SUMMARY/PROJECT REPORT REQUIRED DATA

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

DOE Report Number: DE-FG07-01ER14828
Date: August 1, 2002
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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 Technology Center (SRTC) 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 SRTC 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.

An experimental study, to understand foaminess was carried out using nine Hanford sludge simulants. Foaminess and foam textures during boiling of the simulants at both atmospheric pressure (760 mm Hg) and at low pressure (110 mm Hg) were monitored. The foaminess of all the simulants goes to a maximum at higher solid concentration (>50%). At atmospheric pressure, severe foaminess (600%) was observed in sludge samples AN-107 with entrained solids, AN-107 3M and pretreated AN-107 at the maximum in foaminess. Less foaminess at atmospheric pressure was observed in sludge samples AN-104 3M, AN-104 with entrained solids, AN-105 3M with entrained solids, AN-105 3M, AZ-101 and AZ-102. The AN104, AN105, AZ101 and AZ102 salt solutions are similar to those evaporated at Savannah River HLW Evaporators, therefore we expect the results and conclusions to be beneficial to both Hanford and Savannah River sites.

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 two maxima. The first maxima was observed at 4 wt % solid concentration with a foaminess of 200 vol % and the second maxima was at 15 wt % solid concentration with a foaminess of 150 vol %. Simulant HLW Submerged Bed Scrubber (SBS) showed only one maximum due to negligible amount of surfactant.

The increase in water evaporation flux rate enhances foaminess. Less foaminess was observed at low pressure (110 mm Hg) and low water evaporation flux (0.05 ml/min sq.cm). However, at low pressure and high evaporation flux rate (0.09 ml/min sq. cm) the maximum foaminess was about 1200%. This shows that foaminess increases with increasing water evaporation flux. Extrapolated data for water evaporation flux (0.56 ml/min sq. cm) simulated at the Hanford plant show severe foaminess (>1500%) at low solid concentration (<40%).

The addition of 25 ppm of normal paraffin hydrocarbon (NPH) and tributyl phosphate (TBP) to the sludge samples with severe foaminess did not show reduction in foaminess. However, addition of 100 ppm of NPH increases foaminess at maximum by 25% and addition of 100 ppm TBP reduces foaminess by a factor of three at maximum. NPH and TBP are also encountered in the SR HLW salt solutions, therefore these results will also provide a better understanding of how these compounds effect the foaminess of SR HLW salt solutions.

The Savannah River Technology Center has reported that during the sludge processing some surfactants could be generated or formed inside the sludge. During the boiling of the alkaline sludge, some of the hydrocarbon derivatives present in the sludge decompose and become surface active. Therefore, we carried out a systematic study to understand the effect of the type and concentration of the surfactants on the foaminess. For this study, we used two commercial surfactants, hexadecyl trimethyl ammonium bromide (a cationic surfactant) and sodium salt of dodecane sulphonic acid (an anionic surfactant) and the simulant AN-107. Different concentrations of the surfactants were tested, varying from 1000 to 3300 ppm. Due to the presence of the surfactants, the surface tension decreases and small bubbles are formed during boiling. The gas solubility is high at low temperatures, but with an increase in temperature, the gas solubility decreases and more gas is released. This leads to the generation of more bubbles, as a result of which the foaminess increases. As time progresses, the temperature increases and the gas solubility achieves a steady state, and the foaminess decreases. The nuclei or fine crystals are formed when the solution becomes over-saturated. The crystals attach to the foam lamella surfaces and provide a structural stabilization barrier, which makes the foam lamella stable.

Model systems, which could provide us with more insight into the role of fine particles (nuclei) and surfactants in foaminess, were investigated by us using sodium glyconate (NaOH and glycolic acid) and NaCl concentrated solution. We concluded from these experiments that NaCl showed maximum foaminess due to the formation of nucleic particles. This confirms our observation for the more complex system such as AN-107 simulant where the maximum foaminess was also observed due to the presence of particles. Foaminess due to the sodium glyconate was less.

The antifoaming efficiency of two antifoamers (DOW PULPAID CONCENTRATE 3472 and DOW 1520 US) was tested. The antifoaming performance of DOW 1520 US was poor. Antifoamer DOW PULPAID CONCENTRATE 3472 reduces the foaminess at low pressures by a factor of three (at foaminess corresponding to maximum). However, its antifoaming efficiency needs to be studied at higher evaporation fluxes.

PLANNED ACTIVITIES

1. As recommended by SRTC, we plan to improve the evaporation flux rate since the foaminess seems to increase at a higher water evaporation rate.

2. We plan to use other, non-radioactive simulants from Hanford waste, such as AN-102 and other mixed simulants from mixed recycled waste solutions from the HLW and LAW vitrification process with the LAW feed solutions. Evaporation of these mixtures is likely to differ from the evaporation of only LAW solutions.

3. A short list for potential antifoam solutions will be developed.

4. Hold a coordination meeting of all DOE projects related to foaming problems at Savannah River, Hanford and Oak Ridge sites.

5. The degradation of the selected antifoam agents in caustic media will be investigated. The decomposition (hydrolysis) kinetics and the degradation products will be experimentally determined as a function of hydroxide concentration.

6. A series of papers based on the fundamental results will be prepared for possible publication in refereed journals. One such paper is currently in preparation.

PUBLICATIONS

1. "Stability of Thin Liquid Films Containing Polydisperse Particles," G. Sethumadhavan, S. Bindal, A. Nikolov and D.T. Wasan, Colloids and Surfaces 204, 51-62 (2002).

2. "Confinement-Induced Structural Forces in Colloidal Systems," D.T. Wasan, A.D. Nikolov, A. Trokhymchuk and D. Henderson, in *Encyclopedia of Surface and Colloid Science,* A. Hubbard, Ed., Marcel Dekker, Inc., NY pp 1181-1192, (2002).

3. "Effect of Film Curvature on Drainage of Thin Liquid Films," K. Kumar, A.D. Nikolov and D.T. Wasan, J. Colloid Interface Sci., (in press, 2002).

4. "Foaming Mechanisms in Surfactant Free Particle Suspensions" S.K., Bindal, G. Sethumadhavan, A.D. Nikolov and D.T. Wasan, AIChE J., (in press 2002).

5. "Computer Modeling of Ionic Micelle Structuring in Liquid Films," A. Trokhymchuk, D. Henderson, A. D. Nikolov and D.T. Wasan (Submitted to J. Phys. Chem., 2002)