Title: Nuclear Material Recovery at Los Alamos National Laboratory Using TechXtract® Decontamination Technology

Author(s): Scott Fay (Active Environmental Technologies, Inc.)
David Dennison (LANL)
Keith Fife (LANL)
Wayne Punjak (LANL)

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Abstract

One mission of the Los Alamos National Laboratory (LANL) is to affect pollution prevention and waste minimization surrounding operations at their Plutonium Facility. Efforts are underway and technologies are being deployed to capture the actinide at the source thereby reducing the amount of nuclear material leaving the facility as transuranic waste. Traditional processing alternatives for decontamination, such as strong acid leaching and surface brushing have not achieved the desired recovery efficiencies for plastic or non-actinide metal matrices. Much of the nuclear material present is fixed in the matrix, and is not susceptible to recovery with surface cleaning techniques. In addition, the relatively large secondary waste volumes associated with the acid leaching have persuaded LANL to evaluate alternative recovery methods. The purpose of this paper is to describe the development and testing of a prototype chemical decontamination and co-precipitation process installed at the Los Alamos Plutonium Facility that is based on the patented TechXtract® system developed by Active Environmental Technologies Inc. (AET). The technology was enhanced under a PRDA contract awarded by DOE in 1997.

Introduction and Background

A key effort at LANL is to reduce the global nuclear danger through nuclear weapon stockpile stewardship initiatives and nuclear material inventory disposition activities. Currently, the nuclear material inventories contain a significant amount of non-actinide matrices (plastic, glass, metal, ceramic, and others) contaminated with too much plutonium or other nuclear material to allow disposal as transuranic (TRU) or low-level waste. The current methodology for cleaning these matrices to a suitable level is through a surface leaching technique using strong mineral acids (HCl or HNO3) with or without heat. This technique is sometimes successful but many times is not, especially with porous matrices or matrices that cannot tolerate the acid or the heat. Often, repeated contacts do not affect a sufficient reduction in residual nuclear material content to allow LANL to dispose of the matrix as TRU waste, and LANL has never been able to achieve a reduction to low-level waste. A specific patented decontamination technology (TechXtract®) using a sonication process along with specific cleaning solutions has been
developed by Active Environmental Technologies, Inc. (AET) under a DOE contract. AET, in partnership with LANL, has developed a system utilizing this technology to clean the miscellaneous small scrap accumulated from actinide utilization activities.

TechXtract® Process

The TechXtract® chemical decontamination and co-precipitation system came to the attention of LANL investigators as a result of a PRDA grant report published by the Federal Energy Technology Center (FETC) at Morgantown, WV. Under terms of the grant, TechXtract® was evaluated for scrap material decontamination and volume reduction. Sonication for kinetic energy was proven to be effective. TechXtract® solutions have met or exceeded the following requirements for desired recovery technology:

- Captures smearable and fixed radiation, leaving “free release” levels on/in the matrix
- Does not create a mixed waste
- Minimizes secondary waste volume
- Non-destructive
- Effective for a wide range of radionuclides and substrates
- Attractive total economics
- Generates no airborne contaminants

TechXtract® is a sequential chemical application process, which extracts contaminants from the subsurface pores, capillaries, and voids of hard materials such as concrete, brick, metal, plastic, or wood. The patented combination of chemical constituents and process steps is highly effective in removing radioactive contaminants.

The chemical’s effectiveness is dramatically enhanced by the use of ultrasonic baths. Cavitation caused by the ultrasonics generates both heat and kinetic energy that increase contaminant solubility, and surface volume that aids in the extraction process.

Once the nuclear material is extracted from the scrap matrices it is precipitated from the laden TechXtract® solution by pH adjustment and co-precipitation with ferric chloride as a metal hydroxide. The remaining supernatant is free of radionuclides.

Customized Process for LANL

To satisfy this application AET developed a site-specific TechXtract® protocol to meet the LANL cleanup specifications. LANL requirements were somewhat unique in that they not only wanted to decontaminate the substrate but they also wanted to recover the removed actinides from the chemical solutions to further enhance waste minimization activities. Therefore, AET developed a four-solution process to meet these requirements. These solutions are used in conjunction with sonicator baths to provide the appropriate heat and kinetic energy to increase contaminant solubility and ultimate removal.

Bath 1 consists of TechXtract® 300 solution modified with various reagents. This bath serves to oxidize the various actinide compounds, inorganic impurities, organic impurities, and to alter the
surface chemistry of the various substrates to allow for penetration in subsequent chemical treatments. Bath 2 consists of TechXtract® 200 solution also modified with chemical reagents. This bath is used to reduce the various radionuclides and inorganic impurities. It also forms etching agents responsible for putting oxide forms into solution and promotes radionuclide transport. This solution also introduces complexing agents for Plutonium. Bath 3 consists of TechXtract® 100 solution modified with chemical reagents including NaCl and diatomaceous earth. This bath provides for the elimination of all interfacial tension to allow the charge neutral laden solution to flow out of the substrate. After treatment in Bath 3 the substrate has a net neutral charge. Bath 4 is a rinse bath consisting of distilled water containing about 10% of the TechXtract® 300 solution.

The treatment procedure consists of initially surveying the scrap material to determine the extent of contamination and then placing the loose contaminated material into a tight mesh basket that is compatible with the sonicator bath. It has been shown that a higher solution temperature increases the rate of reaction and system efficiency is enhanced. Therefore, the sonicator baths are brought to a temperature of approximately 43°C (optimum temperature) prior to initiating the treatment process. When the baths are at the proper temperature the basket is lowered into each sonicator in a specific sequence developed by AET. Most of the treatment steps last two minutes or less. After the sequence is complete the object is surveyed to determine if the material is below the appropriate decontamination criteria. If the scrap material meets the release criteria it is loaded into a storage container and the basket is ready for another batch. If it does not meet the release criteria several more steps of treatment are performed to continue the decontamination process.

Additional batches of scrap material can continue to be treated in the same chemical baths until their extraction efficiency decreases to a predetermined level. At that point all the TechXtract® solutions containing the radionuclides can be slowly blended together for pH balance and prepared for further treatment to remove the actinides from the solution. AET has also developed an actinide recovery protocol to recover the actinides from the saturated chemical solutions. This procedure removes the actinides as a metal hydroxide precipitate that is filtered from the mixture. After removal of the precipitate the clear filtrate should contain very low levels of radionuclides and can be disposed of appropriately.

**Glovebox Configuration**

A glovebox-size pilot-scale recovery unit is being assembled at LANL to determine the efficiency of this process in treating the scrap material generated at LANL. Initially only two sonicators will be installed into a selected glovebox to be used for prototype and proof-of-principle tests. Later, if the prototype tests are successful, additional sonicators will be installed to minimize chemical handling within the glovebox. The various chemical reagents will be swapped between the two sonicators as needed to decontaminate the material. Results from this initial testing will be used to forecast production scale throughput. A mass balance will be performed around the process in order to determine nuclear material recovery rates. The prototype setup showing the glovebox configuration is illustrated in Figure 1.
The sonicators used for this project had to meet three specific constraints in order to be accepted for the LANL prototype. First, the maximum external sonicator dimensions had to be small enough to allow the unit to be fit into an existing glovebox. The largest available access opening is a glovebox window opening measuring 12.5” x 20.5”. A special radioactive material control procedure has to be followed in order to remove the glovebox window, install the sonicators, and replace the window. Second, the sonicators had to be capable of providing an ultrasonic transducer density averaging 1500 watts per gallon with a peak output of 3000 watts per gallon. Finally, the units needed to have a multiple frequency capability to minimize the number of standing waves created during cleaning and to minimize “shadowing.”

The sonicators selected for the LANL prototype were custom tabletop Type 800-1 ultrasonic cleaners manufactured by Zenith Ultrasonics in Closter, New Jersey. These sonicators have an internal tank measuring 13.5” long, 16” wide, and 7.75” deep (approximately 7 gallons fluid
capacity) and use the patented Zenith CROSSFIRE ultrasonic system. Each sonicator tank has a total of 12 ultrasonic transducers mounted to the tank bottom and sides. Each sonicator requires the use of two ultrasonic generator modules that are located external to the glovebox. Connections are made to the sonicator tanks inside the glovebox through two electrical feedthroughs that are mounted on a sealed utility plate that is bolted over an opening in the glovebox wall.

Since there are four bath treatments required and only two sonicators installed for this prototype, the various chemical formulations must be swapped out between the sonicator baths and holding tanks installed inside the glovebox during processing. Cylindrical transparent containers compatible with the chemicals are used to store the fluids between uses. The LANL building supplied wet-vac system is used to transfer the fluids between the storage and sonicator tanks. Between transfers the sonicators are rinsed to prevent unwanted chemical reactions between the various solutions. For most operations bath #3 (basic solution) and bath #4 (rinse solution) will be set up first. Then the liquids in those baths will be pumped out and the baths rinsed. Then bath #1 (acid solution) and bath #2 (acid solution) will be set up to continue the treatment process. Then baths #3 and #4 will be set up again to complete the processing of the material.

Conclusion

LANL is currently in the process of ordering the sonicators and other hardware required to set up this prototype system. An existing glovebox has been selected at LANL’s TA-55 Plutonium facility in which to set up this equipment and perform the testing. Minor modifications are being made to the glovebox to accommodate the sonicators and the chemicals. The sonicators will initially be set up on a bench top in a nonradioactive location and checked out for operability prior to installing them into the TA-55 glovebox. Meaningful experimental results are expected before September 1999 and will be presented in detail at this conference.