





# **RETRIEVAL AND TRANSFER OF HIGH-LEVEL** WASTES

# A FUNDAMENTAL UNDERSTANDING OF THE PROPERTIES OF TANK SLUDGES IS REQUIRED TO ENSURE SUCCESSFUL RETRIEVAL AND **TRANSFER OPERATIONS**

A thorough understanding of the chemical properties of waste tank sludges is required to ensure that transfer operations can be made not only without plugging the transfer lines by viscous gels but also with the minimum addition of water to leaking tanks. Even if the relationships between chemical content and flow properties were well understood, it would still be necessary to monitor sludge properties during transfer operations. Some fundamental science issues related to each of these areas are being explored by EMSP projects:

- The objective of one program is to understand the factors that control agglomeration of the colloidal particles that make up the sludge so that strategies for controlling sludge behavior can be devised.
- Another study has focused on the chemistry of aluminate compounds under the • highly alkaline conditions similar to those in the tanks. These studies concentrated on a thorough description of the phase diagram for these materials as well as on the relationships between their microscopic and macroscopic properties. Aluminum and chromium compounds need to be removed from the waste stream prior to immobilization, and some studies related to chromium removal are discussed in the Pretreatment of High Level Waste Fact Sheet.
- Two projects have involved measurements of slurry properties. One research program has examined nuclear magnetic resonance imaging and ultrasonic velocimetry techniques for potential on-line slurry viscosity and concentration measurements. Another has explored fundamental issues related to the use of acoustic probes for determinations of the number density and distribution of particle sizes in slurries.



#### **Measurements of Slurry Properties**

A University of California project (54890) is developing on-line slurry viscosity and concentration measurements. In the graph above (shear viscosity versus shear rate for a 1% polyethylene solution), nuclear magnetic resonance imaging data taken at different spatial resolutions are compared with cone and plate data.

#### **PROBLEMS**/SOLUTIONS

- · Millions of gallons of additional waste may be generated when retrieving tank wastes because of the need to avoid plugging transfer pipes with viscous colloidal gels. Detailed studies of the properties of the colloids that make up the sludges in the waste tanks may lead to chemical treatment methods that can minimize the dilutions necessary for waste retrieval.
- · Process models for sludge treatment require that the chemistry of aluminum in complex electrolytes under highly basic conditions is well understood, particularly with respect to reactions that lead to the formation of solid precipitates. One EMSP project has involved a direct response to this need, which was outlined in a recent Tanks Focus Area Science Needs statement.

### **ANTICIPATED IMPACT**

- It is known that solid volume, viscosity, and ability to retain gases can change dramatically with small changes in composition and temperature, and an ability to predict these properties is particularly important for retrieval of waste from at least 270 tanks with a total volume of about 90 million gallons.
- The formation of colloidal gels can cause a low-viscosity suspension to change into a highly viscous fluid, so models to predict when gels will form in tank sludge retrieval, wash, and leach solutions are crucial to safe tank closure activities.
- Process monitors for high-level waste sludge leaching, washing, and transfer operations are necessary to minimize the volume of wash solutions while avoiding conditions that may lead to precipitate or gel formation. EMSP projects have addressed the particularly important need to develop improved rapid determinations of the density and weight percent of solids in slurries so that the risk of transfer line plugging can be reduced.

## Fundamental Studies of Agglomeration of Tank Sludges

The sludges in the Hanford tanks consist mostly of hydrated oxides of various metals along with insoluble phosphate salts. The objectives of the PNNL/SNL/ University of Washington project (54628) are to understand the factors controlling agglomeration of the colloidal particles that make up the sludge, to determine how agglomeration affects the physical properties relevant to waste processing, and to develop strategies for optimizing processing conditions by control of agglomeration phenomena. Studies of Hanford tank wastes showed that high dilutions may be required when retrieving sludges to keep from plugging pipes and transfer lines with viscous colloidal gels, so their work has focused on possible use of chemical additives to control sludge behavior as well as on understanding the function of certain polymeric steric stabilizers.

#### The Chemistry of Aluminum Compounds in Basic Solutions

An investigation of the relationships between microscopic and macroscopic properties of the highly alkaline salt cakes and slurries in the Hanford tanks is the subject of a LANL/Purdue University study (54773). This project focused on detailed spectroscopic studies of alkaline aluminate chemistry because

## PROJECT TEAMS

#### LEAD PRINCIPAL INVESTIGATOR (AWARD NUMBER)

- Pacific Northwest National Laboratory PI: Joel M. Tingey (54628) Sandia National Laboratory – Albuquerque University of Washington
- Los Alamos National Laboratory PI: Stephen F. Agnew (54773) Purdue University
- University of California Davis PI: Robert L. Powell (54890) Pacific Northwest National Laboratory
- Syracuse University PI: Lawrence L. Tavlarides (55179) Pacific Northwest National Laboratory

large inventories of waste with aluminum are located at Hanford and Savannah River. Aluminate monomor/dimmer equilibria were studied by Raman spectroscopy, and the role of free versus bound water in aluminate speciation was explored. Some gaps in the aluminate phase diagram were being completed so that the consequences of changing composition or temperature could be predicted.

### Measurements of Slurry Properties

The University of California – Davis project (54890) also involves investigations of the properties of slurries, but the emphasis of their work has been on development of on-line slurry viscosity and concentration measurements. They have examined nuclear magnetic resonance imaging and ultrasonic Doppler velocimetry techniques for these measurements. For single phase systems with no suspended solids, comparisons of these techniques with optical velocimetry measurements were made. The ultrasonic monitor is based on simultaneous tomographic pulse-echo ultrasonic flow velocity and time-of-flight speed of sound measurements, and accurate velocity profiles with some fluids with known properties were obtained as the first step in the eventual testing with accurate simulants of slurries in the high-level waste tanks. This group has also made extensive modeling studies of the viscosities of suspensions of particles, and they find that predicted viscosities of systems with particles of different sizes are less than those of systems with uniform-sized particles. They have also worked on understanding the role of colloidal forces on the microstructure in concentrated suspensions.

Acoustic probes are also potential sources of information about the number density and distribution of particle sizes in slurries. Thus, if the acoustic attenuation as a function of frequency and particle size and the volume fraction density of particles of each size are known, then the attenuation as a function of frequency can be calculated. It is, of course, the reverse problem of using the measured attenuation to compute the volume fraction of particles that is of interest, and the Syracuse University/PNNL project (55179) has been directed toward both experimental and computational aspects of this problem in solid-gas-liquid suspensions. They have found that the inverse problem does not have unique solutions for some circumstances, so attenuation measurements alone cannot solve the problem. They have, however, shown that the phase speed changes monotonically with the volume fraction. Thus, future efforts include modifications to the acoustic probe to determine the phase speed of sound waves as well as development of techniques to determine the volume fraction of solids in solid-gas-liquid systems.



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