

Project 94644

Foaming and Antifoaming and Gas Entrainment in Radioactive Waste Pretreatment and Immobilization Processes

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RESULTS TO DATE: SUMMARY/PROJECT REPORT REQUIRED DATE

“Foaming and Antifoaming and Gas Entrainment in Radioactive Waste Pretreatment and Immobilization Processes”

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Number of graduate student/post-doctoral students involved: 2

PROGRESS REPORT

RESEARCH OBJECTIVE

The objectives of this research effort are to develop a fundamental understanding of the physico-chemical mechanisms that produce foaming and air entrainment in the DOE High Level (HLW) and Low Activity (LAW) radioactive waste separation and immobilization processes, and to develop and test advanced antifoam/defoaming/rheology modifier agents. Antifoams/rheology modifiers developed from this research will be tested using non-radioactive simulants of the radioactive wastes obtained from Hanford and the Savannah River Site (SRS).

RESEARCH PROGRESS AND IMPLICATIONS

The Savannah River Site (SRS) and Hanford site are in the process of stabilizing millions of gallons of radioactive waste slurries remaining from production of nuclear materials for the Department of Energy (DOE). The Defense Waste Processing Facility (DWPF) at SRS is currently vitrifying the waste in borosilicate glass, while the facilities at the Hanford site are in the construction phase. Both processes utilize slurry-fed joule-heated melters to vitrify the waste slurries. The DWPF has experienced difficulty during operations. The cause of the operational problems has been attributed to the rheological properties of the process slurries. The rheological properties of the waste slurries limit the total solids content that can be processed by the remote equipment during the pretreatment and meter feed processes. Highly viscous material can lead to air entrainment during agitation and difficulties with pump operations.

During the first year of the three-year project, experimental investigations were pursued which were specifically aimed at discerning the effects of various rheology modifiers or dispersion agents on micro-rheology of the waste slurries/sludges in order to reduce the apparent viscosity and yield stress and mitigate the process difficulties.

The first task accomplished in the grant period involved establishment of the effects of the rheology modifier on the slurry particle-particle interaction and slurry micro-texture of the two slurry samples SRAT (Slurry Receipt and Adjustment Tank) and SME (Slurry Mix Evaporator) simulants and their mixtures prepared at the Savannah River National Laboratory (SRNL). The SRAT slurry is an 18 wt% solid suspension of 1-5 μm sized particles with irregular shapes that tend to form a gel-like particle network. The SME slurry is a 44 wt% solid suspension of SRAT slurry with Frit 418 (irregularly-shaped glass particles ranging in size from 80 to 200 mesh from Fusion Ceramic).

The SRAT and SME slurry particle-particle interactions and slurry texture were probed by both the novel capillary force balance technique in conjunction with differential interferometry, and laser scanning confocal microscopy. The effect of the modifier Dolapix CE-64 (at 4000ppm) on the SRAT slurry micro-rheology was examined using the capillary force balance technique. The micro-rheology measurements reveal that the modifier Dolapix CD-64 efficiently reduces the pressure threshold or yield stress from 650 dyne/cm² without a modifier to 450 dyne/cm² with a modifier. The internal slurry structure is seen to change from a gel-type to a 'cluster' type (i.e., particles form flocs) in the presence of Dolapix CD-64.

The effect of the modifier Dolapix CE-64 on the SME slurry was probed by laser scanning confocal microscopy, which showed that the modifier promotes particle flocculation of frit particles due to the presence of fine SRAT particles, and leads to the grainy texture with the formation of voids. These experimental observations were made at the SRNL facility.

In summary, these experiments reveal that laser scanning confocal microscopy in conjunction with the capillary force balance method is a useful tool to address rheological problems of the waste slurries.

Under task two the interparticle interactions in the processed slurries were probed by the batch sedimentation method. The batch settling (sedimentation) experiments were conducted to verify the role of the modifier Dolapix CD-64 in enhancing slurry particle flocculation. The reference sample of the SRAT slurry without the modifier showed that 25 vol% water was released (separated). The samples of SRAT with Dolapix CE-64 (at 4000 and 6000 ppm) showed that about 38 vol% water was separated as compared to 25 vol% without Dolapix CD-64. Another modifier, Disperse-AYD DA W-28 in concentrations ranging from 2000 to 6000 ppm was also tested and showed only 31.5 vol% water separation. Overall, Dolapix CD-64 was found to have a better water separation performance.

The sedimentation performance of the SME slurry in the presence of Dolapix CD-64 was also studied. The water release is 9 vol% in the presence of 4000 ppm Dolapix CD-64 as opposed to 5 vol% without a modifier after four days, which shows that this modifier also performed well with the SME slurry.

Task three involved the bulk rheological measurements of the waste slurries using the Brookfield viscometer. The effect of the modifier Dolapix CE-64 on the bulk rheology was also investigated. The modifier reduces the shear stress of the SME slurry (at 6 sec-1) by 30%. The shear stress versus the shear rate for both SRAT and SME slurries increases; however, the rate of the increase for the SME slurry is 3.5 to 4 times higher. This result is consistent with the fact that the solid particle concentration of SME product is about two times higher (44 wt%) than the SRAT slurry.

The effect of the frit particle shape on the shear stress was also studied. We replaced the irregularly-shaped frit glass particles (diameter 100 μm) of the SME product with spherical glass particles, with an average diameter of 50 μm . The SME slurry with these new spherical frit particles has a lower shear stress than the original irregularly-shaped slurry particles currently used in the SME product. The shear stress of the SME slurry with the modifier Dolapix CE-64 and with the spherical frit particles was more than 45% less than the SME slurry with irregularly-shaped particles. Therefore, changing the shape of the raw materials added to the Savannah River High Level Waste Immobilization Process decreases the yield stress of the slurries that are processed.

The recommended process changes, based on the research performed by IIT and SRNL researchers in the first year of the project, are expected to result in substantial operational improvements and cost reductions.

INTERACTIONS AND COLLABORATIONS

The principal investigator (Wasan) and his co-investigator (Dr. Nikolov) are currently working with SRNL researchers to further understand surface chemistry involved with the process using SME product to determine the feasibility of implementing the changes recommended by IIT researchers to mitigate process difficulties.

Also during the first year of this project, the co-principal investigator and the IIT graduate student spent more than two weeks at the Savannah River National Laboratory (SRNL) carrying out experiments using the laser scanning confocal microscope.

We held a coordination meeting for all DOE projects related to foaming and gas entrainment and rheology problems at the Savannah River, Hanford and Oak Ridge sites. The main purpose of this meeting was to assure that the results of the basic research conducted at IIT are applicable to foaming/antifoaming and gas entrainment in high level waste pre-treatment and immobilization processes. Also, plans for research to be carried out in the future at IIT were discussed. The technology users at Savannah River and Hanford stated that gas entrainment/foaming and the impact of these phenomena on the rheology and processability of the waste continues to be a high priority technology need.

PLANNED ACTIVITIES

1. Investigate interparticle interactions and microstructure formation using back-light scattering (i.e., Kossel diffraction) method and correlate the results with micro-rheological measurements. 2. Perform sedimentation experiments in conjunction with back-light scattering technique to measure particle settling rates and correlate these results with micro-rheological measurements. 3. Determine the impact of rheology modifiers on particle settling rate. 4. Conduct foaming experiments with a model slurry to monitor bubble dynamics and foam lamella texture in three-phase gas-liquid-solid systems.

DELIVERABLES: PUBLICATIONS

1. "Foaming and antifoaming in boiling suspensions", D. Wasan, A. Nikolov, A. Shah, *Industrial and Engineering Chemistry Res*, 43, 3812-3816 (2004). 2. "A novel method to study particle-air/liquid surface interactions", A. Nikolov and D.T. Wasan, *Colloids and Surfaces A*, 250, 89-95 (2004) 3. "Foaming and Antifoaming in a Gas-Liquid Finely Divided Particulate System" K. Vijayaraghavan, A. Nikolov, D. Wasan, B. Calloway, M. Stone, D. Lambert, J. Chin. *Inst. Chem. Engrs.* 26 (1) 37-41 (2005). 4. "Radioactive Waste Foams: Formation and Mitigation," K. Vijayaraghavan, A. Nikolov, D. Wasan, B. Calloway, M. L. Crowder, M. Stone, Z.H. Quershi, *J. Environ. Eng.* (under review, 2005). 5. "Micellar Films: Thinning and Structure," A. Nikolov, D. Wasan, "Encyclopedia of Surface and Colloid Science", (under review, 2005). 6. "Foaming-Antifoaming in a Three-Phase Gas-Liquid-Particulate System" K. Vijayaraghavan, A. Nikolov, D.T. Wasan, submitted to *Advances in Colloid Interface Science* (under review 2005).

PI Awards and Honors for Research Performed under DOE Grant

American Institute of Chemical Engineers Alpha Chi Sigma Award for Fundamental and Applied Chemical Engineering Research, 2005.

Elected to the U.S. National Academy of Engineering, 2004.