SUMMARY/PROJECT REPORT REQUIRED DATA

"Foaming and Antifoaming in Radioactive Waste Pretreatment and Immobilization Processes"

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Number of graduate student/post-doctoral students involved: 1/2

PROGRESS REPORT

RESEARCH OBJECTIVE

The objective of this research is to develop a fundamental understanding of the physico-chemical mechanisms that cause foaminess in the DOE High Level (HLW) and Low Activity radioactive waste separation processes and to develop and test advanced antifoam/defoaming agents. Antifoams developed for this research will be tested using simulated defense HLW radioactive wastes obtained from the Hanford and Savannah River sites.

RESEARCH PROGRESS AND IMPLICATIONS

Savannah River National Laboratory (SRNL) has reported severe foaminess in the bench scale evaporation of the envelope C tank. Excessive foaming in waste evaporates can cause carryover of radionuclides and non-radioactive waste to the condensate system. The antifoams used at Hanford and tested by SRNL are believed to degrade and become inactive in high pH solutions. Hanford wastes have been known to foam during evaporation causing excessive down time and processing delays.

During the third year of this three-year project, experimental and theoretical investigations of the surface phenomena, suspension rheology, and bubble generation interactions that lead to the formation of foams during waste processing were pursued.

The first task accomplished in the grant period involved establishment of the major mechanisms of formation and stabilization of foams containing very small insoluble particles since the radioactive waste foams are typically stabilized by such particles. The solid particles stabilize foams in two ways: by adsorption of biphilic or amphiphilic (i.e., containing both hydrophilic and hydrophobic parts) particles at the surface of the foam lamella (liquid-gas interfaces) and by layering of the particles trapped inside the foam lamella or film. During bubble generation and rise, hydrophilic solid particles organize themselves into a layered structure due to confinement in between neighboring bubbles, and this structure provides a barrier against the coalescence of the bubbles, thereby causing foaming and foam stability. This novel stabilization mechanism involving particle structuring phenomenon in macrodispersions such as foams opened new vistas in dispersion science and engineering. The results of this research were published in a series of papers (see publications under this grant, 1-2). This research was recognized by the American Institute of Chemical Engineers with its national award in fluid-particle systems presented to the principal investigator, Wasan. Also, he was elected to the National Academy of Engineering this year for his accomplishments in interfacial engineering and colloidal processing.

Fundamental research into the particle structuring phenomenon in the confined boundaries of a film also led unexpectedly to a new mechanism for soil remediation. This work is described in our paper published in the journal *Nature* in 2003 (see ref. 3 in publications under this grant).

Under Task Two, recycle streams (melter off gas condensates) that are being generated by the WTP pilot facilities were evaluated by us and found to contain foam-causing surfactants. Previously, these recycle simulants were thought to be benign in nature and were not included by the baseline WTP R&T tasks. Two simulants of RPP-WTP recycle streams that were generated during pilot scale operations (LAW Submerged Bed Scrubber (SBS) and HLW Submerged Bed Scrubber (SBS)) were studied for foaminess. Due to the presence of the surfactants LAW Submerged Bed Scrubber (SBS) showed a maximum foaminess of 200 vol% after evaporation to 4 wt% total solids.

Task Three involved an investigation of the three major mechanisms of antifoaming of foams stabilized by solid particles, (i) flocculation, which introduces polydispersity in particle size and leads to the weakening of particle layering (i.e., structural barrier), (ii) wetting the surface of biphilic particles to become more hydrophilic (water wet) to aid in particle settling rates, and (iii) disintegration of the structural stabilization barrier by generating a surface tension gradient at the air/slurry surface using a surface-active agent (surfactant).

Based on the mechanistic understanding of the antifoaming action in solid particle-stabilized foams, and working closely with the Savannah River National Laboratory, we have successfully developed and tested antifoam agent IIT-B52 to mitigate foaming in the process being developed for the disposition of SRS HLW radioactive salt solutions. This new and advanced antifoam agent was found to be more effective than those commercially available. Process demonstration studies using advanced antifoam agent IIT-B52 revealed that the antifoam agent also acts as a powerful rheological aid that can dramatically reduce the rheology of SRS HLW potassium tetraphenylborate slurries and also can act to de-entrain air from the slurry. Thus, fundamental understanding of the physio-chemical factors that affect foaming and air entrainment can lead to the development of antifoam and rheological agents that can enhance the processing characteristics of DOE HLW slurries. When the yield stress of these wastes is reduced, the glass-forming solids content of the waste can be increased. If properly developed and deployed, these antifoam agents and rheology modifiers can increase the waste content of the DOE slurries that are immobilized. Increasing the waste content that is immobilized can increase the waste processing throughput which will accelerate the DOE mission.

We held a coordination meeting for all DOE projects related to foaming problems at Savannah River, Hanford and Oak Ridge sites. This meeting was held in December 2003. The main purpose of this meeting was to review the progress made and assure that the results of the basic research conducted at IIT are applicable to foaming/antifoaming in high level radioactive 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 SRS 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. As recommended by SRNL, the effects of soluble and insoluble inorganic compounds on gas entrainment and foam formation in defense waste matrixes will be established.
2. We plan to develop a fundamental understanding of the effect of the interaction of soluble and insoluble waste component on the microrheology (i.e., yield stress) using our capillary force balance method in conjunction with differential interference microscopy.
3. Characterize the three-phase gas-liquid-solid interface structure using a confocal microscope in waste slurry simulations.
4. A short list of potential rheology modifiers will be recommended to the users at Savannah River and Hanford sites.

PUBLICATIONS

1. "Stability of Films with Nanoparticles," G. Sethumadhavan, A. Nikolov, D. Wasan, J. Colloid Interface Sci. 272, 167-171 (2004).

2. "Foaming and Antifoaming in Boiling Suspensions," D. Wasan, A. Nikolov, A. Shah, Ind & Eng Chem. Res. accepted (in press 2004).

3. "Spreading of Nanofluids on Solids" D.T. Wasan and A.D. Nikolov, Nature 423, 156-159 (2003).

4. “Radioactive Waste Evaporation: Current Methodologies Employed for the Development, Design and Operation of Waste Evaporators at the Savannah River Site and Hanford Waste Treatment Plant,” T.B. Calloway, W.R. Wilmarth. J.E. Josephs, R.E. Eibling, D.A. Crowley, C. J. Martino, M.E. Stone, C.D. Barnes, A..S. Choi, M.A. Baich, C.M. Jantzen, R.A. Pierce. W.E. Daniel, T.L. White, J.D. Johnson, K. Vijayaraghavan, A. Nikolov and D.T. Wasan. Proceedings of ICEM ‘03: The 9th International Conference on Radioactive Waste Management and Environmental Remediation. September 21-25, 2003, Examination School, Oxford, England.

5. "Laser Scanning Confocal Microscopic Investigations of Simulated Nuclear Waste Structures," T. B. Calloway, R. L. Brigmon, W.E. Daniel and R. E. Eibling, Journal of Visualization, Vol. 7 No. 2 (2004) 108, April 10, 2004.