

Advanced Remediation Technologies – 9442

Steven L. Krahn, Gary R. Peterson

The United States Department of Energy, Office of Environmental Management

Washington, D.C. 20585

ABSTRACT

The United States Department of Energy (DOE), Office of Environmental Management (EM) is responsible for the cleanup of nation's nuclear weapons program legacy wastes, along with waste associated with nuclear energy programs and research. The EM cleanup efforts continue to progress, however the work continues to be technologically complex, heavily regulated, long-term with a high life cycle cost estimate (LCCE) effort. This article will provide information on the status and relevance of the following Advanced Remediation Technologies (ART) projects that may enhance cleanup efforts and reduce life-cycle costs.

- In-Situ Groundwater Remediation with Enhanced Anaerobic Reductive Precipitation/Enhanced Reductive Dechlorination (ARCADIS G & M Inc., Durham, NC)
- Cold Crucible Induction Melter (CCIM, AREVA NC, Bethesda, MD)
- Near Tank Cesium Removal (Parsons Infrastructure and Technology Group, Inc., Aiken, SC)
- Near Tank Continuous Sludge Leaching (Parsons Infrastructure and Technology Group, Inc., Aiken, SC)
- Steam Reforming Process (THOR Treatment Technologies Inc., Aiken, SC)

INTRODUCTION

EM's mission is one of the most ambitious and far-ranging within DOE, in short, dealing with the environmental legacy of the Cold War. The EM program is one of the largest, most diverse, and technically complex environmental cleanup programs in the world, and includes responsibility for the cleanup of more than one hundred sites across the country. This effort involves site closure activities, processing and disposing of nuclear materials and wastes, and the technology development necessary to facilitate these activities. Progress has been made in the EM cleanup mission, most notably the closure of major sites at Rocky Flats and Fernald. However, the potential for future success at sites with complex nuclear waste and materials issues, such as Hanford and Savannah River, can be enhanced by the successful development and implementation of new technologies.

Some of these technologies are not yet fully tested and others are yet to be developed. It is the mission of the Engineering and Technology program to identify vulnerabilities and reduce the technical risk and uncertainty associated with EM projects and programs. An important source of potential risk reduction strategies and technologies is the private sector. This is where the contribution of the Advanced Remediation Technologies program (ART) comes in.

THE ADVANCED REMEDIATION TECHNOLOGIES PROGRAM, PHASE I

In FY 2005 Congress directed the Department, "... to conduct a competitive evaluation of the various advanced remediation technologies available in the private sector." The resulting Advanced Remediation Technologies program (ART) was structured into two phases. In Phase I, the selected projects were intended to develop a technical approach for demonstration; provide appropriate test results for their technology; develop scale-up plans and an implementation schedule; start detailed system design; and estimate the cost for implementation of the technologies at selected DOE site(s). If Phase I results were promising, DOE then considered a Phase II award(s). Phase II required a pilot/full-scale technology demonstration, preferably at a DOE site.

Improved and advanced technologies, as identified in the EM Engineering and Technology Roadmap, will enable sites such as Hanford, Idaho and the Savannah River Site (SRS) to accelerate their project schedules, reduce life cycle costs, and improve worker safety. Where treatment plants and processing facilities exist, or are already well into design or construction, the technologies are intended to complement the current facilities with minimum disruption.

The Department received 39 proposals for performing work under Phase I of the contract. A total of twelve (12) six-month, fixed price contracts were awarded in 2006. These awards provided \$3.3 million to support the development of technologies that have the potential to reduce cleanup costs and increase the safety and efficiency of treating and disposing of radioactive waste. There was one award to an academic institution; the University of Texas performed work on strontium immobilization in groundwater. Four small businesses won Phase I awards, these included: ARES Corporation for single-shell heel removal, TMR Associates for tank heel removal, North Wind for subsurface characterization, and Commodore Advanced Sciences, Inc., for metals separation. Finally, five large businesses were awarded contracts: Cogema Inc., (2 awards) for cold crucible induction melter technology and tank waste alumina recovery; THOR Treatment Technologies for treatment of Hanford and Savannah River site high level waste, through steam reforming; Parsons Inc., (2 awards) for cesium removal and aluminum and chromium removal; Gas Technology Institute for submerged combustion melting; and ARCADIS G&M Inc., for groundwater remediation.

Upon completion of the six-month contract period the projects were re-evaluated for continuation into a Phase II efforts, based upon those deemed to provide the greatest benefit to the Department's cleanup mission.

THE ADVANCED REMEDIATION TECHNOLOGIES PROGRAM, PHASE II

On September 25, 2007, five ART projects were selected for Phase II. These projects address cleanup of waste stored in High Level Waste (HLW) tanks, Groundwater/Soils remediation and contamination, and vitrification of HLW at Hanford, Savannah River, Idaho and other DOE sites. The five projects were selected for continuation, at an estimated value of \$24.4 M (if all phases are executed) and are as follows.

In-Situ Groundwater Remediation with Enhanced Anaerobic Reductive Precipitation/Enhanced Reductive Dechlorination (ARCADIS G & M Inc., Durham, NC)

This technology is an in-situ bio-reductive process to immobilize contaminant metals and radionuclides within the subsurface at the Savannah River Site. The technique involves the injection of a biodegradable substrate into the subsurface to stimulate the activity of native microorganisms present in the subsurface that will couple the oxidation of the degradable substrate to the production of reduced iron and sulfur species. The reduced iron and sulfur species are known to both chemically reduce and precipitate metal and radionuclide contaminants thereby producing less soluble, and therefore less mobile forms, of these contaminants. This bioremediation technique has been successfully deployed at many locations for a

variety of contaminants. This ART Phase II demonstration focuses on deployment for use in immobilization of DOE contaminants of concern, such as technetium and uranium.

ARCADIS has submitted the permit applications to the State of South Carolina and are scheduled to commence field work this summer.

Cold Crucible Induction Melter (AREVA NC, Bethesda, MD)

Cold Crucible Induction Melter (CCIM) is an alternative vitrification technology that, when successfully deployed, could accelerate the High Level Waste (HLW) vitrification program schedule, reduce lifecycle cost and mitigate technical risks at DOE sites. AREVA and Commissariat à l'Énergie Atomique (CEA) (the French Atomic Energy Commission, equivalent to the Department of Energy) are co-developers of the CCIM technology, while AREVA is the designer and operator of the HLW vitrification facilities at the French fuel reprocessing sites. Extensive testing conducted in France by CEA has resulted in a decision by AREVA to adopt the CCIM technology for use in their HLW vitrification facilities in La Hague, France. Similarly, CCIM technology has the potential to be retrofitted into the Defense Waste Processing Facility (DWPF) at Savannah River, replacing the existing Joule-heated melter (JHM). Notably, this project benefits from the lessons being learned from the La Hague retrofit, which is currently underway and scheduled to be completed in 2010.

CCIM technology offers important potential advantages over the current JHM technology including: higher operating temperatures that support higher waste loadings resulting in fewer canisters produced; a glass pool stirrer that ensures homogeneity of the glass; longer melter life based on the cold shell "skull" created on the melter interior surface that prevents erosion and corrosion of internal structure; and replaceable key ancillary equipment.

The current 18-month project phase (ART CCIM Phase II-A) joins the resources of two US National Laboratories, the Savannah River National Laboratory (SRNL) and the Idaho National Laboratory (INL) as well as two French nuclear engineering and process development organizations, CEA and the Société Générale pour les Techniques Nouvelles (SGN). The project team combines years of experience in the development, design deployment and operation of HLW vitrification processes along with the unique knowledge of the DWPF configuration and operations. The main objectives of the AREVA-led project team are to:

- Perform laboratory studies and testing activities at SRNL and at CEA Marcoule to design a glass matrix which allows high waste loadings;
- Run pilot-scale demonstrations under representative conditions on existing CCIM research platforms at CEA Marcoule and at INL to validate the promising waste processing rates which are expected when processing in the 1,250°C – 1,300°C melt pool temperature range; and
- Perform specific engineering studies of key technical issues identified in Phase I that validate the feasibility of retrofitting CCIM technology into the existing DWPF melter cell.

To date results have been encouraging. The melter feed composition was defined, including the target waste loading level of 46%. Laboratory results indicate that the increased waste loading employed by the CCIM results in acceptable glass. In addition, the project completed preliminary engineering studies. The design inputs and assumptions have been validated, the DWPF melt cell model was created and the preliminary CCIM configuration defined.

Near Tank Cesium Removal (Parsons Infrastructure and Technology Group, Inc., Aiken, SC)

This project develops a portable, modular, and shielded near-tank system to remove cesium from Hanford tank supernates and dissolved saltcake, with a target deployment date before 2012, well before the Waste Pretreatment Facility (WPF) is presently scheduled to be available in 2019. Once the cesium is removed, the low activity waste stream can be vitrified. The ART Phase II technology demonstration is based upon the elutable (re-usable) ion exchange process using the Spherical Resorcinol Formaldehyde resin also planned for cesium removal in the WPF. Successful deployment of this technology will also provide a pilot plant opportunity for the WPF ion exchange technology. The technology also incorporates column elution with cooled nitric acid to prevent resin degradation, and digestion of spent resin using hot nitric acid. After cesium removal, the decontaminated tank farm wastes are likely to be suitable for processing in the Low Activity Waste melter or alternative immobilization processes.

The project has completed Phase II, Task 1 which involved bench-scale testing of batch resin dissolution/destruction tests as well as small column ion exchange performance tests. The results explored operating parameters to achieve desired cesium removal with variable feeds, and also demonstrated that elution with cooled nitric acid (10 to 15° C) did not effect dilution performance. The small column ion exchange tests involved three simulants, two of which bracketed the compositions of the likely first four double-shell tank supernates and the third simulant represented dissolved saltcake. The small column ion exchange tests successfully demonstrated good performance for all target feeds. Resin destruction with hot nitric acid was demonstrated, and some kinetic information was obtained, however in-column destruction was not pursued because of safety issues with rapid gas generation. In these bench-scale tests the reaction was nearly complete after about ten hours, to the extent that total organic carbon destruction was in the range of 43% to 64%, depending on conditions, with no indication of solid residue.

Near Tank Continuous Sludge Leaching (Parsons Infrastructure and Technology Group, Inc., Aiken, SC)

This Continuous Sludge Leaching (CSL) technology develops a modular unit for use in the Hanford tank farms, to be selectively deployed to process tanks with high aluminum oxide concentrations. The technology may reduce the uncertainty and risk to process tank waste sludge which is becoming an increasing concern. Aluminum oxides can be a waste loading limiter for processing certain tanks, and dissolution of aluminum from this stream can reduce the number of HLW canisters. This technology demonstration involves a process to dissolve aluminum, particularly the recalcitrant boehmite mineral (AlOOH) from sludge tanks using a continuous process (versus batch operation) with high sodium hydroxide concentrations and temperatures near 100°C and with concurrent use of cross-flow filtration for separation, thereby reducing the HLW volume requiring vitrification. This technology could assist in early mission completion at Hanford through a reduction in total number of cans processed.

This project has completed Phase II, Task 1 which involves bench-scale testing using boehmite simulant in a one-liter reactor in semi-continuous operation. The kinetic data collected from this testing verified that existing data from the batch operation was applicable and provided a basis for a recommendation of operating conditions to be employed in a pilot scale reactor. Parsons concluded that the reactor should employ a 300 hour residence time, and should operate near conditions in which solution concentrations of aluminum are near 0.5 fraction of the boehmite solubility limit. Parsons also found the reaction to be insensitive to the hydroxide concentrations over the range that was tested.

THOR® Steam Reforming Process for Treatment of Tank Wastes (THOR Treatment Technologies, Inc., Aiken SC)

This fluidized bed steam reforming technology affects the final disposition waste forms for waste streams at Hanford and Savannah River. This process may complement the current treatment using vitrification and may demonstrate several potential advantages over current technologies: the process is low temperature compared to vitrification, potentially allowing retention of key volatile radionuclides that are important to meeting regulatory requirements for groundwater; the final mineralized waste form is a monolith and has similar or better leaching characteristics compared to glass; the process results in no liquid and minimal solid secondary waste streams, NO_x is reduced to Nitrogen; and the process is flexible and can treat various waste streams including those high in organics and sulfur. This technology has been previously tested for Idaho's Sodium Bearing Waste and was selected as the preferred technology.

The project has recently completed the engineering-scale Fluidized Bed Steam Reforming (FBSR) test series using simulated Hanford LAW and LAW recycle. The test unit completed over 200 hours of continuous operation with no unplanned shutdowns. The FBSR process produces a mineralized monolithic product. Early indications are that this product has excellent leach resistance. Materials that have the capability to bind the THOR® mineralized waste product into a monolith waste form are currently being evaluated and tested at Savannah River National Laboratory (SRNL). Laboratory evaluations of the final monolithic waste products will determine volume reductions, waste loading fractions, and chemical and physical properties. THOR will also conduct a bench-scale reformer (BSR) test with actual radioactive LAW or a radioactive spiked LAW recycle simulant to validate radionuclide partitioning and evaluate the performance of the monolithic radioactive waste form. Planning for this activity is complete and a recommended path forward to pursue the LAW recycle application has been provided to DOE.

CONCLUSION

In announcing the ART Phase II awards, Mr. James Rispoli, DOE Assistant Secretary for Environmental Management noted:

“These advanced remediation technologies will enable the Department to demonstrate and implement processes to accelerate high level waste and groundwater/soil cleanup missions across the Department's complex...”

Project kickoff meetings were held at the Office of River Protection and Savannah River sites during the week of October 22, 2007. The status of each project is monitored by a lead DOE engineer, with assistance from technical personnel at major field offices. A mid-year review of ART projects was held in May 2008 where the status of each project was presented. The presentations from this review can be found on the EM-21 web-site at: <http://www.em.doe.gov/Pages/MidyearReviews.aspx>. Subsequently, the various ART projects were presented to the broader EM research community at a multi-year program planning meeting in July.