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Cross Flow Filtration of Aqueous Radioactive Tank Wastes

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INTRODUCTION

The Tank Focus Area (TFA) of the Department of Energy (DOE) Office of Science and Technology addresses remediation of radioactive waste currently stored in underground tanks. Baseline technologies for treatment of tank waste can be categorized into three types of solid liquid separation: (a) removal of radioactive species that have been absorbed or precipitated, (b) pretreatment, and (c) volume reduction of sludge and wash water. Solids formed from precipitation or absorption of radioactive ions require separation from the liquid phase to permit treatment of the liquid as Low Level Waste. This basic process is used for decontamination of tank waste at the Savannah River Site (SRS). Ion exchange of radioactive ions has been proposed for other tank wastes, requiring removal of insoluble solids to prevent bed fouling and downstream contamination. Additionally, volume reduction of washed sludge solids would reduce the tank space required for interim storage of High Level Wastes. The scope of this multi-site task is to evaluate the solid/liquid separations needed to permit treatment of tank wastes to accomplish these goals. Testing has emphasized cross flow filtration with metal filters to pretreat tank wastes, due to tolerance of radiation and caustic.

Savannah River Site

The Savannah River Site High Level Waste treatment flow sheet includes (a) the Extended Sludge Process (ESP), (b) In-Tank Precipitation (ITP), and (c) the Defense Waste Processing Facility (DWPF). The stored wastes are slurries containing varying quantities of sludge, salt cake, and supernate. The ESP includes in-tank washing and settling of sludge, followed by decantation of the wash water. The ITP process involves precipitation of Cs with tetraphenylborate (TPB) ion and sorption of Sr and actinides on monosodium titanate from salt solution. In the ITP process, a 1 wt % slurry is concentrated to 10 wt % using the cross flow metal filter and then washed. The solids removal efficiency (>99.9975%) is accomplished with a 0.5 micron stainless steel Mott filter.

A second washing of the slurry to remove sodium nitrite is performed in the Late Wash (LW) process, which is part of the DWPF.

The solid/liquid separation techniques used in the ITP and LW processes at SRS required significant testing in support of start up of the facilities. The filter apparatus used for radioactive waste testing was designed for remote operation and to minimize the volume of sample required. The apparatus has been highly useful in evaluating the filter performance of the production-scale unit during radioactive waste treatment operations. The filtrate from this test unit was shown to meet the product requirements and exhibited flow rates comparable to those in the full scale facility[1] (0.1 to 0.5 gpm/ft^2 from 5 to 25 psid). The apparatus has also been replicated for use at other DOE sites, and utilized in testing at Pacific Northwest National Laboratory (PNNL) and Oak Ridge National Laboratory (ORNL). A modified apparatus is being constructed for use in the Idaho National Engineering Laboratory (INEL). A similar unit is being constructed at the Khlopin Radium Institute in Russia for comparison with Russian filter technologies.

Hanford

The Hanford tank waste is composed of a variety of wastes from nuclear fuel processes. Similar to SRS wastes, the Hanford wastes are alkaline slurries containing varying quantities of sludge, salt cake, and supernate. The goal for Hanford research is to identify the appropriate solid/liquid separation (SLS) process type and approximate size of the equipment. The Tank Waste Remediation System (TWRS) Pretreatment flow sheet for treatment of Hanford high-level tank waste specifies locations for SLS processes. The SLS processes are not yet selected but baseline techniques include concentration and washing of sludges and polishing of Ion Exchange influents.

Testing has been completed on simulated Hanford tank waste at SRTC, and on simulated and actual tank waste at PNNL. For both the concentrated (8.0 wt %) and dilute (0.05 wt %) simulants, a sintered metal filter produced the highest filtrate flow rate.[2] Furthermore, the filtration processes could be divided according to the mechanism that dominated the filtration performance. For dilute simulant, filtrate flow rate is controlled by the pressure drop across the filter due to the formation of filter cake within the filter pores. For concentrated simulant, the filtrate flow rate is controlled by axial velocity across the filter due to the limited back-transport of solids from the surface of the filter. The filtrate quality for all filter types was comparable, and exceeds the estimated criteria for downstream processes. For the actual tank wastes, the filtrates were analyzed for TRU/alpha, Tc, Sr, and Cs. For TRU, the maximum decontamination factors were as high as 2460 at 8 wt%, 1450 at 1.5 wt%, and 48 at 0.05 wt%.[3] For Tc and Sr, decontamination factors were not available, however, the activity levels were significantly lower than the Class A limits. The results support earlier findings that virtually all of the technetium is in a soluble form and Sr is predominantly insoluble. As expected, low decontamination factors for Cs were observed, ranging from 0.69 to 2.02. Filtrate samples were also measured for particle size distributions by photon correlation spectroscopy. Virtually all of the filtrate samples were devoid of solid particles.

Oak Ridge National Laboratory

The Gunite Tanks at ORNL are no longer used for storage of waste but do contain residual heels of sludge which will be sluiced with water and transferred to the Melton Valley Storage Tanks (MVSTs). Since the available MVST tank capacity is limited, sluice waters must be separated and evaporated to conserve tank space. In addition, it will be necessary that sludge contaminated with transuranic materials be contained within the MVST system and not permitted to carry over to newly constructed waste storage tanks, evaporation equipment, or other supernate liquid treatment systems. Tentative plans are to use settling and decanting to separate the sludge from the supernate, however, this is expected to be slow and inefficient. If settling is not sufficiently fast or effective, filtration may be needed. Filtration of simulated Gunite tank waste has been completed at SRTC, and actual waste has been tested at ORNL. The SRTC studies evaluated one dead-end and two cross-flow filters for use in processing the retrieved waste. With simulants, the filtrate flow rate from cross-flow filtration with sintered metal and ceramic/stainless steel filters ranged from 0.02 to 0.14 gpm/ft^2 throughout the range of 0.1 to 15 wt %.[4] Diatomaceous earth prevented clogging of the dead-end filter and yielded a filtrate flow rate of 0.12 gpm/ft^2 for the 0.1 wt % slurry. The filtrate quality for all filter types was comparable, and satisfies the criteria for downstream processes. Furthermore, the filtration processes could be divided according to the mechanism that dominated the filtration performance. For simulants containing 5 wt % or lower solids loading, filtrate flow rate is controlled by the transmembrane pressure across the filter due to the formation of sub-surface foulant. For the 15 wt % simulant, the filtrate flow rate is controlled by axial velocity due to the limited back-transport of solids. At ORNL, the cross-flow filtration tests with radioactive waste were performed in a shielded cell using two sludge composites from the Gunite tanks.[5] The filtrate flux results were in the 0.02 to 0.14 gpm/ft^2 range, which is consistent with measurements from the SRTC simulant studies. No indication of significant solids plugging was observed. Statistical analysis of the cross–flow filtration data showed a significant effect of backpulse on filtrate flux, as expected. Also, the analysis showed significant effects of solids concentration, transmembrane pressure, and axial velocity on filtrate flux. Filtrate flux was lower for increasing solids concentration and higher for increasing transmembrane pressure and axial velocity. As predicted by the SRTC simulant tests, transmembrane pressure had a greater effect on filtrate flux for lower solids content and axial velocity controlled filtrate flux for higher solids content samples. This illustrated the ability to use simulant tests to predict filter performance with actual waste.

Idaho National Engineering Laboratory

The wastes at INEL are highly acidic and therefore significantly different from alkaline wastes at the other sites. The planned disposal method and pretreatment process (liquid/liquid extraction) will require pretreatment to remove insoluble solids. Solids characterization and filter testing will be completed in 1997. Research will focus on both filter performance and chemical cleaning.

Two types of wastes exist at the INEL, with the majority as a solid, granular calcine material, and a small amount of liquid tank waste that is highly acidic (1-2 M). The calcine will be dissolved in nitric acid and the radionuclides separated using liquid/liquid extraction or ion-exchange. Filtration will be used to pretreat the process feed streams to ensure that the waste meets the disposal criteria. Initial characterization of the insoluble tank solids and undissolved calcine solids will be completed in 1997. A cross flow filter, fabricated at SRS, will be tested on simulated and actual waste solutions at the INEL with a focus on filter performance and chemical cleaning. This work will help define the required solids removal efficiency. Cleaning of solids from the filter may require dissolution, however, because they are already insoluble in nitric acid, a more aggressive chemistry will be required. The cleaning procedure must be accomplished with minimal corrosion to the filter media.

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