
by

D. J. McCabe
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808
DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P. O. Box 62, Oak Ridge, TN 37831; prices available from (423) 576-8401.

Available to the public from the National Technical Information Service, U. S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161.
DISCLAIMER

Portions of this document may be illegible electronic image products. Images are produced from the best available original document.
Technology Status Report of the Applicability of Solid-Liquid Separation Methods to Radioactive Tank Wastes (U)

December 22, 1997

D.J. McCabe, 773-43A

Authorized Derivative Classifier

Authorized Reviewing Official

Savannah River Technology Center
Westinghouse Savannah River Company
Aiken, SC 29808

Prepared for the Department of Energy, Office of Science and Technology, Technical Task Plan SR16-WT-41
Summary

Solid-Liquid separation of highly radioactive underground storage tank wastes has been examined, and cross-flow filtration has been shown to be viable. Although sludge settling is often preferred due to ease of operation, cross-flow filtration is needed for many waste streams due to downstream system requirements. This multi-site task has integrated cross-flow filter testing at SRS, PNNL, INEL, and ORNL. Comparison of U.S. and Russian filter technologies has also been completed. Large-scale cross-flow filter technology has been implemented with radioactive wastes at SRS at the In-Tank Precipitation (ITP) facility. Experience gained during testing of the ITP filter system allowed rapid implementation of test programs at the other sites. Pilot-scale testing of cross-flow filtration is planned at ORNL with MVST sludges. Pilot-scale demonstration of cross-flow filtration with waste tank sludges is needed at ORNL and Hanford to prove the long-term viability of the process and to obtain engineering data. Testing at INEL has recently been initiated and additional lab-scale studies are needed.

Introduction

The Tanks Focus Area (TFA) of the Department of Energy Office of Science and Technology addresses treatment methods for radioactive wastes stored in underground tanks. Baseline technologies for removal of insoluble solids from aqueous wastes are either undefined or focus on settling. While settling of insoluble solids is recognized as the preferred method, this approach may not always produce an aqueous phase that is acceptable to downstream processing. Cross-flow filtration has been used at the Savannah River Site for both high and low level aqueous waste solid-liquid separation. The Tanks Focus Area leadership requested examination of the applicability of cross-flow filtration, as well as other filtration methods, to treating tank wastes. This examination was initiated in 1995 and the preliminary work has been completed.

This task sought to:

- Evaluate technologies applicable to the various tank wastes.
- Construct filtration test units comparable to the SRS unit for High Level Waste performance evaluations, if applicable.
Develop consistent guidelines for testing of the technology at DOE sites.

Test the technology on a variety of tank wastes at DOE sites.

Determine if the technology is applicable for pilot-scale testing of DOE tank wastes.

Evaluate use of simulants to mimic filter performance with radioactive tank waste.

A miniature-scale cross-flow filter test unit was developed at the Savannah River Technology Center in support of the In-Tank Precipitation process as part of the DOE EM-30 funded commissioning of that process. The “Cells Unit Filter” (CUF) is used in the Shielded Cells facility at SRTC. The CUF is designed to minimize the quantity of radioactive waste needed to establish the relative filterability of a sample. The actual ITP process filter sizing and engineering data were obtained using larger-scale equipment. The CUF served as the basic design for filter test units constructed at SRS and provided to PNNL, ORNL, and INEEL. The design for the unit was also provided to Russian researchers at the V.G. Khlopin Radium Institute in St. Petersburg, Russia. A similar unit was constructed in Russia and used to compare the U.S. and Russian filtration technologies. The CUF testing provides preliminary performance data, and larger scale equipment is needed to obtain engineering data for plant design.

Cross-flow filtration was the predominant technology tested because of its unique ability to maintain high filtrate production rates and due to the method’s compatibility with highly radioactive slurries. Testing was performed at the sites using simulated and actual radioactive tank wastes. The testing used a statistically-designed test matrix that varied the axial velocity and transmembrane pressure. This test matrix minimized the manpower time needed for each test while producing data that enabled comparisons between different tests and different sites. The filtrate production rate is one of the most important parameters obtained during these tests, but the fouling tendency, filtrate quality, and chemical cleaning protocols also provide important insights. Additional details on the test results can be found in the referenced technical documents.
Hanford Status

A CUF was constructed at SRTC and provided to PNNL researchers in March, 1996. Modifications to the unit were made at PNNL, such as installation of a larger pump to increase the axial velocity and increasing the size of the feed tank. The larger pump size also required installation of an apparatus to cool the slurry. An operating procedure, statistically-designed matrix, and training were also provided by SRTC. A sintered metal filter (0.5 micron pore size) manufactured by Mott Metallurgical was initially used, and a titania-coated sintered metal filter (0.1 microns) manufactured by Graver Separations was used for additional testing.

Experiments using the Parallel Rheology Experimental Filter (PREF) in support of the Tank Waste Remediation System (TWRS) were performed at SRTC. These tests used simulants to establish general parameters for radioactive waste testing. The testing used slurries with a wide range of insoluble solids (0.05 - 8 wt %). The dilute samples represent the estimated solids loading for a decanted supernatant salt solution, where the cross-flow filter is used as a polishing step prior to subsequent treatment such as ion exchange. The concentrated samples (8 wt %) represent the estimated composition of sludge wastes which would be concentrated, washed, and vitrified.

The simulant test results at SRS indicate that moderate filtration rates could be expected and that chemical cleaning was viable. The filtrate quality of all samples exceeded the projected criteria for downstream processes. One of the simulants tested, S-103, was used as a reference standard for testing at the other sites to ensure consistency among researchers.

Testing performed at PNNL using the CUF indicated that cross-flow filtration is a viable option for tank wastes. This data shows that pilot-scale testing is needed to ensure long-term process viability and predict filter fouling. Two waste sludges peptized, resulting in decreased filtrate flow rates. Larger scale testing would help to discern the cause of this flux decrease and determine if it is unique to the high recirculation rates in the small-scale equipment.

As a result of the observed deagglomeration behavior in testing at PNNL, another simulant was tested at SRTC. In addition, this
testing provided a side-by-side comparison of a Mott and Graver filter. Minimal peptizing was observed, indicating that further details on the characterization of the actual waste are needed.

A phosphate-containing precipitate has been observed in caustic wash water from Hanford sludges by Ed Beahm at ORNL. The phosphate is co-dissolved with aluminum during high temperature sludge washing and precipitates on cooling. The phosphate may need removal prior to treatment of the caustic wash water. The filter testing showed that filtration of the solids is feasible, but inadequate time following the solution cooling cycle caused post-filtration precipitates to form. The kinetics of the precipitation must be well understood prior to further testing of this phosphate precipitate.

Oak Ridge Status

A CUF was constructed at SRTC and provided to ORNL researchers in April, 1996. Minor modifications to the unit were made at ORNL to increase the axial velocity. An operating procedure, statistically-designed matrix, and training were also provided by SRTC. A sintered metal filter (0.5 microns) manufactured by Mott Metallurgical was used for all testing at ORNL. Testing at ORNL was performed using two Gunite and Associated Tanks (GAAT) sludges.

Testing of GAAT simulated sludges was also done at SRTC using a sintered metal filter in a dead-end configuration. The separation method utilized diatomaceous earth as a filter precoat and body feed. This method was shown to be impractical for solutions containing 0.1 wt % insoluble solids, but may prove practical at lower concentrations.

Tests in support of the Radiological Engineering Development Center (REDC) and the Newly Generated Low Level Liquid Waste (NGLLLW) were performed at SRTC using simulants to establish general parameters for radioactive waste testing. The NGLLLW program has been canceled at ORNL. The REDC facility has implemented cross-flow filtration of radioactive sludges. In-cell testing with actual REDC waste is underway.

Testing was also performed at SRTC in support of the GAAT clean out and closure. A simulant of GAAT waste was prepared and tested in Mott and Graver filters. Results of the simulant test aided in development of the testing of actual GAAT waste at ORNL.
Testing was also done at SRS to determine if bentonite impacts the filter performance. This information was required because of a proposal at ORNL to combine the GAAT sludges into the Melton Valley Storage Tanks (MVST). An MVST simulant was prepared and tested with and without bentonite. These results were used to address concerns that bentonite has the potential to foul the filters. A significant finding during bentonite-containing simulant testing was that more vigorous chemical cleaning is needed to restore filter performance.

Idaho National Engineering Lab Status

A CUF was constructed at SRTC and provided to INEEL researchers in March, 1997. That CUF was specially-designed to fit into the restrictive entry port of the shielded cells at INEEL. An operating procedure, statistically-designed matrix, and training were also provided by SRTC. A sintered metal filter (0.5 microns) constructed of Hastelloy™ and manufactured by Mott Metallurgical was purchased for use in all testing of this highly acidic waste stream.

Testing completed at INEEL with radioactive sodium-bearing waste and simulated dissolved calcine waste. Filtrate flow rates were slightly higher than those observed with caustic tank wastes, such as at Hanford. Moderate filter fouling occurred over time. Filter cleaning was done by rinsing with water, and chemical cleaning has not been attempted. The Hastelloy™ filter was apparently corroded by the chromate ion, as indicated by the change in color of the solution from yellow (Cr⁶⁺) to blue (Cr³⁺) as it passed the filter. This suggests that the filter may exhibit fouling due to corrosion with long term exposures. Further research is planned in 1998 to examine radioactive calcine waste and chemical cleaning.

Savannah River Site Status

The Cells Unit Filter (CUF) was used at the Savannah River Site to verify the performance of the full-scale filter in the In-Tank Precipitation (ITP) facility. The filter was tested with actual radioactive waste from the SRS storage tanks. Results showed adequate filtration rates and that the decontamination factor was acceptable. This work was funded by EM-30 at the Savannah River Site.
Other filtration units were constructed using EM-30 funds for use with simulants. The Parallel Rheology Experimental Filter (PREF) is a ~1/125th scale unit (compared to ITP) with two parallel filters. This unit is uniquely capable of measuring variances in the performance of filter elements connected in parallel and monitoring changes in the slurry rheology. This unit was used in support of the later Hanford and MVST programs. The Experimental Lab Filter (ELF) is a ~1/700th scale (compared to ITP) single tube filter used for screening experiments. The ELF was used in early testing with Hanford simulants and in testing of the GAAT simulant.

Future programs at SRS will benefit from a much larger knowledge base and retention of cross-flow filtration expertise. This experience can be used at SRS to aid in design of potential future facilities, such as a process to treat Recycle from DWPF. Technical support of the ITP and Late Wash filters will also continue. These filters are critical to the treatment of High Level Waste at SRS. Maintenance of this core competency will be beneficial if problems arise with these filters.

**Russian Research Status**

Design of the CUF was provided to Russian researchers at the V.G. Khlopin Radium Institute. A Mott filter, simulant recipe, and test matrix were also provided. The simulant recipe sent to Russia was for the GAAT sludge simulant tested at SRTC. Results obtained by the Russians with the Mott filter were comparable to those obtained at SRTC. Comparison of results with the Russian-designed filter elements indicates 2-50% higher filtrate flow rates with the Russian technology, depending on conditions. However, the Russian filter had larger pore sizes and allowed a trace amount of solids to pass. Larger pore Mott filters may show comparable filtrate flow rates, but this has not been demonstrated.

**Conclusions**

Cross-flow filtration of tank waste sludges and precipitates has been shown to be feasible. Testing was done with both simulated and actual radioactive tank sludges and precipitates. Larger-scale testing is needed to examine deagglomeration, long-term viability, and to obtain engineering data on filtrate flow rates.
Acknowledgments

This work was done as part of the DOE Office of Science and Technology, Tanks Focus Area program (TTP #SR16-WT-41): C.P. McGinnis, Pretreatment Technical Integration Manager; T. Gutman, DOE-SR Technical Program Office; D. Geiser, Office of Science and Technology Program Manager. Construction of the simulant test units at SRTC and testing in support of the ITP facility was funded by DOE EM-30. The research performed at the V.G. Khlopin Radium Institute was supported by the Tanks Focus Area.

Design Check

C.A. Nash 12/28/97

References


Acronym Definitions

CUF - Cells Unit Filter
DOE - Department of Energy
ELF - Experimental Lab Filter
GAAT - Gunite and Associated Tanks
INEEL - Idaho National Engineering and Environmental Lab
ITP - In-Tank Precipitation
MVST - Melton Valley Storage Tank
NGLLLW - Newly Generated Low Level Liquid Waste
ORNL - Oak Ridge National Lab
PNNL - Pacific Northwest National Lab
PREF - Parallel Rheology Experimental Filter
REDC - Radiochemical Engineering Development Center
SRS - Savannah River Site
SRTC - Savannah River Technology Center
TFA - Tanks Focus Area
TWRS - Tank Waste Remediation System

Distribution

C.P. McGinnis, ORNL
W.B. Van Pelt, 703-T
B.W. Walker, 773-42A
T.E. Kent, ORNL
J. Tripp, INEEL
W.E. Stevens, 773-A

C.A. Nash, 773-42A
W.L. Tamosaitis, 773-A
R.A. Peterson, 773-A
B.A. Reynolds, PNNL
S.D. Fink, 773-A