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LABORATORY SIMULATION OF HIGH-LEVEL LIQUID WASTE EVAPORATION AND STORAGE

by

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The reprocessing of nuclear fuel generates high-level liquid wastes (HLLW) which require interim storage pending solidification. Interim storage facilities are most efficient if the HLLW is evaporated prior to or during the storage period. Laboratory evaporation and storage studies with simulated waste slurries have yielded data which are applicable to the efficient design and economical operation of actual process equipment.

Simulation of HLLW

Simulation of reprocessing waste streams is a practice dictated by the difficulties and expense of experimenting extensively with actual radioactive solutions. The chemical compositions of solutions used in these studies were based upon those used in the waste fixation program at Battelle Pacific Northwest Laboratory[1,2,3], government reports[4], information from commercial reprocessors[5], and the ORIGEN Computer Code. The slurries used for most of these studies represented first-cycle raffinates from the Purex reprocessing of LWR fuel irradiated to 33,000 MWd/MTU and cooled for 150 days. An initial raffinate concentration of 4900 L/MTU was assumed.

Simulated HLLW solutions were prepared by combining a series of stock solutions to closely approximate the introduction of components into an actual process solution. The validity of substituting and omitting certain rare and expensive elements was investigated. Fission products with concentrations less than 0.1% of the total fission product content were omitted. For some experiments, the fission product ruthenium was omitted because it is expensive and no available substitute is known to simulate its unique properties. Less expensive elements were substituted for rhodium, palladium, and technetium. When compared to slurries without these substitutions or omissions, no differences were observed in any of the physical and chemical properties which were studied, including the amounts and composition of undissolved solids.

Evaporation

Simulated HLLW slurries were evaporated in both a batch type and a continuous glass evaporator. In both cases, the limit to which HLLW can be evaporated and still remain a fluid with easily suspendable solids was approximately 340 L/MTU. As shown in Figure 1, the quantity of total undissolved solids

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increased rapidly as the slurry was evaporated to volumes less than 800 L/MTU. At all degrees of evaporation, the quantities of total undissolved solids were greater in solutions with lower acidities. Freshly prepared HLLW slurries contained lesser quantities of undissolved solids than slurries that had been heated to boiling.

Filtered samples of undissolved solids were analyzed using X-ray fluorescence, X-ray diffraction, emission spectroscopy, and scanning electron microscope (SEM) techniques. Under all conditions studied, the major components in the undissolved solids were zirconium and molybdenum. Phosphorus was always present, but frequently in lesser amounts. Some of the wide variety of crystalline forms and particle sizes which were observed are shown in Figure 2. Most samples yielded identical X-ray diffraction patterns which correspond to the pattern for $ZrMo_2O_8 \cdot xH_2O$. Undissolved solids of this composition have been identified in concentrated extraction raffinates in hot cell studies from irradiated LWR fuels.[6] One or more still unidentified compounds of zirconium, molybdenum, and phosphorus were observed in the undissolved solids from HLLW containing higher concentrations of phosphate.

Other physical and chemical properties which were obtained for simulated HLLW slurries at various degrees of evaporation include densities, viscosities, boiling point rises, acidities, and nitrate concentrations.

Storage

Various concentrations of HLLW slurries containing possible process additives were stored up to two years at 60°C with no significant changes in quantities of undissolved solids. The median settling rate for undissolved solids particles in simulated HLLW evaporated to 340 L/MTU was approximately 4×10^{-5} m/s.

The undissolved solids are easily kept suspended by frequent gentle agitation. If permitted to settle for several days, more vigorous agitation is required to resuspend the undissolved solids.

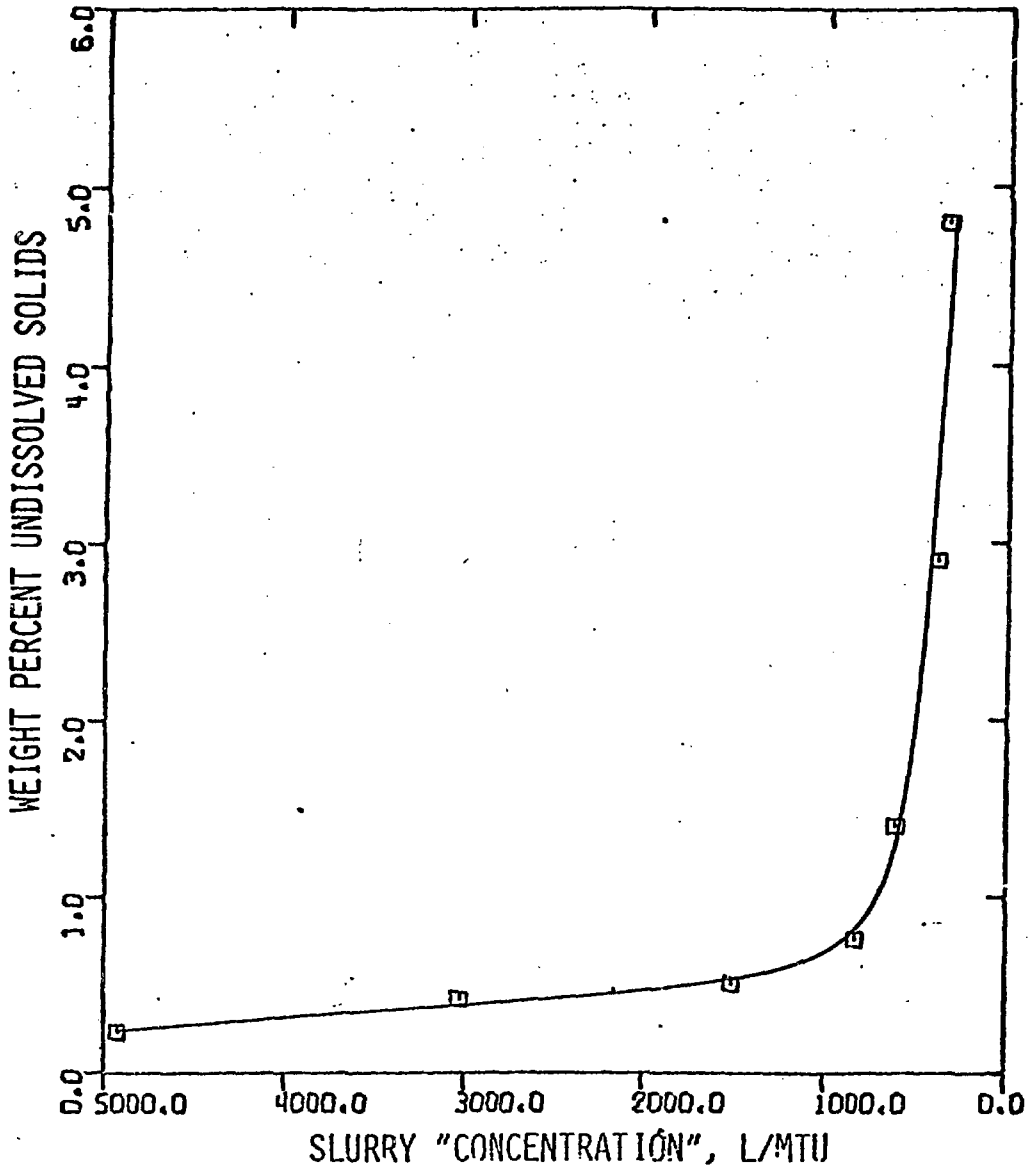

Figure 1. Relative undissolved solids in evaporated HLLW.

a) Solids from HLLW without soluble poison.

b) Solids from HLLW containing gadolinium as soluble poison.

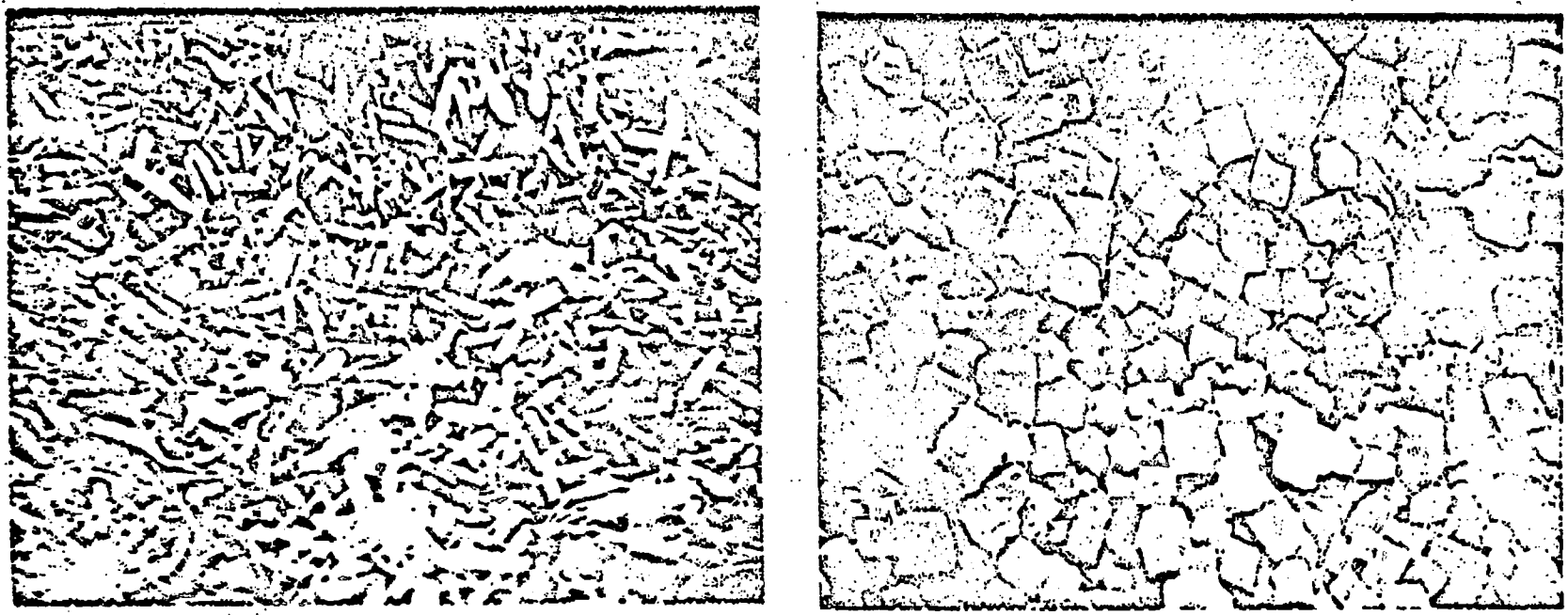

FIGURE 2. Undissolved Solids from Evaporated Simulated HLLW (scanning electron microscope at 5000X magnification).

References

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