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## **SESSION 4B: Separation Technologies for Mercury-Bearing Waste**

## Title: Removal and Stabilization of Mercury From Mixed Waste Debris Using Polymer Filtration Technology

We have developed and successively demonstrated a new technology for washing elemental and ionic forms of mercury from solid debris for both high and low subcategories of mercury-containing mixed-waste debris (>260 ppm and <260 ppm Hg, respectively). A full-scale treatability study was completed where the parameters of operation and the performance of the process was determined. Data was obtained on engineering parameters, throughput and waste production, and information for a cost analysis and technology comparison. Many of the debris types tested in this study required sorting, segregating, and size reduction followed by a water prerinse/density separation step to remove interfering chemicals and to collect some free dense elemental mercury liquid. The debris-washing step consisted of a solution of specially prepared synthetic water-soluble metal-binding polymer, in combination with a mild oxidizing agent and dilute acid. The washing solution, when aided by good mixing, assisted in the mild oxidative dissolution of elemental mercury, Hg(0), to ionic mercury, Hg(II). Mercury(II) was desorbed/leached from the debris by the soluble polymer and the soluble polymer prevented the newly generated mercury(II) ions from sorbing onto the debris. The debris was postrinsed in dilute acid and water after leaching and spun to remove excess moisture.

The resulting aqueous mercury-containing leach solution was treated by ultrafiltration, using commercially available membranes, to concentrate the soluble polymer-mercury complex. Permeate, containing water and the remaining oxidizing agent and acid, was recycled to the leaching unit for reuse in the next batch of debris leaching. The ability to reuse the aqueous leach solution multiple times, and the ability to readily remove the ionic mercury from the leach solution and to concentrate it to very small volumes, alleviates the inherent problem of excess waste production associated with typical leaching or solids-washing operations.

The mercury ion concentrate can be separated from the soluble polymer concentrate by adjusting the pH of the system to release the mercury from the soluble polymer followed by a second ultrafiltration process (diafiltration) to collect the mercury. The regenerated soluble polymer can be recycled for further debris leaching. The recovered mercury concentrate was precipitated as the stable sulfide, HgS, with NaHS for solid waste management. Alternatively, the mercury was also stabilized on the soluble polymer concentrate with a sulfide-containing reagent and the small volume of the concentrate was solidified as a final waste form. In cases where the Universal Treatment Standard (UTS) for burial as low-level radioactive waste was not met by leaching alone and the debris met the criteria of being < 260 ppm Hg remaining, the debris was post-treated with a dilute sulfide solution, then meeting the UTS in 100% of the samples tested.

Thirty-six batches of light debris (plastic, Tyvek, paper) were treated in the leaching unit, using conditions developed to readily oxidize and dissolve elemental mercury and leach ionic mercury from debris. The conditions of time, temperature, reagents, pH and general operating conditions were developed from prior experiments with 25 batches of mixed waste debris, and

the developed conditions were used in the final assessments of the treatability study. The total mercury content of each debris batch was >260 ppm for about half of the batches. Data analysis for total mercury concentrations on the debris indicated that in almost every test the mercury level was lowered to below the 260 ppm mercury level after leaching for one hour. Total mercury removal levels of 74% to 99% were observed, with < 2 ppm total remaining mercury reached in 8 out of 34 tests (24%), leaving debris that could be incinerated.

High levels of mercury removal were observed, and the debris batches met the target UTS with the Toxicity Characteristic Leaching Procedure (TCLP) of  $\leq 25$  ppb mercury about 39% of the time with only one-hour of leaching. A treatment target of  $\leq 200$  ppb mercury was an original goal of the project and was met 86% of the time with one-hour leaching. This <200 ppb target was chosen because it represented the treatment limit required for retorting debris that is >260 ppm mercury (EPA standard method), and it was the target at the time this treatability study was initiated. Sulfide post-treated samples met the UTS 100% of the time.

The process operates at near room temperature, at ambient pressure, and uses dilute or low hazard chemical reagents. The process could easily be integrated with other unit operations to remove other RCRA metals. Because of the oxidizing nature of the leach solution other RCRA organics could potentially be treated. The final debris volume was not increased by the leaching operation, but it had some weight increases due to water absorption of certain materials such as paper. In the sorting, segregating, and size reduction process, we observed a 1 in 4 to sometimes a 1 in 10 volume reduction of the as-received debris. The leaching unit could treat combustible-type debris or light debris such as plastic, rubber, personal protective equipment, fabric, paper, cardboard, and other cellulosics. The unit could also treat heavy or noncombustible debris such as glass, stainless steel, fiberglass, and epoxy coated surfaces. Certain materials were incompatible with the process such as reactive metals (e.g., sodium, aluminum, magnesium, potassium, etc). Objects composed of other metals would have their surfaces corroded (e.g., copper, lead, zinc, carbon steel, etc.), which would consume some amount of the oxidizing agent and acid plus contaminate the leachate with their metal ions. Though compatible with the processing conditions, other mixed waste types were considered to be inconsistent with the equipment configuration used in this study including soil, crushed fluorescent lamps, crushed cement, resins, sludge, and other finely divided, dense materials.

Large differences were observed in the cost and throughput estimates between light and heavy debris. For the leaching unit used in this treatability study, throughput could be 2.5 tons/yr of light debris (equivalent to 100 m<sup>3</sup>), or 50 tons/yr of heavy debris (equivalent to 200 m<sup>3</sup>). We estimated that if a ton of light and heavy debris were treated in our unit together, that 12 L of stabilized sludge would be generated in the prerinse step per 2-tons of debris (1% on a weight basis), and 81 L of stabilized solution would be produced in the leach/postrinse/post-treat steps per 2-tons (4.4% on a weight basis) for a total secondary process waste production of 5.4%.

At energy costs at LANL of \$0.055/kW-h, a total energy cost to operate our unit would be approximately \$880/yr. To treat light debris in our unit would cost \$404.80/ton, and heavy debris would cost \$21.12/ton in energy costs. For chemical costs using reagents from Fisher Scientific or Aldrich Chemical (we can not obtain bulk chemical at LANL), light debris would cost \$859/ton and heavy debris \$312/ton with no polymer regeneration (sacrificing the polymer after one week). If bulk chemicals could be used it would cost \$784/ton of light debris and \$92/ton of heavy debris. This new technology (PFT) is cost-comparable to any other leaching technology, generally being less expensive in the initial cost of reagents. It is also competitive in that it produces less secondary waste.