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Colloidal Agglomerates in Tank Sludge: Impact on Waste Processing

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Research Objective

Insoluble colloidal sludges in hazardous waste streams such as tank wastes can pose serious problems for waste processing, interfering with retrieval, transport, separation, and solidification procedures. Properties of sediment layers and sludge suspensions such as slurry viscosities, sedimentation rates, and final sediment densities can vary by orders of magnitude depending on the particle types present, the degree to which the particles agglomerate or stick to each other, and on a wide range of processing parameters such as solution shear rates, pH, salt content, and temperature. The objectives of this work are to:

- 1) understand the factors controlling the nature and extent of colloidal agglomeration under expected waste processing conditions;
- 2) determine how agglomeration phenomena influence physical properties relevant to waste processing including rheology, sedimentation, and filtration; and
- 3) develop strategies for optimizing processing conditions via control of agglomeration phenomena.

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Research Progress and Implications

This project summarizes work performed after almost two years of a three year project. Significant findings include:

Particles in Actual Tank Wastes - Transmission electron microscopy of actual wastes shows that most sludges consist of agglomerates of submicron ($< 10^{-6}$ m) primary particles of hydrated oxides and insoluble salts. Model colloid suspensions for this work were selected to duplicate the compositions and particle morphologies in actual waste.

Agglomeration of Primary Particles - Static light scattering measurements on both model suspensions and actual wastes show that in the basic salt solutions found in most tank wastes, primary particles undergo extensive aggregation to form fractal agglomerates. The fractal nature of the agglomerates has an enormous impact on slurry properties because fractal objects occupy much more space than dense objects at the same solids loading.

Effect of Agglomeration on Slurry Viscosity - The viscosities of both simple suspensions and actual wastes can vary by over five orders of magnitude (1 centipoise to over 10,000 centipoise) depending on the extent of agglomeration and solids loading. Slurries can exhibit Newtonian (viscosity independent of shear rate), shear thinning (viscosity decreasing with shear rate), or shear thickening (viscosity increasing with shear rate) behavior. The most viscous suspensions contain agglomerates of submicron particles. To achieve desired viscosities for retrieval and transport, sludges of such particles may have to be diluted by as much as a factor of twenty, producing enormous quantities of wastes.

Effect of Agglomeration on Sedimentation - Measurements on simple suspensions and on actual wastes show that sedimentation rates can vary by over three orders of magnitude (from no measurable sedimentation to over 30 cm/hr) depending on primary particle and agglomerate sizes. Measurements on actual wastes indicate that in lab scale tests, final sediment densities of from 1-8 vol% are common. This means: 1) that particulate layers will occupy large processing volumes during steps such as sludge washing and leaching, and 2) that the volume occupied by sediment layers is predominantly interstitial water which cannot be removed during solid-liquid separations.

Effect of Salts on Agglomeration and Sedimentation - Experiments on small gibbsite particles indicate that the solids loading in the sediment layer is highly dependent on the degree of agglomeration and the interparticle interaction potential. If particles are dispersed, particle packing is highly efficient (up to 50 vol%). For agglomerated systems, sediment densities can be increased by as much as a factor of three by weakening interparticle interactions, which increases the compressibility of the sediment. The sediment compacts under its own weight. Experiments have shown that in the high-salt regime of most tank wastes, sediment densities decrease, then increase with salt content due to the interplay of electrical double layer and hydration forces. The presence of calcium and other divalent cations is particularly effective in promoting sediment compression. Salts such as apatite also appear to promote compression by disrupting hydrogen bonding between hydrated oxide particles. Manipulation of such forces could be used to improve tank utilization and the efficiency of solids-liquids separations during steps such as sludge washing and leaching by as much as a factor of four.

Planned Activities

While work done so far in FY98 suggests that short range forces are important in controlling the properties of tank sludge, quantitative models do not yet exist to predict sludge properties such as sediment densities or slurry viscosities based on interaction potentials. Work in FY99 will focus on the following tasks aimed at developing quantitative models for sludge properties based on tank waste processing chemistry:

Measurement of Short Range Forces - Interaction potentials will be measured between surfaces relevant to tank waste disposal using a surface forces apparatus.

Measurement of Agglomerate Sized and Structures - Small-angle x-ray scattering (SAXS) and light scattering will be performed on model suspensions in both static and sheared solutions to determine how different salts influence the fractal dimension and size of agglomerates of submicron particles.

Modeling of Agglomerate Properties - Computer codes developed at PNNL and SNL will be used to calculate the kinetics of aggregation, provide information regarding agglomerate sizes and structures, and calculate the ease of deformation of agglomerates as a function of the magnitude of interparticle interaction potentials.

Once the framework relating interparticle forces, agglomeration, and sludge properties is established, the project will shift to deliberate modifications of sludge suspensions to optimize properties for retrieval, transport, and sedimentation processes.

Other Access To Information

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