The U.S. Department of Energy-Office of Environmental Management’s International Program

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ABSTRACT

The U.S. Department of Energy’s (DOE) Office of Environmental Management (EM) has collaborated with the various international institutes (e.g. Russian Federal Atomic Energy Agency – Rosatom and Ukraine government’s Chornobyl Center-International Radioecology Laboratory (CC-IRL)) for many years on radioactive waste management challenges of mutual concern. Currently, EM is cooperating with Rosatom and the Ukraine’s CC-IRL to explore issues related to high-level waste and to investigate experience and technologies that could support EM site cleanup needs. EM is currently implementing five collaborative projects with other international institutes on nuclear waste issues:

- Application of the Cold Crucible Induction Heated Melter to DOE Wastes; SIA Radon, Moscow.
- Design Improvements to the Cold Crucible Induction Heated Melter; Electrotechnical University, St. Petersburg.
- Improve Retention of Radionuclides in Cement Type Waste Forms; Khlopin Radium Institute, St. Petersburg.

- Improved Solubility and Retention of Troublesome Components in SRS and Hanford HLW Glasses; Khlopin Radium Institute, St. Petersburg.
- Long-term Impacts from Radiation / Contamination within the Chornobyl Exclusion Zone, Chornobyl Center, International Radioecology Laboratory, Ukraine

The purpose of this paper is to provide an overview of EM's objectives for participating in cooperative activities with various international institutes. Additionally, this paper presents programmatic and technical information on these activities, and outlines specific technical collaborations currently underway and planned to support DOE's cleanup and closure mission.

INTRODUCTION

The Office of Environmental Management (EM) and EM's Office of Engineering & Technology, U.S. Department of Energy are responsible for implementing EM's international cooperative programs. The Office of Engineering & Technology's international efforts are aimed at supporting EM's mission of accelerated risk reduction and cleanup of the environmental legacy of the nation's nuclear weapons program and government-sponsored nuclear energy research. To do this, EM pursues collaborations with government organizations, educational institutions, and private industry to identify technologies that can address the site cleanup needs of the U.S. Department of Energy. The Office of Engineering & Technology currently works with the Russian Federation and Ukraine CC-IRL through cooperative bilateral arrangements to support EM's accelerated cleanup and closure mission. The Office of Engineering & Technology is also currently evaluating the potential benefits to the U.S. seeking collaborative agreements and contracts with governments and research institutes in other parts of the world (e.g. Asia).

Through international arrangements, EM engages in the cooperative exchange of information, technology, and data on technology development. EM works closely with foreign governments, industry and universities to acquire innovative environmental technologies, scientific and engineering expertise, and operational experience that support DOE's objectives. DOE-EM will also seek out additional international institutes possessing technical expertise to provide assistance to DOE in addressing the highest priority technology needs for cleaning up our sites across the DOE complex

The following discussion provides an updated overview from that presented last year at Waste Management [1]. The latest results from the current bilateral collaboration projects being implemented by EM in support of the EM accelerated cleanup and closure mission are presented. A new task related to the Chernobyl Exclusion Zone is introduced and involves research performed by the International Radioecology Laboratory in the Ukraine. Each of the five tasks listed in the Abstract will now be discussed in detail.

APPLICATION OF THE COLD CRUCIBLE INDUCTION HEATED MELTER TO DOE WASTES

Background and Application

The U.S. technical lead for this task is Dr. James Marra of Savannah River National Laboratory (SRNL). DOE's partner on this project is the Radon Institute with Dr. Sergei Stefanovsky the technical lead on the Russian side. The advanced melter evaluations requested for testing during the contract period are focused on taking the results of prior testing in Russia (at both the SIA Radon and Khlopin Radium Institute facilities) and then applying what we have learned to the processing of difficult Savannah River waste compositions. Once the improvements in melt rate and waste loading have been demonstrated in Russia, then a retrofit of the technology into the U.S. DOE Defense Waste Processing Facility (DWPF) can be evaluated. The accelerated mission objectives continue to emphasize maximizing waste throughput (i.e., to increase waste loading to minimize the number of canisters produced and the melt rate to accelerate project completion). The CCIM appears to be a very robust technology and some of the existing designs appear compatible with the DWPF. Therefore, a thorough evaluation of the existing design and technology should be performed.

Scope of Work and Results

In previous testing, the Radon Institute conducted three (3) 216 mm melter tests and two (2) longer duration tests in the 418 mm CCIM (See Figure 1) using a DWPF sludge batch 2 (SB2) waste composition and Frit 320 glass composition. The testing was conducted jointly by SRNL and Radon conducted at the Radon Institute's laboratory. The previous testing at the Radon facility demonstrated that waste loadings up to 50 wt % (on an oxide basis) for a surrogate DWPF sludge batch 2 (SB 2) feed could be readily processed in the CCIM [2]. It should be noted that DWPF achieved an approximate 38 wt % maximum loading with the SB2 feed. The resulting glass products were of good quality. The results are presented in detail in reference 2.

The objective of testing in FY05 was to determine the maximum waste loading achievable with the SB2 waste composition and the Frit 320 glass with a high end target of 60 wt % waste loading. The test was initiated using 50 wt % waste loading glass "cullet" prepared in previous testing. Slurry feed representing a 60 wt % waste loading was added to the melter for a cumulative period of approximately 52 hours. This strategy resulted in a sequential increase in waste loading in the melter until 60 wt % waste loading was achieved (i.e. after the initial 50 wt % glass cullet was completely "flushed" out of the melter, 60 wt % waste loading was achieved). As expected, an increase in melt viscosity was observed as waste loading increased. The glass was readily processed through the melter up to the point that 60 wt % waste loading was reached. At this point, the melter drain tube had to be manually cleared to facilitate glass pouring. Glass samples from the testing were characterized to determine chemical composition, identification of crystalline phases, and determination of durability. In general, the glass compositions were very near the target values and the glasses exhibited good durability. At waste loadings equal to or greater than about 55 wt % there was an apparent decrease in durability. This is likely due to destruction of the glass network and formation of a nepheline phase in the glass. It should be noted, however, that the glass durability for all samples as measured by the Product Consistency Test (PCT) was better than the reference Environmental Assessment (EA) glass used for repository acceptance.

From Left to Right: 1) **Cold Crucible Melter at SIA Radon** Before Insulation 2) After Insulation with Induction Heating Coils

From Left to Right: 1) Inside Melter Before Startup 2) Inside Melter After Startup

Figure 1 – Increasing Nuclear Waste Loading in DOE High Level Waste Glass Using a Cold Crucible Induction Melter

Significant FY2006 Accomplishments:

- Vitrification tests on a SRS defense waste SB2 surrogate using the 418 mm inner diameter CCIM have demonstrated the high efficiency of the CCIM technology and processing of these feeds up to 60 wt % waste loading.

- Normalized releases of B, Li, Na, and Si from vitrified products with waste loading up to 60 wt % were determined using the ASTM PCT-A procedure and were found to be at least 5 times lower than those from the EA glass used for waste form repository acceptance

Path Forward

Future testing is planned to evaluate processing of other DWPF feeds in the CCIM with specific evaluation of processing the Sludge Batch 4 composition. This waste feed is high in aluminum which limits waste loading due to nepheline formation and a corresponding reduction in durability. The higher process temperatures afforded by the CCIM may alleviate some of the processing difficulties associated with high aluminum feeds and lead to a durable product at higher waste loadings. Glass formulation testing is currently underway with a goal to perform melter testing in the summer of 2007.

DESIGN IMPROVEMENTS TO THE COLD CRUCIBLE INDUCTION HEATED MELTER

Background and Application

The U.S. technical leads for this task are John Richardson and Jay Roach of the Idaho National Laboratory (INL). DOE's partner in this effort is the Electrotechnical University and the Russian technical lead is Dr. Dmitry Lopukh. The INL and Electrotechnical University (ETU LETI), St. Petersburg, Russia, have collaboratively developed two sophisticated mathematical models of cold crucible induction melter (CCIM) operation using both finite-element and bulk-properties approaches. The models will help to design more efficient melters, and to develop a more complete understanding of CCIM theory and how to effectively control this type of melter. This work was only partially validated in 2003 using the INL CCIM, and further validation was accomplished jointly with ETU LETI faculty throughout 2004 and 2005, as a natural extension of the work. Further, if proven effective, this work would result in an implementable and novel control technology that could greatly enhance CCIM operation in both countries.

Scope of Work and Results

During FY2006, the INL researchers continued their collaboration with ETU LETI to design and validate sensors and methods to characterize and control the operation of a CCIM using feedback signals from various operating parameters within the power supply system. Several accomplishments were achieved, including 1) development and testing of enhanced unique sensors that can determine the power factor in a high frequency generator, which is a key parameter in the proposed control scheme (see Figure 2), 2) collecting more comprehensive temperature distribution data in the melt volume during processing, and, most significantly, 3) validating the ability to measure key signals resulting from specific operational parameters in the generator that are driven by the melt properties, which are not directly measurable, and thus determine the operational regime (i.e. temperature profile within the melt) through correlation.

Figure 2 - Configuration and location of phase angle (current component), voltage, and current sensors, respectively, on the ETU LETI Test Melter

In collaboration with INL, ETU LETI designed, manufactured, and installed instrumentation in a CCIM system to measure key parameters (voltage, current density and phase angle) necessary for INL to validate the CCIM electro-thermal models and measurement scheme. The Khlopin Radium Institute also supported this effort through development of models to predict the magnetic flux on the inductor coil, which was used to help develop the sensor operational parameters (See Figure 3). INL developed the bulk properties model and provided technical oversight during key data gathering tests. INL also provided technical consultation during test data analysis and assisted with report writing for the project.

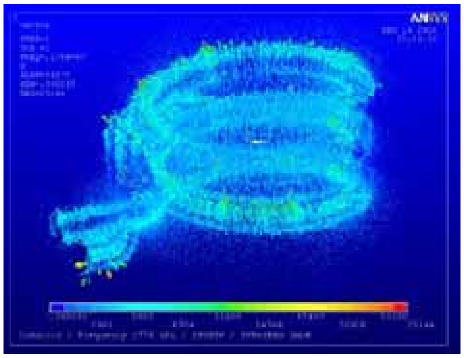

Figure 3 - Model Prediction of the Magnetic Flux on the Inductor at Khlopin Radium Institute.

Figure 4 - Deployment of High Temperature Thermocouples in the Test Melter at ETU LETI

Significant FY2006 Accomplishments

- Continued to enhance/refine the unique sensors designed to collect operational signals in the high frequency environment, which can be used as a feed back control system. Focus was on clarity of signal, repeatability, and validation.

- Designed and constructed a complex, highly instrumented cooling water distribution system for conducting calorimetry measurements for comparative evaluation of the sensor measurement results to determine accuracy and repeatability.

- Developed a more comprehensive temperature profile within the melt for correlation with the electrical signals (phase angle, grid, anode current and voltages) using specially-designed platinum-platinum/rhodium thermocouples.

Path Forward

This task will continue to utilize the investment in unique testing equipment with enhancements to allow testing at higher temperatures, as necessary. The key parameters to be measured include extending the temperature profile to more comprehensive, and potentially higher temperatures; as well as further characterization of the power factor measurement and the use of calorimetry for comparative validation. Several activities are proposed that will allow implementation of an automated feedback control system for CCIM operation.

- The signal transformation model should continue to be developed. Appropriate measurement windows and sample frequency need to be determined to quantify and control the accuracy of the results. The capability to directly input the raw data from the ETU LETI sensors into the software for analysis and interpretation also needs to be added.
- A comprehensive series of experiments need to be conducted to validate the application range, operability, maintainability, and repeatable of the sensors.
- Further understanding of the melt height and its effects on the temperature distribution needs to be developed and quantified for optimizing designs and their operation.
- An integrated melter system needs to be constructed that has automated control such that the feedback system can be demonstrated and its accuracy quantified. This should eventually include the ability to also automate frequency adjustment for optimizing energy deposition as feed compositions and desired operating parameters change.

IMPROVED RETENTION OF KEY CONTAMINANTS OF CONCERN IN LOW TEMPERATURE IMMOBILIZED WASTE FORMS

Background and Application

The technical leads are Dr. Albert Aloy of the Khlopin Radium Institute and Dr. John Harbour of the SRNL. Russian researchers have significant experience in grout waste forms and in measuring the leach rates of radioactive and hazardous (RCRA) metals. In addition, they have the knowledge base to identify existing reagents and technical capabilities to develop new technologies for immobilization of contaminants of concern (COC). Innovative approaches to immobilizing COC is crucial to the expanded use of grout and/or other non-thermal waste forms to treat radioactive/hazardous wastes and to dispose of the resulting waste forms in shallow landfills in the DOE complex [3]. The project objectives in 2006 were to develop and test improved ambient temperature waste forms that provide for stabilization of (> 10,000 yr.) Tc-99 and Se-79. An additional objective was to measure the heat of hydration of Saltstone formulations and determine whether alternative cementitious components can be used to reduce the heat of hydration. The increase in temperature due to heat of hydration of the waste form in the Saltstone vaults can limit the processing rate of the facility.

Scope of Work and Results

Tc-99 A salt solution (doped with Tc-99), that simulates the salt waste stream to be processed at the Saltstone Production Facility, was immobilized in grout waste forms with and without (1) ground granulated blast furnace slag and (2) pretreatment with iron salts. The degree of immobilization of Tc-99 was measured through monolithic and crushed grout leaching tests. Although Fe (+2) was shown to be effective in reducing Tc-99 to the +4 state, the strong reducing nature of the blast furnace slag present in the grout formulation dominated the reduction of Tc-99 in the cured grouts. An effective diffusion coefficient of 4.75×10^{-12} (Leach Index of 11.4) was measured using the ANSI/ANS-16.1 protocol. The leaching results show that, even in the presence of a concentrated salt solution, blast furnace slag can effectively reduce pertechnetate to the immobile +4 oxidation state. The measured diffusivity was introduced into a flow and transport model (PORFLOW) to calculate the release of Tc-99 from a Saltstone Vault as a function of hydraulic conductivity of the matrix.

Heat of Hydration The objective of this task is to develop Saltstone mixes with an overall heat of hydration that is reduced relative to current design mixes. Specifically, the objective is to achieve a reduction in heat generation through partial or complete substitution of inert materials (or materials having reduced hydraulic or pozzolanic activities) for the fly ash and/or the blast furnace slag. The resultant mixes with lower heat of hydration must also have fresh and cured grout properties that meet the Saltstone specifications. The initial phase of this task was to set up the isothermal calorimeter and measure the heat of hydration of a Saltstone mix for comparison with a previous measurement of the heat of hydration using adiabatic calorimetry. The results are shown in Figure 5.

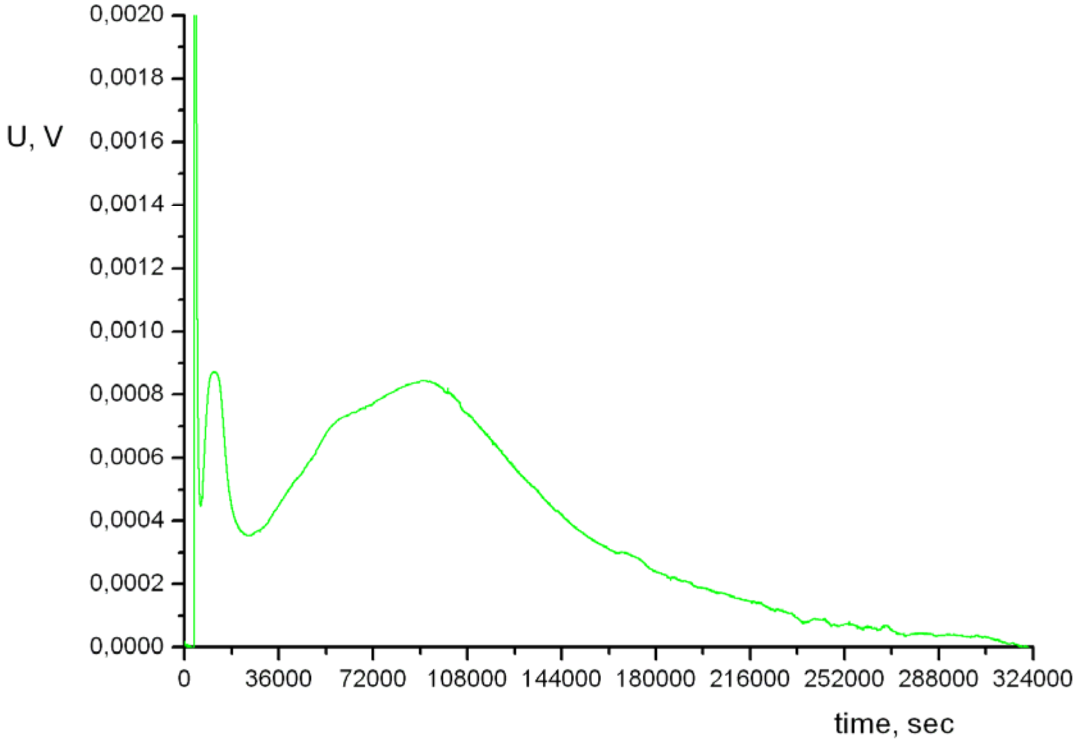

Figure 5 - Isothermal Calorimetry Data for a Saltstone Grout Mixture (Water/Premix=0.6). The units of the ordinate are volts.

There was good agreement between the isothermal measurements of heat of hydration performed in this task with previous measurements of heat of hydration of Saltstone obtained adiabatically over the same 80 hour time period. For the isothermal data, a value of 81 Kj/kg of cementitious material was obtained.

Se-79 The objective of this task was to determine the degree of sorption (i.e., retention) of soluble selenium ions (originally present in the salt waste stream) within the cured Saltstone. The approach will measure the leaching of selenium from both monolithic and crushed Saltstone samples. In addition a determination of the role of reductant in the Saltstone (present in slag) in this sorption process (reducing Se^{+6} to Se^{+4}) will be made.

Initial leaching tests using high levels of selenium demonstrate that essentially all of the selenium is released during leaching. Therefore, at high levels of selenium, the Saltstone does not retain much of the selenium. The next step in the process will be to measure the release through leaching of selenium at the lower levels of selenium that are more representative of levels anticipated for actual waste streams.

Significant FY 2006 Accomplishments:

- The ANSI/ANS-16.1 protocol was used for a monolith leaching test using a formulation containing blast furnace slag obtained from the same vendor that supplies Saltstone Production Facility. The average Tc-99 effective diffusion coefficient, D, and leaching index, L, determined by this method are Deff of 4.8 x 10^{-12} cm^2/sec and L equal to 11.4. This data was used to support Performance Assessment Modeling.

- The experimental setup for measuring heat of hydration for Saltstone mixes has been accomplished and the measurement of the heat of hydration for the Saltstone formulation using a water to premix ratio of 0.60 corresponded well with previous measurements using adiabatic calorimetry.

- Leaching tests were completed with Saltstone formulations containing high levels of selenium. These results indicate that at these levels, most of the selenium is released.

Path Forward

Future work will focus on the substitution of materials for blast furnace slag and Class F fly ash in the Saltstone formulation that will reduce the heat of hydration and the consequent temperature rise of the waste form in the Saltstone Vaults. Other important properties of the grout will be measured to ensure that all grout properties still meet the Saltstone requirements. Lower levels of selenium will be incorporated into the waste stream and grouted. Leach testing of these waste forms will be performed to determine the retention of selenium in the waste form as a function of oxidation state of the selenium.

IMPROVED SOLUBILITY AND RETENTION OF TROUBLESOME COMPONENTS IN SRS AND HANFORD HLW GLASSES

Background and Application

The Russian technical lead is Dr. Albert Aloy of the Khlopin Radium Institute and the U.S. technical leads are Dr. John Vienna (Pacific Northwest National Laboratory) and Dr. David Peeler (Savannah River National Laboratory). The U.S. Department of Energy (DOE) is currently processing (or planning to process) high-level waste (HLW) through Joule-heated melters at the Savannah River Site (SRS) and Hanford. The process combines the HLW sludge with a prefritted additive or with mined mineral glass forming additives. The combination is subsequently melted, and the molten glass is poured into stainless steel canisters to create the final waste form. In preparation for the qualification and receipt of each sludge batch, development and definition of various tank blending and/or washing strategies are often evaluated. The various strategies are contemplated in an effort to meet critical site objectives or constraints such as those associated with tank volume space, transfer options, and settling issues. Although these objectives or constraints are critical, one must not lose sight of both process and product performance issues associated with the final waste form. The product performance issue relates to the durability of the glass waste form. Process related issues (e.g., liquidus temperature, viscosity, electrical conductivity, and melting rate considerations) ultimately dictate the efficiency and effectiveness of the melter operation.

Tank retrieval and blending strategies at both SRS and Hanford have identified high Al_2O_3 waste streams that are scheduled to be processed through their respective HLW vitrification facilities. For example, the Liquid Waste Organization (LWO) at SRS has provided compositional projections for the next two sludge batches (Sludge Batch 4 and Sludge Batch 5) to be processed in the Defense Waste Processing Facility (DWPF). These streams have Al_2O_3 concentrations between approximately 25 and 40 wt% (on a calcined oxide basis). In addition, physical limitations in the Tank Farms and/or settling issues associated with the sludge have prevented “advanced” washing which has resulted in relatively high Na_2O (between ~ 22 – 26 wt%) and SO_3 (between ~ 0.8 – 1.6 wt%) concentrations. Current Hanford projections suggest the Al_2O_3 concentrations in sludge could be much greater than those currently projected for DWPF, with Al_2O_3 concentrations as high as 80 wt%.

Under this task, researchers from the Pacific Northwest National Laboratory, Savannah River National Laboratory, and Khlopin Radium Institute are collaborating to conduct experiments that will develop glass formulations for specific DOE waste streams to maximize Al_2O_3 concentration in glass while maintaining or meeting waste loading and/or waste throughput expectations as well as satisfying critical process and product performance related constraints. Secondary objectives of this task are to assess the SO_3 solubility limit for the DWPF composition and spinel settling for the Hanford composition.

Scope of Work and Results

To meet the stated objectives, an integrated work scope has been developed in four primary areas (see Figure 6):

1. Test matrix development
2. Laboratory scale glass fabrication and testing
3. Melter testing
4. Documentation

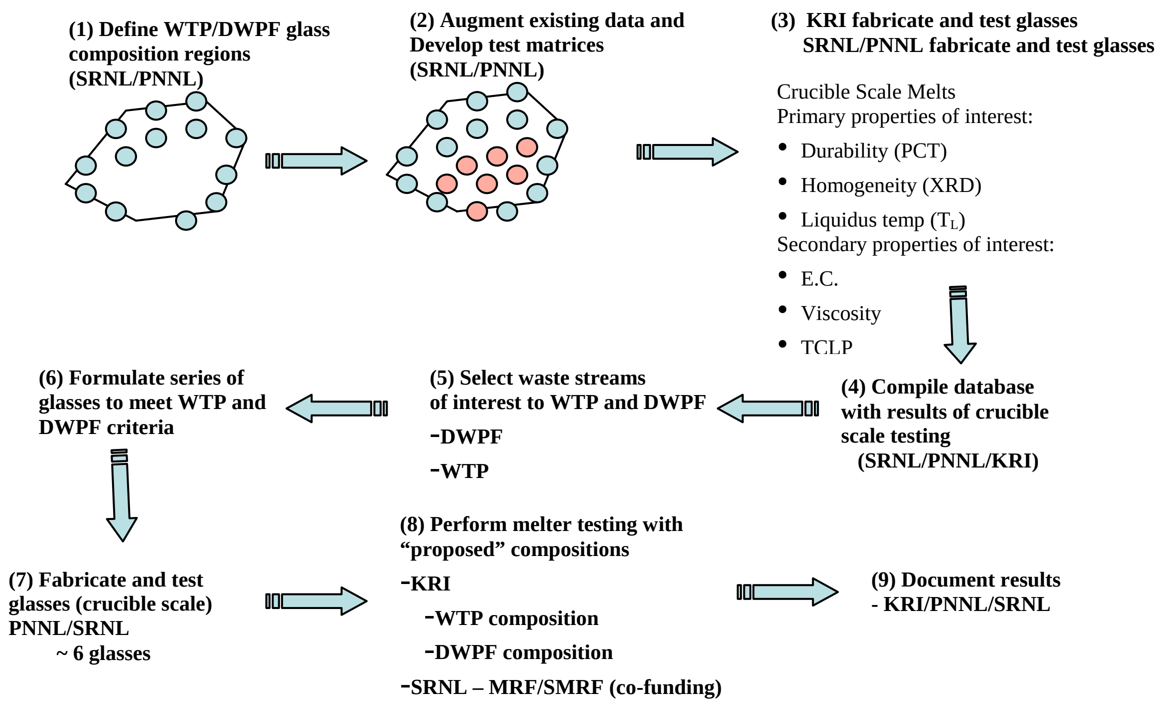

Figure 6 – Integrated Work scope for HLW Glass Solubility Testing

The International Team has developed an integrated test matrix of 75 glasses which will provide valuable compositional – property relationship data from which optimization studies can be based. The 75 glasses were statistically designed to cover glass composition regions of interest to both DWPF and Hanford. The International Team has completed fabrication of the glasses and are currently testing specific properties of interest including durability, liquidus temperature (see Figures 7 and 8), homogeneity and viscosity.

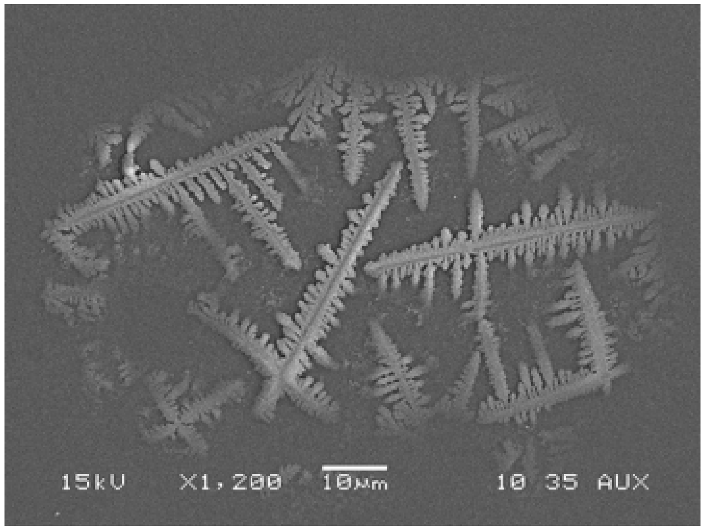

Figure 7 - Example of Nepheline Formation.

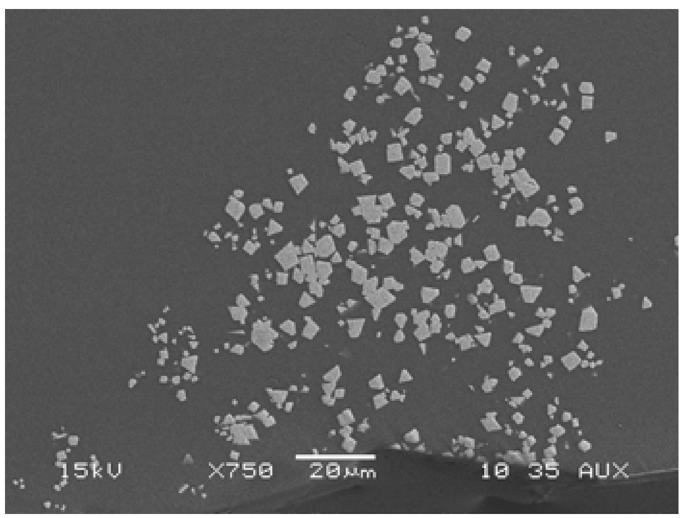

Figure 8 - Example of Spinel Formation.

Path Forward

Once data compilation on the 75 glasses is complete, specific high Al_2O_3-based waste streams of interest to DWPF and Hanford will be identified. The International team will develop specific glass compositions to support the KRI EP-5 and/or SMK melter runs. Development of the specific glasses will be based on the results of the laboratory test matrix glasses and will leverage any existing data that may support the development process. Existing and new composition-property models will be used to support glass formulation efforts. The models will be a guide with respect to the acceptability of the specific glass composition relative to the acceptance criteria for Hanford and/or DWPF. Once the specific DWPF and Hanford glasses have tested in the laboratory to confirm model predictions, KRI will process the selected compositions through their EP-5 melter (see Figure 9). The KRI EP-5 melter is a rectangular Joule-heated melter that will be used for evaluation of the selected high-Al_2O_3 simulated HLW glass compositions.

Significant FY 2006 Accomplishments:

- Developed integrated test matrix covering compositional regions of interest to both DWPF and Hanford
- Completed fabrication of 70 test matrix glasses
- Initiated development of composition – property relationship database
- Completed durability testing of 45 test matrix glasses

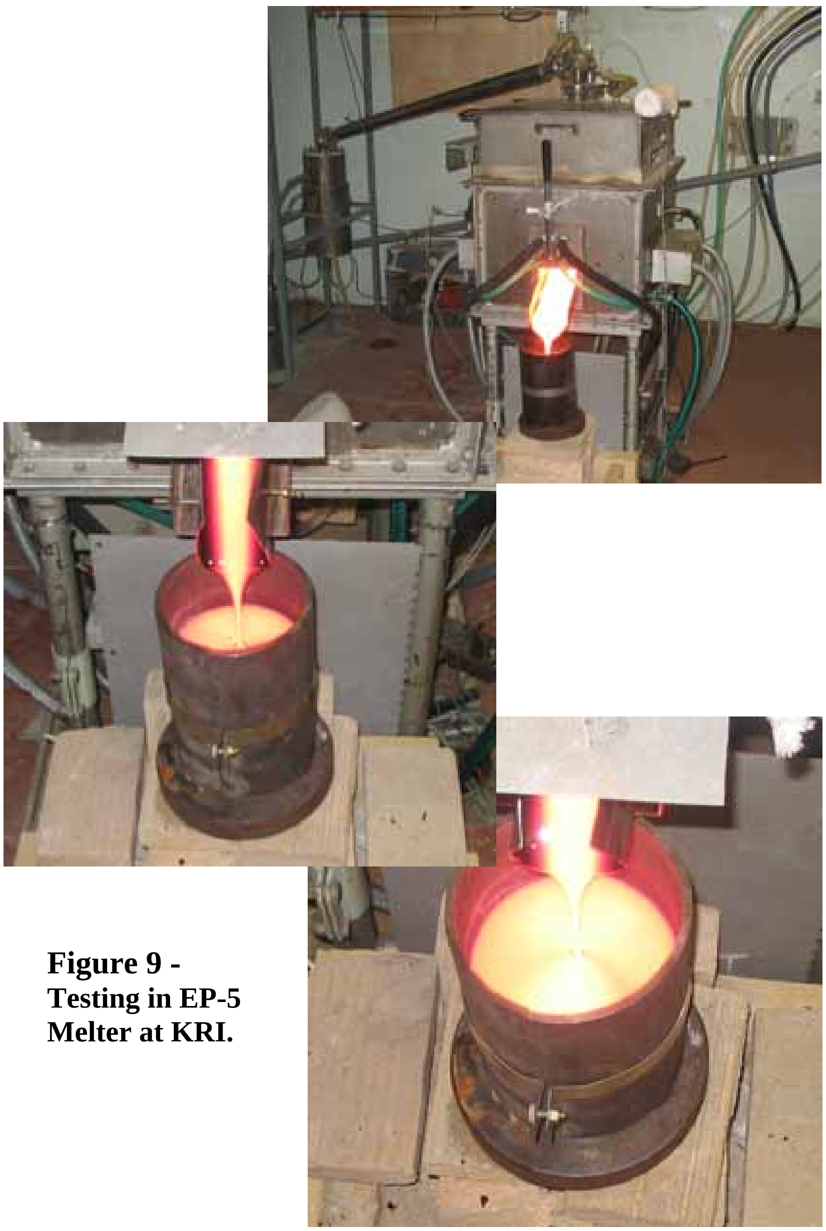

Figure 9 - Testing in EP-5 Melter at KRI.

INTERNATIONAL RADIOECOLOGY LABORATORY – CHERNOBYL

Background & Application

The Russian technical lead is Dr. Mikhail Bondarkov of the International Radioecology Laboratory (IRL) in Slavutych, Ukraine and the U.S. technical leads are Dr. John Harbour and Bond Calloway of the Savannah River National Laboratory.

The overall objectives of the project are:

- Assess the long-term impacts to the environment from radiation exposure within the Chornobyl Exclusion Zone.
- Provide information on remediation guidelines and ecological risk assessment within radioactively contaminated territories based on the results of long-term field monitoring, analytical measurements, and numerical modeling of soils and groundwater radioactive contamination.
- Recommend the development and testing of effective cleanup technologies to reduce environmental and health risks.

Scope of Work and Results

There are four subtasks that comprise the overall task:

Task 1 Characterization and Monitoring Methods

Goals*:* Evaluate the performance of existing characterization and monitoring methods that are deployed at various field sites within the Chornobyl Exclusion Zone (ChEZ). The results of this task will be used to provide recommendations related to long-term radio-ecological monitoring of contaminated lands and sites which are designated for recovery.

Task 2 Understanding of the Processes and Modeling of Fate and Transport of Radionuclides through Environmental Pathways (e.g., soils, groundwater, surface waters, microbiota, plants and animals etc.)

Goals: Perform data analysis to obtain a better understanding of radionuclide fate and transport processes in the ChEZ, and demonstrate the applications of computer databases and geographic information systems. The results of this task will be used to build confidence in conceptual, mathematical and numerical models of flow and radionuclide transport between compartments of ecological systems (soil, ground and surface water, microbiota, plants, and animals.

Task 3 Evaluation of Risk Assessment Methods

Goals: Evaluate and document the use of risk assessment methods within the ChEZ. The results of this task will be used to build confidence in conceptual, mathematical and numerical models to be used for the risk (dose) assessment in different parts of the environment and population groups.

Task 4 Evaluation and Demonstration of Cleanup Technologies for Radioactively-Contaminated Sites

Goals: Evaluate the areas of potential application of monitoring methods, modeling techniques, and cleanup technologies used in the ChEZ at radioactively contaminated DOE sites. The results of this task will be documented in a series of joint DOE-IRL reports and peer reviewed publications, which will be distributed among DOE scientists, engineers, managers, and contractors.

Significant FY 2006 Accomplishments:

- The test plans that lay out the specific research activities for each of the 4 tasks identified above have been written, reviewed and approved.

Path Forward

A series of deliverables has been developed and agreed upon that will summarize the research results from each of the four tasks. Efforts in all four tasks were initiated in September, 2006.

CONCLUSION

DOE-EM has shown again that its current program of cooperation with other international institutes will allow EM to leverage international expertise in support of an accelerated risk reduction and cleanup of DOE sites through the identification of innovative technologies focused on user needs. It is EM's goal to achieve a program with international centers of excellence that is solution- and cost savings-oriented. EM has shown that the five projects discussed in this paper have extensive application to current DOE high-level waste challenges and will contribute greatly to this goal. EM also looks forward to the possibility of enhancing the scope of the EM-Rosatom cooperation program to investigate other areas of mutual concern.

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