# THE U.S. DEPARTMENT OF ENERGY–OFFICE OF ENVIRONMENTAL MANAGEMENT TECHNICAL PROJECTS WITH THE RUSSIAN FEDERAL ATOMIC ENERGY AGENCY (ROSATOM)

K.D. Gerdes
Office of Environmental Cleanup and Closure
Office of Environmental Management
U.S. Department of Energy

## ABSTRACT

This paper outlines specific technical collaborations between the U.S. Department of Energy's (DOE) Office of Environmental Management (EM) and the Russian Federal Atomic Energy Agency (Rosatom) currently underway to support DOE's cleanup and closure mission, specifically in the area of high-level waste.

## INTRODUCTION

Representatives from DOE's Office of Environmental Management (EM), DOE national laboratories, Florida State University, Rosatom and Russian institutes conducted meetings in March and April 2004 to reinitiate EM-Rosatom cooperation, which had been stalled as the result of the expiration of a Government-to-Government agreement. Through preliminary discussions between DOE and Rosatom, one of the most prevalent environmental challenges, High-Level Waste (HLW), was agreed upon as the focus of the initial projects. The March 2004 meetings were conducted in Moscow and St. Petersburg, Russia, to review technical details of 6 new technical projects. Technical scopes for 6 projects were agreed upon, in principle, with all of the Russian institutes.

A meeting was held in Moscow in April 2004 with Rosatom to finalize the path forward for the new framework of cooperation and formalize DOE and Rosatom approval of the initial 6 projects. A Record of Meeting was signed that approved the EM path forward and the 6 initial projects, which are listed below:

- Advanced Melter Technology Application to the Defense Waste Processing Facility (DWPF) –Cold Crucible Induction Heated Melter (CCIM)
- Design Improvements to the Cold Crucible Induction Heated Melter
- Long-term Performance of Hanford Low Activity Glasses in Burial Environments
- Improved Retention of Key Contaminants of Concern in Low Temperature Immobilized Waste Forms
- Documentation of Mixing and Retrieval Experience at Zheleznogorsk
- Low-Activity-Waste (LAW) Glass Sulfur Tolerance

All 6 contracts have been finalized and approved by DOE and Rosatom and work is ongoing. This paper presents technical details on 5 of the 6 ongoing projects in the area of high-level waste.

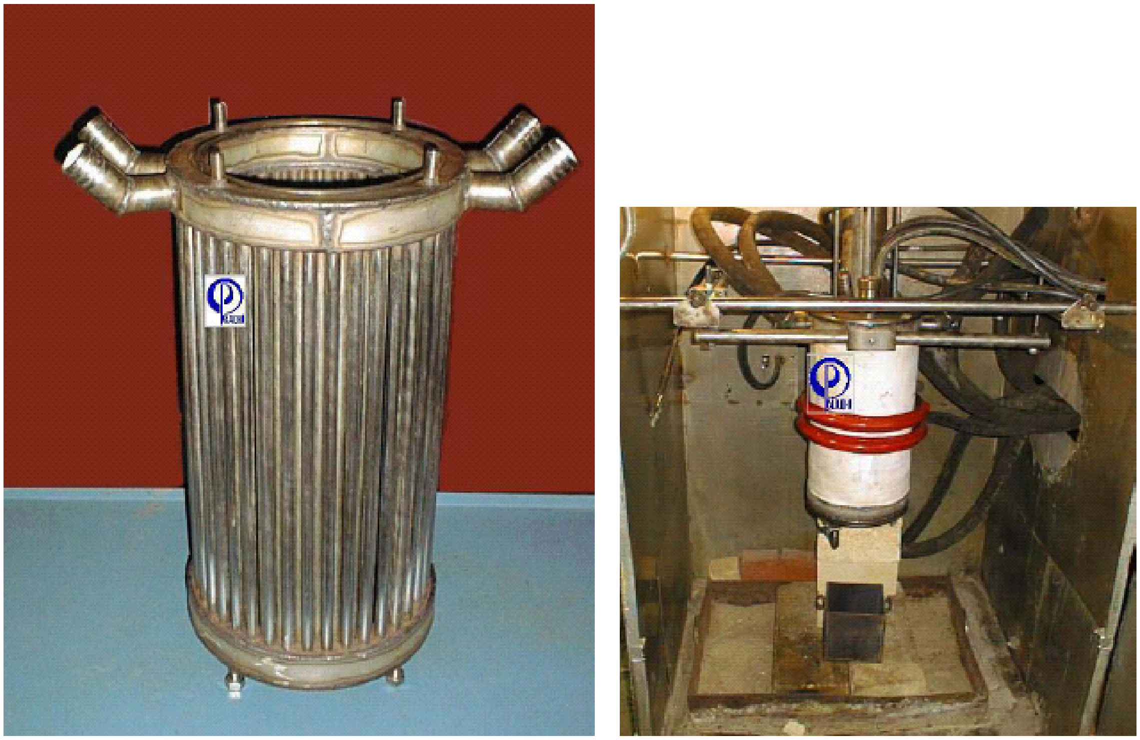
**Task 1 - Advanced Melter Technology Application to the U.S. DOE Defense Waste Processing Facility (DWPF) – Cold Crucible Induction Heated Melter (CCIM)**

**Period of Performance:** May 1, 2004 – October 31, 2005
**Russian Partner:** Radon Institute, Moscow, Russia (Principal Investigator. - Sergei Stefanovsky)
**U.S. Technical Lead:** Savannah River National Laboratory (Principal Investigator **-** James C. Marra)

**Background & Application**
The advanced melter testing initiated in October 2004 and planned to continue through October 2005 will utilize the results of prior testing in Russia (at both the SIA Radon and Khlopin Radium Institute facilities) and then apply what we have learned to the processing of difficult Savannah River Site waste compositions. The issue at the Savannah River Site is that as waste loading is increased, the melt rate in the 1150°C Joule-heated melter begins to decrease; ultimately reducing throughput. The benefit of putting more waste in a single can and reducing the number of canisters going to the HLW repository is still achieved, but the operating time and costs increase.

This program will demonstrate the extent of improvement in waste loading and melt rate that can be achieved with Savannah River Site waste. 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). Further increases in waste throughput and waste loading will exceed the capability of the 1150°C melter in the next several years. The stirred 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.

**Russian Scope of Work**
- Prepare chemical simulants from waste compositions provided by SRNL
- Review and adjust formulation for glasses for use in the CCIM testing
- Prepare a test plan for testing the simulated SRS waste and Radon glass formulations in the 216 mm and 400 mm CCIM
- Perform melter tests according to the plan developed: four (4) 216 mm melter tests and one (1) longer duration tests in the 400 mm CCIM.



(a) (b)

**Fig. 1. CCIM Construction Details and Deployed Configuration-Water-Cooled Shell with Induction Heating to the Melt.**

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**U.S. Scope of Work**

- Develop projections of waste compositions that will bound waste streams that are projected to be difficult to process
- Provide initial glass formulations with increased waste loading that are compatible with the DWPF process flowsheets and equipment
- Define test matrix requirements, project deliverables, and support Radon’s final test plan; ensure test matrix allows for demonstration of feasibility, advantages, and disadvantages of CCIM versus joule heated melter
- Provide expert technical oversight and participation in the CCIM tests in Russia and direct the approach and tools used in interpreting data, test results and deliverables. This role ensures that the final result is compatible with the existing DWPF.
- Provide technical consultation during test data analysis and report writing

**Expected Benefits**

- Approximately a ten per cent improvement in waste loading over current operations (life cycle cost improvement measured in the hundreds of millions of dollars- 1% is approximately $150M)
- A more robust technology. Current technology is limited to 1150°C; the Cold Crucible Induction Heated Technology is capable of much higher temperatures, but will likely operate between 1200°C and 1300°C to limit volatility of cesium and other semi-volatiles.
- The melter is smaller than the current melter with reduced hold up of HLW glass. This will facilitate dismantlement and decommissioning of the melter when operation is complete.

**Status**

- October 29, 2004 completed the initial two 216mm melter tests. Conducted 96 hours of melter operation with DWPF surrogate sludge and frit 200 and frit 320 glass compositions. Glass former additives were used instead of frit in these tests.
- Achieved waste loadings of 45 wt% with both glass compositions. Radon personnel indicated that melter operations were consistent with previous melter operation experience.
- A strategy to transition from frit 200 to frit 320 was employed which provided data for intermediate glass compositions between frit 200 and frit 320.
- The resulting glass products were generally homogeneous with only a small amount of surface crystallization noted (identified as spinels by x-ray diffraction).
- Glass sample analysis was completed including: chemical analysis, physical analysis and durability testing. The results showed that the analyzed glasses were very similar to the targeted compositions indicating very little volatility occurred. The glasses were also very durable as measured by the Product Consistency Test (PCT) with elemental releases for key elements approximately an order of magnitude lower than the Environmental Assessment (EA) repository qualification reference glass. Additionally, off-gas particulate and gaseous species collected during the testing are being analyzed.
- Agreement was obtained on a path forward for follow-on testing:
    - Perform two 216 mm CCIM test with actual frit and simulant sludge to further quantify melt rate and compare to DWPF operations.
    - Perform a large-scale, extended duration test in the 400 mm CCIM with the preferred composition using glass frit and surrogate sludge.

**Task 3 - Long-term Performance of Hanford Low-Activity Glasses in Burial Environments**

**Period of Performance:** July 2004- July 2005
**Russian Partner:** Radon Institute, Moscow, Russia (Principal Investigator – Natalya Ozhovan)
**U.S. Technical Lead:** Pacific Northwest National Laboratory (Principal Investigator – B. Peter McGrail)

**Background & Application**

The Radon Institute has carried out long-term burial testing since 1987 comparing waste form performance in shallow land burial configurations. One of the glasses tested is a high sodium glass similar to those of the Hanford Low Activity Waste (LAW) glasses to be prepared in both the Waste Treatment Plant and by the Bulk Vitrification Alternative. Performance results from that test have been provided to Pacific Northwest National Laboratory to support validation of models used in the LAW performance assessment. Simulants of the actual Russian glass buried at the Radon Institute, Moscow, Russia have been tested by Pacific Northwest National Laboratory (PNNL) and SRNL over the past several years. Non-radioactive materials were provided by Radon to make simulants. The evaluations to date indicate that an analysis of the actual glass buried at the Radon site would greatly benefit tying the data and analysis together. This task will obtain a sample of the RBMK glass that was buried by the Radon Institute in the long-term burial studies and perform a thorough chemical analysis of the sample and conduct single pass flow through testing to determine the forward rate of reaction of the alteration processes. This data will be used by PNNL as field test data to support performance assessment model validation.

**Russian Scope of Work**

- Perform a full chemical analysis of a portion of a glass sample of the RBMK glass
- Perform the Single Pass Flow-Through (SPFT) test on multiple samples of the glass and analyze the leachate for Na, B, Si, and Al [Ca if possible]

**U.S. Scope of Work**

- Based upon an on-site inspection of the Russian facilities and equipment, specify technical data requirements consistent with Russian analytical equipment and interpretation capability for the glass sample that has been buried for over 12 years
- Provide the equipment for performing the single pass flow through test (SPFT)
- Provide training and instruction in SPFT equipment operation and analysis and assist in performance of the initial testing. This test has several performance options. It is essential that the test be performed consistent with PNNL testing.

**Benefits**

- The long-term test performed by SIA Radon on a glass similar to the Hanford Low Activity Waste glass provides validation data that would otherwise be very costly in both schedule and dollars.
- The test was performed with a Low Activity Waste Glass. The leaching data includes radioisotopes.

**Status**

- One joint experimental session was completed in November 2004. The Single Pass Flow-Through Test equipment was set up, tested and processing initiated. Testing will continue for several months with interim analysis of the leachate solutions.

### Task 4 - Improved Retention of Key Contaminants of Concern in Low Temperature Immobilized Waste Forms

**Period of Performance:** August 1, 2004 – September 30, 2005
**Russian Partner:** Khlopin Radium Institute, St. Petersburg, Russia (Principal Investigator – Albert Aloy)

**U.S. Technical Lead:** Savannah River National Laboratory (Principal Investigator **–** Christine Langton)

**Background & Application**

Russian researchers have significant experience in thermodynamic and kinetic calculations that can be applied to grout or other non-thermal waste forms. They also possess the capabilities for radioactive and hazardous (RCRA) testing. In addition, they have the knowledge base to identify existing reagents and technical capabilities to develop new technologies for immobilization of contaminants of concern (COCs).

Innovative approaches to immobilizing COCs 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. This in turn provides additional options to meet accelerated closure goals.

This task aims to develop and test improved ambient temperature waste forms and waste form/engineered landfill systems to achieve immobilization comparable to high-level waste glass for at least Tc-99, Np-237, Se (Se-79), and Cs (Cs-137), with the objective of improving stabilization of mobile/volatile long-lived radionuclides for final disposal. During the first phase of this task, Tc-99 and Cs-137 will be used in the testing. Other radionuclides will be added sequentially in future testing.

**Russian Scope of Work**

- Select and document suitable waste form matrices for thermodynamic and kinetic evaluation in the shallow land fill configuration; identify and document potential suitable/stable phases that can be produced by pretreatment and incorporated into waste form matrices
- Test desorption of Tc as a function of temperature and aging. This will identify the most favorable conditions for stabilizing Tc.
- Develop the selected grout with stabilizers for testing. One test will use water and one will use simulated supernate.
- After a 30-day cure initiate leaching tests before and after $CO_2$ and $O_2$ exposure.
- In parallel with the Tc testing, test Cs sorption using inorganic sorbents followed by incorporation into a hydraulic waste form.
- Construct a mathematical model to evaluate long-term performance. Use the property data developed in the experimental task for the model calculations. Use the model to predict the impact of time and temperature.

**U.S. Scope of Work**

- Review all waste form material constituents to ensure that materials comply with stringent US disposal sites standards and requirements
- Prepare the initial test plan objectives and requirements for review with KRI
- Provide consultation to KRI in the selection of systems and techniques
- Provide technical support for the experiments and guidance on performing the leaching tests to ensure consistency with prior testing.
- Review all data and results and assist in interpreting the data.

**Status**

- Reductants have been screened and selected for Tc-99 stabilization testing.

- Initial test plans have been completed and an "Evaluation of Durability of Mortars and Concretes Used in Ancient Structures" has been completed. That study indicated evidence of Oliva mortars exposed to wind erosion for greater than 2400 years with incredible resistance to the effects of air. Hadrian's Wall in northern England was sampled after 1700 years with the further hardening evidenced by significant amounts of calcium silicate hydrates.
- The grout matrix for the initial testing has been selected and samples were fabricated on January 12, 2005. Once the samples have cured for ~30 days, leach testing will be initiated.

**Task 5 - Documentation of Mixing and Retrieval Experience at Zheleznogorsk**

**Period of Performance:** May 1, 2004 – January 31, 2005
**Russian Partner:** Mining and Chemical Combine, Zhelezhogorsk, Russia
(Principal Investigator – Knostantin Kudinov)
**U.S. Technical Lead:** Pacific Northwest National Laboratory (Principal Investigator – Michael Rinker)

**Background & Application**
Removing consolidated, hardened radioactive waste from high level waste (HLW) tanks has been as much a problem for Hanford as it has been for the Mining and Chemical Combine (MCC) in Zheleznogorsk, Russia. U.S.-Russian cooperation has led to four generations of retrieval technologies. Advancements in Russian mixing and retrieval technologies, such as the pulsating mixer/sluicer, may be applicable to retrieving radioactive waste from HLW tanks in Hanford. A first generation of the sluicer was tested for potential application at Oak Ridge, which prompted the second-generation system design that was ultimately used to clean out the Oak Ridge TH-4 tank. The third and fourth generation systems are under development and testing for use at Hanford and Zhleznogorsk sites respectively. The fourth generation pulsating mixer pump (PMP) will be used at Zheleznogorsk to clean active tanks. MCC stores its waste in a 3,200 $m^3$ tank that has very similar conditions to the Hanford tanks. The MCC retrieval process has been in progress for several years and has a number of the difficult to retrieve characteristics expected as both Hanford and Savannah River continue to retrieve High Level Waste sludge. The U.S. is currently considering the use of the Russian Dual Nozzle PMP in potentially leaking tanks at the Hanford Site. This task aims to provide documentation of the MCC retrieval experience to date. The objective of this task is to leverage lessons learned by MCC in HLW tank waste retrieval.

**Fig. 2. Russian Pulsating Mixer Pump-Cleaning footprint of initial test at PNNL's quarter-scale test facility.**

**Russian Scope of Work:**
- Document operational characteristics of the retrieval equipment and control systems
- Document quantitative and qualitative characteristics of the retrieved sludge
- Document analysis of the gases and vapor space in the tank and
- Document temperature values of the waste during retrieval operations
- Document radioactive contamination in the tank, shielded box and the room where the tank is installed
- Document dose exposures for the personnel directly involved in sludge retrieval

**U.S. Scope of Work:**
- Provide an onsite summary of the current status of High Level Waste retrieval in the U.S. and the performance of sluicing and waste removal equipment.
- Provide expert review and analysis of Russian MCC retrieval equipment, observe and evaluate limitations to site operations, and provide technical guidance to MCC to ensure the MCC reports will be useful to Hanford Office of River Protection
- Review draft reports and provide technical advice and guidance in the preparation of the final three (3) reports

**Status**
- This task was initiated in Mid-October 2004.

**Task 6 - Low-Activity-Waste (LAW) Glass Sulfur Tolerance**

**Period of Performance:** May 1, 2004 – September 30, 2005
**Russian Partner:** Khlopin Radium Institute, St. Petersburg, Russia (Principal Investigator – Albert Aloy)

**U.S. Technical Lead:** Pacific Northwest National Laboratory (Principal Investigators. **–** John Vienna –Glass Formulation and Michael Elliott-Melter Testing)

**Background & Application**

Roughly 50 Million gallons of high-level radioactive waste is stored in 177 underground tanks at the Hanford Site. The US Department of Energy (DOE) has contracted for the design, construction, and demonstration of a waste treatment and immobilization plant (WTP). The Hanford WTP will separate tank waste into low-activity waste (LAW) and high-level waste (HLW) fractions and vitrify them separately into borosilicate glasses. The LAW fraction is composed primarily of sodium salts. A large fraction of the LAW streams have sufficient concentrations of sulfur to negatively impact their loading in glass. Efforts on this task will focus on increasing the loading of high-sulfur LAWs in borosilicate glass by better understanding the relationships between salt formation and the controllable chemical and physical parameters of waste glass melting. Sulfate incorporation is sensitive to feed composition and possibly bubbling/mixing. Under this task, researchers from the Pacific Northwest National Laboratory and Khlopin Radium Institute are collaborating to conduct experiments that will evaluate the impacts of chemical and physical parameters on the incorporation of sulfate into Hanford glasses.

**Fig. 3. Visible Salt Segregation**

**Russian Scope of Work**

- Develop glass formulations with systematic variation in key chemical parameters such as concentrations of alkaline-, alkaline-earth-, and other oxides
- Perform furnace melter testing of the formulations to determine the allowable sulfur loading for each of the formulations both with and without bubbling
- Modify KRI melter to include the ability to bubble the melt pool
- Perform five melter runs to evaluate the five compositions selected
- Perform analyses of the feed composition and glass composition

**U.S. Scope of Work**

- Develop initial test plan for defining the glass formulations that have the highest probability of improving sulfate solubility
- Review results of formulation evaluations and recommendations for additives, etc. to ensure compatibility with the Waste Treatment Plant Process at Hanford. Perform performance testing and model evaluation of critical glass properties and performance.
- Provide oversight and evaluation to guide the large crucible testing
- Provide oversight and consultation during melter tests in Russia to ensure data and interpretation that will be directly applicable to Hanford LAW glass production.
- Provide technical consultation during test data analysis and report writing and ensure that the final report satisfies test matrix requirements

**Status**

- The initial test plan for small and large crucible work has been completed. Melter test plan for larger scale and bubbler testing has been drafted.
- Tin and Manganese additives are being evaluated both at the Khlopin Radium Institute in St. Petersburg, Russia and at Pacific Northwest National Laboratory to determine their impact on sulfate solubility in these glass compositions. Small crucible scale testing will continue in parallel to larger scale testing.
- Using the above additives larger scale crucible testing will be initiated in late January 2005.