CLEAN UP

This section describes several successful EMSP projects that have already produced results that sites have used in their decision making to solve tank remediation issues.

Impact of Three EMSP Projects at the Savannah River Salt Processing Project (8): *Next Generation Crown Ethers (EMSP #55087, #73803); New Silicotitanate Waste Forms (EMSP #60345, #73748); Foaming in Radioactive Waste Treatment (EMSP #60143)*

All waste tanks at the Savannah River Site (SRS) must be empty by 2028 to comply with the Site Treatment Plan and Federal Facilities Agreement. To complete this mission, the HLW system at SRS must retrieve the tank waste and convert the HLW into solid waste forms suitable for disposal. Both the long-lived and short-lived radioisotopes in the waste will be incorporated into borosilicate glass (vitrified) in the Defense Waste Processing Facility as a precursor to transporting the material for disposal at the national HLW repository. Because the HLW vitrification process is very expensive, the SRS implementing technology must limit the volume of high-level waste glass produced by removing a significant portion of the non-radioactive salts.

These salts can then be classified as incidental waste for subsequent on-site low-level waste disposal, which is far less expensive.

The In-Tank Precipitation (ITP) process for salt waste treatment was originally selected as the treatment baseline based on successful demonstrations with actual SRS salt waste in the 1980s. The ITP process separates cesium isotopes from solution using tetraphenylborate precipitation. The decontaminated salt solution with nearly all of the radioactivity removed can be grouted into a low-level waste form at the site's Saltstone Facility. During radioactive startup of the ITP facility in 1995, higher than predicted releases of benzene occurred due to catalytic degradation of the tetraphenylborate. In January 1998, WSRC informed the DOE that extensive chemistry testing demonstrated that the existing system configuration could not cost-effectively meet the safety and production requirements for the ITP facility. WSRC recommended that a systems engineering team conduct a study of alternatives to the current system configuration. In February of the same year, the DOE Assistant Secretary for Environmental Management (EM) approved a DOE-Savannah River (DOE-SR) plan-of-action to suspend startup-related activities and undertake a systems engineering alternatives study to ITP. Subsequently, DOE-Savannah River (SR) directed WSRC to perform an evaluation of alternatives to the current system configuration for high-level waste salt removal, treatment, and disposal. On March 13, 1998, the WSRC high-level waste management division chartered a team to systematically develop and recommend an alternative method and/or technology for disposition of high-level waste salt. The team conducted an extensive systems engineering evaluation of over 140 alternative cesium removal processes and reduced the list of candidates to four:

- Crystalline Silicotitanate Non-Elutable Ion Exchange
- Caustic Side Solvent Extraction
- Small Tank Tetraphenylborate Precipitation
- Direct grouting (with no cesium removal).

Further review eliminated direct grouting as an option. In 1999, DOE-Headquarters (DOE-HQ) asked the National Academy of Sciences (NAS) to independently review the Department's evaluation of technologies to replace ITP. As a result of the NAS review, DOE agreed that further research and development on each alternative was required to reduce technical uncertainty prior to a down select. DOE-SR also held back the issuance of the Draft Supplemental Environmental Impact Statement on SRS HLW treatment alternatives pending further development of salt processing technology alternatives (9).

In March 2000, DOE-Headquarters requested that TFA assume management responsibility for the Salt Processing Project (SPP) technology development program at SRS. In essence, TFA was requested to review and revise the technology development roadmaps, prepare a comprehensive Research and Development Program Plan, and develop down-selection criteria for three candidate cesium-removal technologies for the SPP at SRS. The three-cesium removal candidate technologies were:

- Crystalline Silicotitanate (CST) Non-Elutable Ion Exchange
- Caustic Side Solvent Extraction (CSSX)
- Small Tank Tetraphenylborate Precipitation (STTP). (9, 10)

The importance of integration with EMSP is clear considering that two (CST and CSSX) of the three potential processes directly relate to research conducted under EMSP. The researchers from these EMSP projects were funded through TFA to bring their expertise and creativity to the development and down selection process for this critical DOE project. The third process (STTP) experienced foaming issues and the EMSP researcher investigating foaming was brought in to help evaluate and select optimum anti-foaming agents (see projects #60143 & #81867).

EMSP Project Numbers: 55087, 73803, *Next Generation Extractants for Cesium Separation from High-Level Waste: From Fundamental Concepts to Site Implementation*, **Lead PI:** Dr. Bruce A. Moyer, Oak Ridge National Laboratory

Referred to as Alkaline-Side CSEX (Cesium Solvent Extraction), the process provides the first practical application of calixarene-crown extractants to treatment of HLW by solvent extraction. An effective form of the extractant was first synthesized at ORNL, and a recent centrifugal-contactor demonstration at Argonne National Laboratory proved to have economic viability. Results from the batch and engineering tests showed that the stringent requirements of a 40,000-fold reduction in Cs-137 activity in the waste and a 12-fold concentration increase in the Cs stream going to HLW could be readily met. In addition to meeting these SRS decontamination and concentration needs, key advantages of the CSSX process include:

- It does not require adjustment of the waste feed stream.
- Extraction is very selective.
- Scrubbing and stripping of the solvent can be accomplished with very dilute acidic solutions.
- It is compact and involves liquid streams.

These advantages reduce costs by minimizing consumption of chemicals, secondary waste production, volume of vitrified waste form, and plant space. The cesium-concentrated stream produced by the process is expected to be so pure that it will require negligible downstream processing and will have negligible impact on the volume of the final vitrified waste form, which is costly to produce and store.

Based upon the down selection R&D and subsequent management recommendations, DOE selected the CSSX technology as the preferred cesium removal technology. This selection was documented in the SRS Supplemental Environmental Impact Statement (11,12) and the official Record of Decision (13).

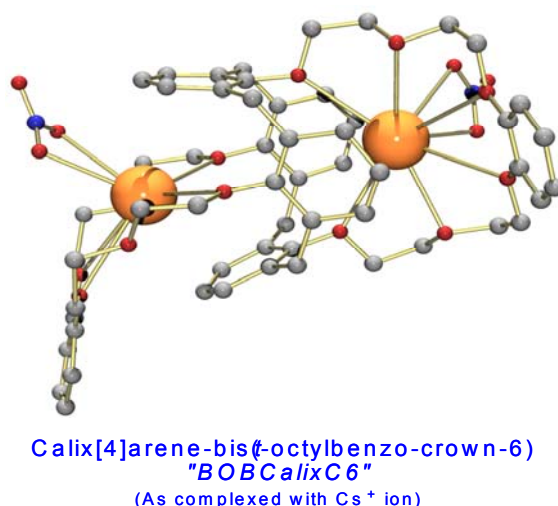


Fig. 2. Calixarene-crown extractant developed under EMSP funding.

The selection of CSSX as the preferred cesium removal technology validates a tremendous success for the technology developers, champions, and programs that supported the technology development over several years. The concepts, which ultimately became CSSX, were originally investigated under basic DOE research programs, Basic Energy Sciences (BES) research, and later under the Efficient Separations and Processing Program (ESP). Under a FY96 EMSP award, “*Design and Synthesis of the Next Generation of Crown Ethers for Waste Separations: An Inter-Laboratory Comprehensive Proposal*” (#55087), researchers investigated the fundamental mechanism of cesium extraction so as to understand the nature of the complexes formed between the cesium ion and the extractant ligand. This fundamental information played a crucial role in successful process development. At the time of the 1998 assessment of 140 cesium separation alternatives, the CSSX process had a critical process obstacle, the Calixarene ligand bound the cesium so tightly that it could not be readily removed from the organic to regenerate and recycle the extractant. The EMSP project provided the pivotal insight that nitrate addition to the stripping solution would enable proper stripping of the cesium from the extractant. Through this insight, the process development could advance and be tested so that it could qualify for consideration in the Salt Processing down selection (14).

In FY00 the EMSP project was renewed as “*Next Generation Extractants for Cesium Separation from High-Level Waste: From Fundamental Concepts to Site Implementation*” (#73803). The EMSP projects have kept the scientists actively engaged developing the fundamentals of the process and maturing the process sufficiently to be considered among the candidates for down selection in the SPP project. During that critical time in 1998, ESP and EMSP were supporting the development of the process and a viable concept to strip the extracted cesium from the organic solvent was discovered. Nevertheless, the technology was judged to be “too immature” and that further scientific development was necessary (15). In the meantime, EMSP scientists continued to support the engineering developers to bring the technology from one considered “too immature” to the prime candidate upon which to base the design of the Savannah River SPP, a DOE investment approaching \$1 billion.

EMSP Project Numbers: 60345, 73748, **Project Title:** *New Metal Niobate and Silicotitanate Ion Exchangers: Development and Characterization*, **Lead PI:** Yali Su, Pacific Northwest National Laboratory, **Co-PI(s):** Dr. Alexandra Navrotsky, University of California at Davis; Dr. Tina M. Nenoff, Sandia National Laboratories - Albuquerque

Crystallinesilicotitanate (CST) was also a down selection candidate for cesium separation at Savannah River. CST, like CSSX, had a history of development through several programs including EMSP before a process based on CST was intensively developed during the SPP down selection program. In that process, CST was used as a cesium absorber only. The full system was based upon subsequent vitrification of the CST into borosilicate glass in the Defense Waste Processing Facility (DWPF). The results of EMSP investments were used to make decisions and brought forward a viable alternative for DOE Environmental Management (EM) to solve a significant clean up problem. Above and beyond that, there are still other potential applications of CST that may yet emerge from scientific research. New research is focusing on its suitability as a final waste form for some wastes. Another EMSP project, “*Strategic Design and Optimization of Inorganic Sorbents for Cesium, Strontium, and Actinides*” (#81949) is

investigating methods to modify the crystal structure to make it selective for Sr and/or actinide separations.

EMSP Project Number: 60143, *Project Title: Foaming in Radioactive Waste Treatment and Immobilization Processes*, **Lead PI:** Dr. Darsh T. Wasan, *Illinois Institute of Technology*

During the intensive SPP development of the third candidate cesium separation technology, STTP, severe foam problems occurred that hindered processing. Earlier work by EMSP research project #60143, which developed a fundamental understanding of the physical mechanisms that produce foaming in the DOE high-level waste and low-activity radioactive waste separations processes, led to the development of a radiation-resistant anti-foam agent for the DWPF. An improved anti-foam agent was developed based upon a better understanding of the chemistry, rheology, and physics that lead to the formation of foam during waste processing. The new anti-foam was successfully tested in laboratory scale experiments at the Illinois Institute of Technology and SRS and in a pilot plant at SRS and has proved to be superior to the current anti-foam agent. Other applications at SRS include Waste Tank evaporation, sludge processing, and salt processing. Other sites with similar evaporation processes, primarily Hanford, ORNL, and INEEL, are all in need of better antifoam agents to improve process throughput.

EMSP Project Number: 65411, 81887, **Project Title:** *Precipitation and Deposition of Aluminum-Containing Phases in Tank Wastes*, **Lead PI:** Dr. Shas Mattigod, *Pacific Northwest National Laboratory*, **Co-PI(s):** Dr. Daniel M. Dabbs, *Princeton University*; Dr. David Hobbs, *Savannah River Technology Center*

The plugging of an evaporator at SRS caused premature shutdown of the DWPF facility in October 1999. To solve this urgent problem, the site mobilized all available resources, including a TFA project. In turn, TFA mobilized its resources including calling on an EMSP researcher working on a related project to study the crystal formation of aluminum compounds in tank sludge. The original EMSP project plan was to focus on the boehmite to gibbsite transformation, but this was quickly changed to study the formation of aluminosilicates as evaporator plugging compounds. The new studies are aimed at the investigation of AlSiO formation. The data gathered is being used by the TFA to develop a thermodynamic model to predict solids formation and establish a safe operating envelope for evaporator composition.

The original EMSP planned work on transformation of boehmite to gibbsite, is also critical to a long-term solution for processing waste with high aluminum content at DWPF (or later Hanford). Historically, tank washing has been able to remove 75% of the aluminum from the sludge, thereby reducing HLW volume and allowing operation within acceptable aluminum concentrations. However, several tanks to be processed contain aluminum in a boehmite form, which is not readily soluble using normal sludge washing conditions. This EMSP task will determine whether the boehmite form can be transformed to the more soluble gibbsite structure and achieve the necessary aluminum removal.

EMSP Project Number: 54621, 73749, **Project Title:** *Chemical Speciation of Strontium, Americium, and Curium in High Level Waste: Predictive Modeling of Phase Partitioning During Tank Processing*, **Lead PI:** Dr. Andrew R. Felmy, Pacific Northwest National Laboratory, **Co-PI:** Dr. Gregory Choppin, Florida State University

EMSP research has yielded new thermodynamic data, Pitzer parameters, to develop a new thermodynamic model to predict equilibrium phase partitioning of radionuclides and other chemicals in high electrolyte solutions. The new Pitzer model allows better prediction of the fate of radionuclides during processing of the many different and complex solutions that are in the DOE wastes. The Pitzer model is also utilized almost exclusively in the scientific community so thermodynamic data from other sources could be readily incorporated into the model.

This information is valuable for plant design and predicting performance of process unit operations. The first application of the science results was seen to better predict the dissolution and precipitation of solids important in tank processing and cross-site transfer line plugging. However, the DOE sites use another software model, the Environmental Simulation Program (ESP), to predict conditions that could cause plugging problems in line transfers. The ESP couldn't use the Pitzer parameter data. Although the Pitzer model better predicted the important phase equilibria, the sites couldn't routinely take advantage of the better thermodynamic model.

To overcome the disconnect and make his EMSP research results more usable, the researcher worked through the TFA and with the ESP software vendor to create a software upgrade that would use the Pitzer based model and could integrate all the Pitzer based thermodynamic data available from the EMSP research and other programs worldwide. Access to such a wealth of available data will significantly improve tank process modeling at Hanford and Savannah River.

EMSP Project Numbers: 55042, 73859, **Project Title:** *Quantify Silica Reactivity in Subsurface Environments: An Integrated Experimental Study of Quartz and Amorphous Silica to Establish a Baseline for Glass Durability*, **Lead PI:** Dr. Patricia M. Dove, Virginia Polytechnic Institute & State University

EMSP Project Number: 60362, **Project Title:** *Ion-Exchange Processes and Mechanisms in Glasses*, **Lead PI:** Dr. B. Peter McGrail, Pacific Northwest National Laboratory, **Co-PI(s):** Dr. David K. Shuh, Lawrence Berkeley National Laboratory

These two EMSP projects have been working closely together to investigate waste glass properties. Project #73859 is investigating the waste glass dissolution kinetics. Project #60362 developed a better understanding of how glass structure impacts sodium ion exchange so that improved waste glasses can be developed. This example demonstrates another benefit of the two-way interchange between TFA programs and EMSP developments. In the previous examples, EMSP researchers were funded directly by TFA to perform additional tasks related to their EMSP work. In this example, the knowledge developed through EMSP projects modified the TFA program for evaluating issues associated with long-term stability of waste glasses and impacting the Hanford performance assessment of its proposed Low Activity Waste (LAW) disposal.

EMSP Project Number: 65371, **Project Title:** *Numerical Modeling of Mixing of Chemically Reacting, Non-Newtonian Slurry for Tank Waste Retrieval*, **Lead PI:** Dr. David A. Yuen, University of Minnesota, **Co-PI:** Dr. Yasuo Onishi, Pacific Northwest National Laboratory

Many highly radioactive tank wastes will be retrieved by installing mixer pumps that inject high-speed jets to stir the sludge, saltcake, and supernatant liquid in the tank, blending them into a slurry. This slurry will then be pumped out of the tank into a waste treatment facility. This EMSP project investigated the chemical reactions, waste rheology, and slurry mixing occurring during the retrieval operation to provide decision-making support tools needed to determine the safety, design, and operational conditions of waste retrieval.

This project has successfully integrated a computational fluid dynamics code with state-of-the-art equilibrium and kinetic chemical models and non-Newtonian rheology. This new transport code, ARIEL, is a state-of-the-art non-Newtonian reactive transport computer code that is applicable to a time-varying, three-dimensional hydrothermal field with multi-phase, multi-component, high ionic-strength, and highly basic chemical conditions. This is unique and is already being applied to the complex phenomena of tank waste retrieval. The ARIEL code explicitly accounts for interactions of aqueous chemical reactions, adsorption/desorption, and dissolution/precipitation under high ionic-strength conditions and associated rheology (viscosity and yield strength) changes.

This modeling capability can be used to determine how many pumps are needed to mobilize the tank waste, and with a given number of pumps, how much and how fast tank waste will be mixed. Developments in this project, including computer code and staff expertise, are being applied directly to Hanford site needs. The ARIEL code was used to model the mixing of waste with two 300-hp mixer pumps in Hanford's Double-Shell Tank 241-AZ-102 and is currently being applied to 241-AN-104 and 241-AN-105 for Hanford's W211 (Tank Waste Retrieval).

EMSP Project Numbers: 65435, 81897, **Project Title:** *Millimeter-Wave Measurements of High Level and Low Activity Glass Melts*, **Lead PI:** Dr. Paul P. Woskov, Massachusetts Institute of Technology, **Co-PI(s):** Dr. William E. Daniel, Savannah River Technology Center, Dr. S Kamakshi Sundaram, Pacific Northwest National Laboratory

This project won the prestigious R&D 100 award as one of the 100 most significant new technologies in 2001. An instrument was developed to perform multiple measurements of high temperature chemistry on solid surfaces that take place during melting such as vitrification of HLW. This is an innovative high temperature viscosity measurement technology for process monitoring of hot molten materials for HLW, as well as numerous industrial applications. The EMSP project targets HLW treatment processes to maximize waste loading in glasses for wastes stored at Hanford, Idaho, and Savannah River. Real-time sensors characterize glass melts containing high-level waste and low activity waste using millimeter-wave sensor technology to simultaneously measure temperature, conductivity, and viscosity. These sensors will decrease

the uncertainty in current property models and allow for improved operation and control of melters. It enables better process control to improve reliability and efficiency of high level waste melters and is applicable to DWPF, WVDP, and vitrification efforts at Hanford, Oak Ridge, and INEEL sites.

The researchers demonstrated the technology's temperature measurement capability during a pilot run of a melter at Clemson University during a TFA/SRS melter trial, and are preparing to demonstrate the technology's viscosity measurement capabilities as part of the renewed EMSP project. By demonstrating their technology on a real melter used for processing development engineering and site personnel have become more comfortable that the technology can be implemented in real plant situations.

EMSP Project Numbers: 55110, 73976, **Project Title:** *Iron Phosphate Glasses: An Alternative for Vitrifying Certain Nuclear Wastes*, **Lead PI:** Dr. Delbert E. Day, University of Missouri-Rolla

Borosilicate glass is the only material currently being used and approved as a host matrix to vitrify HLW. Some HLW contains components that are either insoluble or incompatible with borosilicate glasses; and they dramatically decrease the waste loading capacity of the borosilicate glasses. Pretreatment processes to remove these components are costly. Iron phosphate glasses promise advantages over a borosilicate waste form for certain HLWs and may reduce the vitrified waste volume by as much as 50%, with comparable chemical durability, and lower melting temperatures and shorter melting times (which translates to smaller, less expensive furnaces) (16). Since no single type of glass will ever be totally satisfactory and cost effective for all of the many different nuclear waste feeds now in existence, development of an alternative waste form is clearly justified.

INEEL is leveraging EMSP research performed on iron phosphate glass to develop a waste form that has the potential to reduce the final waste form volume and weight. The EMSP researcher is helping to determine iron phosphate glass compositions that would be compatible with INEEL waste. There is the potential to load the iron phosphate glass with significantly more waste than the borosilicate glass. INEEL is testing several candidate glass compositions in a Russian cold crucible furnace and is proposing to build a similar furnace at the INEEL.

The EMSP researcher's expertise and EMSP data were also called upon to contribute to a DOE-HQ sponsored study to review all the currently available vitrification technologies and waste forms for applicability to DOE wastes. The purpose of the study was to determine if the enormous cost of DOE site clean up could be reduced by employing alternate immobilization technologies or wasteforms (17).

EMSP Project Number: 60424, **Project Title:** *High Temperature Condensed Phase Mass Spectrometric Analysis*, **Lead PI:** Dr. James E. Delmore, Idaho National Engineering and Environmental Laboratory

The project developed an integrated mass spectrometric analysis system capable of analyzing materials from room temperature up to high temperatures, with the practical upper temperature limit to be experimentally determined. A primary objective of the program was to develop

techniques to analyze waste materials during vitrification processing to produce waste forms. As a result of this research, this analytical tool is now being used at the INEEL to support subsurface science research. The tool is capable of performing mass spectral analysis on solid surfaces. It performs multiple analyses in rapid succession versus the current tools, which utilize single analytical techniques.

SUMMARY

These examples show that fundamental scientists involved in EMSP projects have already made, and will continue to make, important contributions to the HLW program. These cases also show that the usefulness of EMSP research is maximized when there is close integration with the decision-making and technology deployment arms of the DOE EM program, TFA, and site users. Sometimes the technology deployment arms (TFA or DOE sites) sought out EMSP researchers because their work was known and their relevant expertise was needed. In other cases, such as the CSSX technology or Pitzer model, the EMSP researchers translated their results in a readily understandable form and made extra efforts to make their scientific results usable.

Several EMSP projects were advanced using TFA funding to perform additional tasks. Applied research and technology development funding are critical to the ability to deploy some of the cost-saving, scientific breakthroughs.

The value of pursuing alternatives to current baselines was clearly demonstrated in the SPP. It takes years for basic research to advance to the stage where it can be useful in operations. It is important that DOE invests to develop viable alternatives in case baseline technologies fail, such as cesium separation at Savannah River, or for improved performance and cost reduction during the years of operation to complete the HLW clean up.

The standard products from basic research programs are reports and peer reviewed papers and journal articles. These are important to maintain the scientific credibility of the research and data upon which decisions are made. It is interesting to note, however, that actual use of EMSP results hasn't occurred through such an academic mechanism. That is, a DOE problem holder hasn't read an EMSP report or journal article and found a solution for his problem. Rather, real use of EMSP results and success addressing DOE problems has occurred when EMSP researchers and DOE technology deployment arms have worked together to address the multitude of details that are necessary to consider and could never be completely captured in a broad call for proposals. It is through these skilled scientists making timely connections with DOE problem holders, people connecting with people, that tangible contributions have been realized from EMSP.

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FOOTNOTES

¹ In this paper, high-level waste (HLW) refers to the highly radioactive by-product generated by the chemical reprocessing of spent fuel and target materials to recover plutonium and uranium for weapons, research, and new fuel (1).

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