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## SOLIDS CONTROL IN SLUDGE PRETREATMENT

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### INTRODUCTION

Sludge pretreatment will likely involve washing, followed by caustic or acidic leaching and washing of sludge residues after leaching. The principal goal of pretreatment is to obtain a low-volume high-activity waste stream and a high-volume low-activity waste stream. Also, some waste constituents such as chromium and phosphate can be included in glass formulations only at very low concentrations; therefore, it is desirable to remove them from high-level waste streams.

Two aspects of sludge treatment and subsequent separations should be well delineated and predictable: (1) the distribution of chemical species between aqueous solutions and solids and (2) potential problems due to chemical interactions that could result in process difficulties or safety concerns. Before any treatment technology is adopted, it must be demonstrated that the process can be carried out as planned. Three pretreatment methods were considered in the Tri-Party (Washington State Ecology, U.S. Environmental Protection Agency, and U.S. Department of Energy) negotiations: (1) sludge washing with corrosion-inhibiting water, (2) Enhanced Sludge Washing, and (3) acidic dissolution with separations processes. Enhanced Sludge Washing is the baseline process. In Enhanced Sludge Washing, sludge is first washed with corrosion-inhibiting water; it is then leached with caustic (sodium hydroxide solution) and washed again with corrosion-inhibiting water. The initial concern is whether a pretreatment technique is effective in separating sludge components. This can be evaluated by bench-scale tests with sludge specimens from underground storage tanks. The results give data on the distribution of important species such as aluminum, phosphate, and radionuclides between wash and leach solutions and solid sludge residues. In addition, it is very important in these processes to prevent formation of solids by uncontrolled precipitation (autoprecipitation). Failure to do so will result in the formation of crystalline solids and gels, which are unacceptable because they will (1) prevent mixing; (2) prevent pumping; (3) retard separations; (4) coat surfaces; and (5) clog pipes, equipment, and filters.

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## ENHANCED SLUDGE WASHING

### Enhanced Sludge Washing and the Bayer Process

It is very important to assess implementation of a pretreatment process. Failure to do so will result in process operating parameters and unit operations that are not well matched to chemical and physical changes that are actually occurring. This assessment requires an evaluation of the overall process, not simply the distribution of species between solids and liquids. Figure 1 is a simplified diagram of Enhanced Sludge Washing and the Bayer process. In the Bayer process, a bauxite slurry is leached with caustic to dissolve alumina from the ore. The goal is to produce alumina in the form of gibbsite,  $\text{Al}(\text{OH})_3$ . This process has been used for more than 100 years, and the continuous changes it has undergone as new technologies become available make it an excellent starting point for evaluating Enhanced Sludge Washing and for avoiding potential problems.

The Bayer process is similar to Enhanced Sludge Washing. First, the material being treated, either sludge or bauxite, is digested at some temperature ( $T_1$  in Figure 1) by a caustic solution. This is followed by a solid-liquid separation at a temperature  $T_2 \leq T_1$ . The solids are then washed, and another solid-liquid separation takes place. The liquid from digestion and washing is combined at a temperature  $T_3 \leq T_2$ . In the Bayer process, controlled precipitation of gibbsite occurs at this point. A number of techniques can be used to control the formation of solids. Temperatures can be maintained in the process to prevent solids formation that occurs when wash solutions or leachates that are at or near saturation are cooled. After digestion at temperature  $T_1$ , either of two things can happen if the temperature decreases, assuming that the leachate is at or near saturation in at least one species: (1) reprecipitation back to the sludge solids or (2) supersaturation of the leachate. Supersaturation is a metastable state that can result in reprecipitation elsewhere in the process. Thus, in either case, allowing the system to cool after digestion and before solid-liquid separation leads to an undesirable result. If the material reprecipitated is a major component such as alumina, the heating during digestion enhances only the rate that the saturated concentration referred to ambient temperature is reached, not the amount of material ultimately in solution.

In another option for controlling solid formation, excess caustic could be used to avoid exceeding solubility limits, even when the solutions are cooled or when leachates and wash solutions are mixed. In addition to these techniques, controlled precipitation, as in the Bayer process, may improve process control. Of course, in the Bayer process, controlled precipitation is used to obtain the gibbsite product. In Enhanced Sludge Washing, the combination of leachate and wash solutions occurs in a wash accumulation tank rather than in the controlled precipitation portion, as in the Bayer process. Clearly, if some of the three techniques for controlling solids formation in Enhanced Sludge Washing are not in place, precipitation is likely in the wash accumulation tank or in associated piping.

### Thermodynamic Calculations

Equilibrium calculations of alumina behavior in Enhanced Sludge Washing of Hanford S-104 and T-104 sludge were carried out. Details of the calculational method and thermochemical data are provided by Weber and Beahm.<sup>1</sup> For this series of calculations, the species set included  $\text{H}_2\text{O}$ ,  $\text{Na}^+$ ,  $\text{OH}^-$ ,  $\text{H}^+$ ,  $\text{Al}(\text{OH})_4^-$ ,  $\text{AlOOH}$ ,  $\text{Al}(\text{OH})_3$ , and  $\text{NaNO}_3$ .

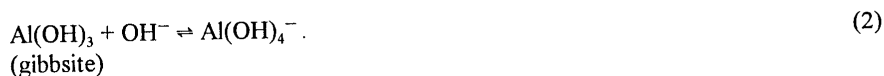
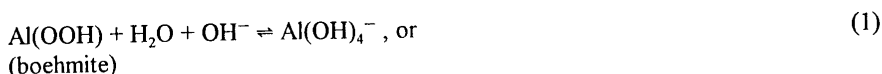
**Excess Caustic and Caustic Consumption.** The calculations on S-104 sludge are described first. The caustic solution was chosen so that the resulting solution at equilibrium was exactly 3 molal ( $m$ ) in hydroxide ( $\text{OH}^-$ ). The initial concentration required was  $3.1 < m < 3.8$ . In Figure 2 caustic leaching is represented as a function of liters of caustic per kilogram

**Figure 1.** Simplified diagram of Enhanced Sludge Washing and the Bayer process.

**Figure 2.** Calculation of S-104 caustic leaching at 75°C: sufficient caustic added so that mixture is 3 *m* OH<sup>-</sup>; ambient temperature, 25°C.

of aluminum (L/kg Al). The line labeled "Undissolved in Sludge" gives the percentage of undissolved aluminum as a function of (L/kg Al). The percentage of undissolved aluminum reaches zero at an (L/kg Al) of about 75. This is also the point at which the percentage of aluminum precipitated back to the leachate reaches its maximum. In addition, a relatively small amount of aluminum would reprecipitate back to the sludge residue. (Note: This curve is 10 times the calculated percentage.) At (L/kg Al) values  $\geq 75$ , the percentage of aluminum precipitated in the leachate or reprecipitated in residue decreases and becomes zero at  $\sim 200$ . If the sludge were leached at  $100^\circ\text{C}$  instead of  $75^\circ\text{C}$ , the percentage of aluminum "Undissolved in Sludge" would reach zero at an (L/kg Al) of  $\sim 48$  rather than  $\sim 75$ . This phenomenon results from the greater solubility of alumina at the higher temperature. However, the (L/kg Al) value at which no aluminum precipitated in the leachate or reprecipitated in the residue would be similar in both cases because the ambient temperature of  $25^\circ\text{C}$  was the same and the same amount of aluminum was involved in the process.

It should be emphasized that only a fraction of the caustic would be consumed in the reaction with alumina:



Most of the caustic indicated in the large (L/kg Al) values would be needed to maintain the  $\text{OH}^-$  concentration in saturated alumina solutions or would be the excess necessary to prevent precipitation when the temperature is lowered or when leachates and wash solutions are mixed.

Other materials in sludge, such as silicates and acid phosphates, can also consume caustic. The T-104 sludge contains a relatively high concentration of phosphorus and has a pH of  $\sim 10.5$ . At this pH the phosphate would be  $\text{HPO}_4^{-2}$ . In caustic leaching this would consume  $\text{OH}^-$  and convert to phosphate  $\text{PO}_4^{-3}$ :



This effect was seen experimentally in a test in which the final caustic concentration could be related to the amount consumed by both aluminum and acid phosphate. This is shown in the calculations represented by Figure 3. In these calculations the initial sodium hydroxide concentration was 3 molal ( $m$ ). The final caustic concentration was  $\sim 2.1 m$ , due to consumption of caustic by aluminum and acid phosphate. In Figure 3 the slope of the line "Undissolved in Sludge" is initially somewhat flat because the caustic is being consumed in the conversion of acid phosphate to phosphate. With the addition of more caustic, the percentage of aluminum undissolved in sludge decreases more rapidly. The (L/kg Al) value at the point at which the percentage precipitated in leachate or reprecipitated in sludge approaches zero is again  $\sim 200$ .

In the *Status Report: Pretreatment Chemistry Evaluation - Wash and Leach Factors for the Single-Shell Tank Waste Inventory*,<sup>2</sup> N. G. Colton gives the expected inventories of aluminum and sodium in Hanford single-shell tanks as 6,280 metric tons and 43,000 metric tons, respectively. If 200 (L/kg Al) is used as the estimated caustic-to-aluminum ratio needed to avoid precipitation in solutions or residues, then  $\sim 1 \times 10^9$  L of  $\sim 3 m$  sodium hydroxide is indicated. This would contain twice the amount of sodium presently in the single-shell tanks.

**Figure 3.** Calculation of T-104 caustic leaching:  $\text{OH}^-$ , initially 3 *m* at 75°C; ambient temperature, 25°C.

### Laboratory Studies

Enhanced Sludge Washing tests were performed on sludge from Hanford underground storage tanks T-104, S-104, C-105, C-107, C-108, SX-113, B-202, and C-104 to evaluate the formation of solids in leachates and wash solutions and the treatability of these materials by caustic processing. The test sequences were similar to the process shown in Figure 1. Thus, the sludge samples, 2–10 g, were leached at temperature  $T_1$ , and the solid-liquid separation was carried out at  $T_2$ . The sludge solids were then washed, and another solid-liquid separation was performed. After the test the leachates and wash solutions were examined for solid formation and chemical composition. The samples were filtered through 0.45- $\mu\text{m}$  Teflon syringe filters. In the tests in which filtration was done at a temperature above ambient, the filter and syringe were preheated. The tests were performed in high-density polyethylene containers, and leachates and wash solutions were placed in clear polystyrene tubes for observation.

These tests demonstrated that solid formation in process solutions takes a variety of forms: very fine particles, larger particulate solids, solids floating in solution like egg whites, gels, crystals, and coatings on sample containers. Solid formation has been seen in wash solutions before and after leaching as well as in leachates. Colloids appeared in the second wash (0.01 *M* NaOH + 0.01 *M* NaNO<sub>2</sub>) before caustic leaching of Hanford T-104 sludge (30 wt % solids). The sludge was washed twice (4 g wash solution/g of initial sludge) at ambient temperature. No solids were seen in the first wash solution. The ionic strength of the first wash solution was 0.45, and the ionic strength of the second wash solution was 0.1. In this same test sequence, a gel formed in the second wash after leaching with 3 *M* NaOH. The ionic strength of the second wash after leaching was 0.07.

Two test sequences were conducted on sludge from T-104 under conditions that were identical except for the temperature. The treatments started with a wash with inhibited water ( $0.01\text{ M NaOH} + 0.01\text{ M NaNO}_2$ ). After the liquid was centrifuged and decanted, the sludge was leached with  $3\text{ M NaOH}$  and centrifuged; the leachate was then decanted. The sludge was next leached with  $3\text{ M NaOH}$ , centrifuged, and decanted. The treatment concluded with three washes with inhibited water, with centrifugation and decantation after each wash. The initial wash and the two leaches were carried out at  $60^\circ\text{C}$  in one test sequence and at  $95^\circ\text{C}$  in the other test sequence. The centrifugation, filtration, decantation, and the three washes after leaching were performed at ambient temperature.

One day after leaching, gels were observed in both the first and second leachates from both test sequences. A sample of the gel that formed in this sequence was examined by X-ray diffraction and scanning electron microscopy. The X-ray diffraction pattern identified the material as natrophosphate,  $\text{Na}_7(\text{PO}_4)_2\text{F}\cdot 19\text{H}_2\text{O}$ . When this substance was wet, it looked like a gel; it was soft and stuck to the sample container walls. When it was dried, it appeared as a white mass. Chemical analysis of the gel gave the relative mole values as 7 for sodium, 2.2 for phosphate, and 0.93 for fluoride and indicated a trace of potassium and sulfate. This is close to the ratios from X-ray diffraction. It is noteworthy that there was very little aluminum in the gel. The sodium-to-aluminum ratio was  $4 \times 10^4$ , even though the leachate in contact with the gel had an aluminum concentration of  $0.2\text{ M}$ .

Two test sequences were carried out on sludge from Hanford underground storage tank T-104 to evaluate the behavior of phosphate. In one test sequence, the temperature was  $75^\circ\text{C}$  throughout all steps of the process. In the other test sequence, the  $75^\circ\text{C}$  temperature was maintained only during leaching; after that time, the process was carried out at room temperature. The two processes were carried out in parallel to ensure that temperature was the only variable.

A marked difference in the behavior of phosphate was observed in the two tests. When the leachate was maintained at  $75^\circ\text{C}$  during settling and filtration, the amount of phosphate in the leach solution was approximately 10 times greater than when the leaching was performed at  $75^\circ\text{C}$  and the settling and filtration were conducted at room temperature. This is direct evidence that the phosphate reprecipitated into the sludge residue-leachate mixture when the temperature was reduced to ambient. Because of this reprecipitation more phosphate was observed in this sludge residue after leaching than in the case where temperature was maintained at  $75^\circ\text{C}$ . This resulted in higher phosphate concentrations in the wash solution processes at ambient temperature than in those at  $75^\circ\text{C}$ . In the test sequence maintained at  $75^\circ\text{C}$ , most of the phosphate remained in the leachate and was not present in the sludge residue at the time of washing.

These results have several implications. First, the elevated temperature of the leachate enhanced the solubility of the phosphate. Second, running the process with leaching at an elevated temperature and the rest of the process at ambient could possibly increase the rate of dissolution of phosphate, but reprecipitation would return the concentration to that consistent with room temperature. The reprecipitated phosphate is rather gelatinous and sticks to container walls. Third, if the leaching temperature is not maintained throughout the process, phosphate solids will form whenever it is lowered.

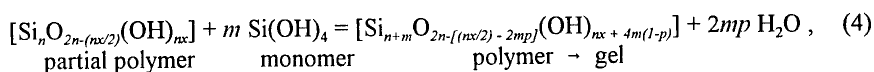
Enhanced Sludge Washing tests have been conducted with sludge from Hanford tanks C-105, C-107, C-108, SX-113, B-202, and C-104, and the filtered process solutions were examined for solid formation. The C-108 and SX-113 sludge were separately leached with  $3\text{ M NaOH}$  and washed three times with inhibited water. The temperature throughout the leaching, settling, washing, and filtration was  $75^\circ\text{C}$ . The ratio of leachate or wash solution to sludge weight was 10, based on the original mass of sludge. The C-105 and C-107 sludges were separately leached at  $70^\circ\text{C}$  in  $6\text{ M NaOH}$  as part of the Sludge Partitioning Chemistry program managed by B. Z. Egan. After leaching, the sludge residue was washed three times

at ambient temperature. In the tests with C-108 and SX-113 sludge, the process solutions were first examined for solids ~30 min after filtration. The tests with the material from C-105 and C-107 were conducted in a hot cell, and a 2-day delay occurred before the first examination could be carried out.

Solids formed in filtered process solutions from each of these sludges. With C-105 and C-107, a clear gel-like material formed in the leachates. Wash solutions from the C-105 and C-107 tests also had some clear masses. The leachate from the C-108 test developed a mass of gel-like material as well as some material that appeared to be more crystalline. The wash solutions from the C-108 test developed a small amount of a filmy fibrous material. The filtered leachate from the SX-113 test produced a significant amount of particulate material, which appeared to be semigelatinous when suspended. Scanning electron microscopy with energy-dispersive X-ray analysis showed that these particles contained sodium and silicon. This could be any of a host of sodium silicates in the NaOH·SiO<sub>2</sub>·H<sub>2</sub>O system. The test with sludge from B-202 also produced particulate material. This material contained bismuth, a principal component of this sludge.

## ACID TREATMENT

Sludge solids contain silica, aluminum, and a variety of metal ions that may participate in gels if they enter solution during processing. At pH levels ≤7, silica in solution as silicic acid, Si(OH)<sub>4</sub>, is stable for long periods of time if the concentration is ≤100 ppm. At greater concentrations, silicic acid polymerizes according to the general equation,<sup>3</sup>



where  $n$  = the number of silicon atoms in a polysilicic acid molecule or particle or polymeric network,  $x$  = the number of OH groups per silicon atom in the polymer (not exceeding 4),  $m$  = the number of monomeric silicic acid molecules added to the polymer, and  $p$  = the fraction of the hydroxyl groups per monomeric silicic acid molecule that is converted to water during the polymerization reaction. Polymerization of Si(OH)<sub>4</sub> leads to particles or to the formation of gels.

Acidic treatment of sludge was carried out with material from Hanford underground storage tank B-202. This material was selected for testing because it contains a relatively high fraction of transition metals in the sludge solids and because Enhanced Sludge Washing tests conducted at Pacific Northwest National Laboratory and at Los Alamos National Laboratory showed that only a small fraction of aluminum dissolved in caustic treatment. Two test sequences were run with the B-202 sludge. In both sequences the sludge was leached successively with 1 M, 3 M, and 6 M HNO<sub>3</sub> and then washed with water. In one case, the entire sequence was carried out at 75°C; in the other case, it was performed at ambient temperature. This test plan enabled an evaluation of both acid concentration and temperature. Before treatment the sludge looked like a dark shoe polish. The ratio of leachate or wash to sludge weight was 10, based on the original weight of the sludge.

In the sequence performed at 75°C, sludge solids remained after leaching with 1 M and 3 M HNO<sub>3</sub>. However, the sludge was completely dissolved by the 6 M HNO<sub>3</sub> step in the sequence. This was not the case in the sequence at ambient temperature. In that case sludge solids remained at all times. Thus, the temperature difference between 75°C and ambient was the important factor in the complete dissolution of this material.

The leachates and wash solutions were all filtered through 0.45-μm Teflon syringe filters. Solid formation was observed in all of the leachates. This varied from halo-like deposits on the

glass sample tubes to gels and particulates. Analyses of the solids revealed that a gel material contained silica with some potassium, calcium, iron, and manganese. Two phases were embedded in the gel. One was barium sulfate. The other could not be identified, but it was determined that the only metal it contained was bismuth. In addition, particles of a reddish precipitate separate from the gel were found to contain bismuth and chromium as the only metals.

Hydrofluoric acid can be used with nitric acid to avoid the formation of silica gel and to enhance the dissolution of siliceous materials. Hydrofluoric acid will convert silica to the  $\text{SiF}_6^{-2}$  ion, which is soluble. This requires six fluoride ions for each silicon. A series of tests was run with sludge from Hanford underground storage tank SX-113. This sludge has a silicon concentration of 0.2 g per gram of sludge. Three samples of SX-113 sludge were leached at 75°C. After leaching, the leachate was filtered through 0.45- $\mu\text{m}$  syringe filters at 75°C. Sample A was leached with 3 M nitric acid, sample B was leached with 3 M nitric acid plus 3 M hydrofluoric acid, and sample C was leached with 3 M nitric acid plus 6 M hydrofluoric acid. The leachate-to-sludge ratio was 10 mL per gram in each case. In sample A, the fluoride-to-silicon ratio was zero; in sample B, it was 4; and in sample C, it was 8. Leaching the SX-113 sludge with 3 M nitric acid alone (sample A) was not effective in dissolving sludge solids. A mass of solids remained after treatment.

In sample B, which had a fluoride-to-silicon ratio of 4, a porous silica deposit formed in the airspace above the liquid during leaching. It is likely that this vapor-assisted deposition of silica occurs when the fluoride in solution is consumed by reaction with silica in the sludge. This would result in conversion of some of the  $\text{SiF}_6^{-2}$  to silicon tetrafluoride,  $\text{SiF}_4$ , which is a gas. This gas would decompose to silica and hydrofluoric acid in a cooler region.

The test with sample C, which had a fluoride-to-silicon ratio of 8, resulted in essentially complete dissolution of the sludge without transport of silica from the sample.

This series of tests revealed the important features associated with the use of hydrofluoric acid in acidic sludge treatment. First, hydrofluoric acid enhanced the dissolution of sludge solids. Second, it is necessary to have a fluoride-to-silicon ratio greater than 6 to prevent silica deposition in areas such as tank domes, ventilation systems, and sample lines. This excess of fluoride must be preserved in all parts of the system to avoid local regions of low fluoride content.

## DISCUSSION

### Enhanced Sludge Washing

Solid formation in filtered leachates and wash solutions was seen in seven of the eight sludges treated. The gel material was identified as natrophosphate,  $\text{Na}_7(\text{PO}_4)_2\text{F}\cdot 19\text{H}_2\text{O}$ . Sludge from tank S-104 was the only material that did not produce solids. This is a redox sludge that contains alumina in the form of boehmite.

The composition of phosphate fluorides can vary by substitution of hydroxide,  $\text{OH}^-$ , for fluoride and vice versa. The substitution of F for OH in apatite,  $\text{Ca}_5(\text{PO}_4)_3\text{OH}$ , is the basis for fluoride toothpaste. The substitution of OH for F in natrophosphate,  $\text{Na}_7(\text{PO}_4)_2(\text{F})_x(\text{OH})_{1-x}$ , would extend the range of conditions at which solids can form.

Phosphate and fluoride concentrations in solution can be reduced by the addition of lime,  $\text{CaO}$ , to form calcium phosphates and calcium fluoride. This addition would be a means of avoiding gelation, but it could also introduce problems in Enhanced Sludge Washing. If lime is added during retrieval, gelation would likely be prevented throughout processing, but little or no phosphate removal from sludge solids would occur. Also, experience in Enhanced Sludge Washing of Oak Ridge Melton Valley Storage Tank sludge, which contains high

calcium concentrations, showed that little aluminum was dissolved. This was most likely due to the formation of calcium aluminum hydrogarnets. Thus, lime treatment would have to occur after leaching, settling, and decantation. It is also possible that lime could assist in controlled precipitation of alumina.

Alumina reprecipitation can be controlled by temperature, by excess caustic, or by controlled precipitation. At this time the options for controlling alumina are better defined than those involving phosphate solids. Combinations of caustic and temperature that avoid alumina reprecipitation can be delineated. However, the close temperature control and excess caustic may make this approach impractical, and controlled precipitation may be the most viable option. Of course, any methodology for dealing with alumina reprecipitation will have to be compatible with mitigating formation of phosphate gels and sodium silicate precipitates.

### Acid Treatment

The concentration of silica in leachates and wash solutions must be kept quite low, <100 ppm, to prevent the formation of silica gel. This could be done in two ways. The liquid-to-sludge ratio in acid treatment could be maintained at a level that ensures that only very dilute silica solutions would occur. Alternatively, a combination of nitric acid and hydrofluoric acid could result in silicon in solution as  $\text{SiF}_6^{-2}$ , rather than as  $\text{SiO}_2$ .

A combination of caustic leaching followed by acidic leaching could reduce the amount of fluoride necessary to maintain a fluoride-to-silicon ratio greater than 6 by dissolving some of the siliceous material in the caustic. However, the caustic leaching steps of such a process would have to be done with the same control of temperature and concentration and amount of caustic, and possibly with controlled precipitation, as in Enhanced Sludge Washing, to prevent solid formation.

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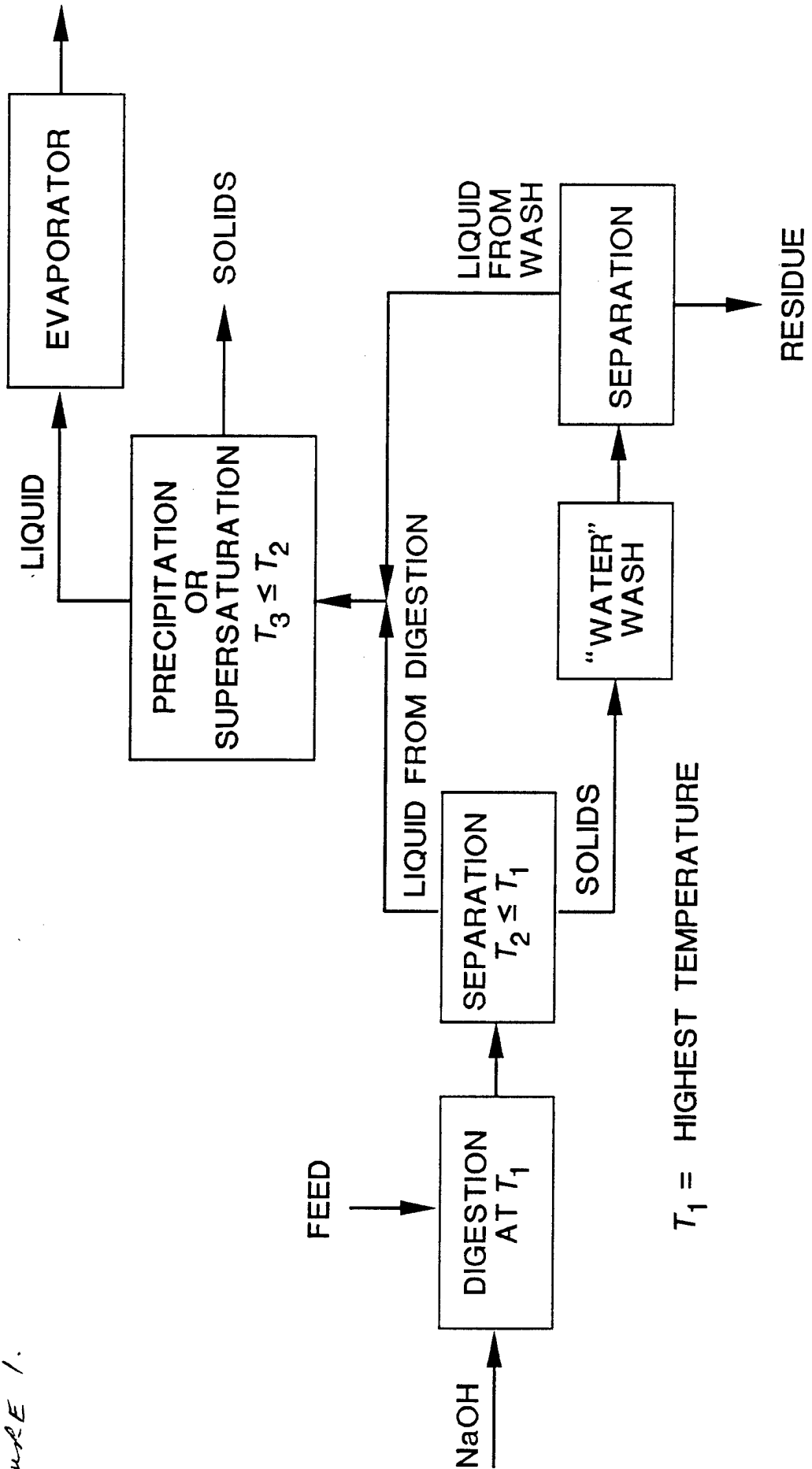
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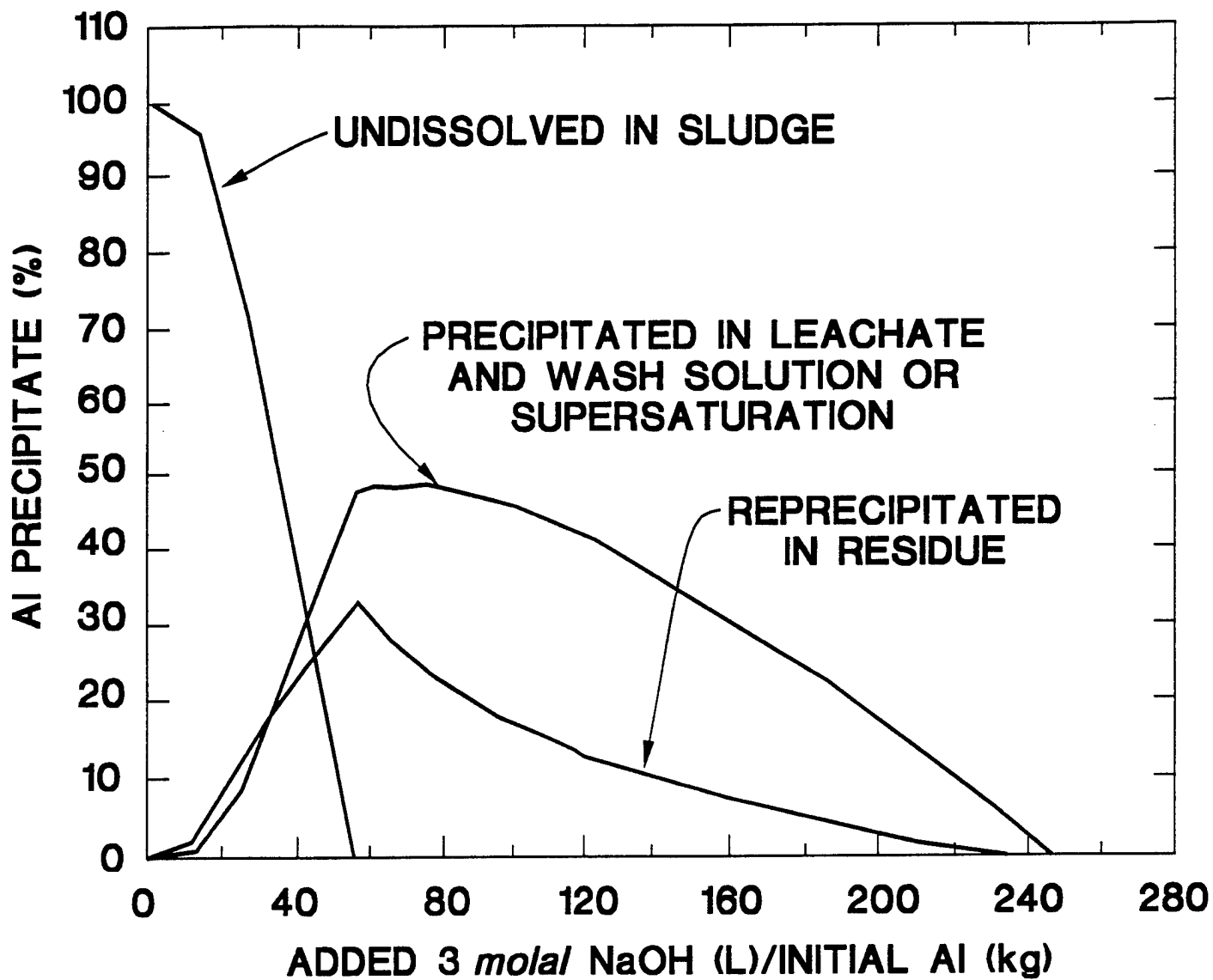
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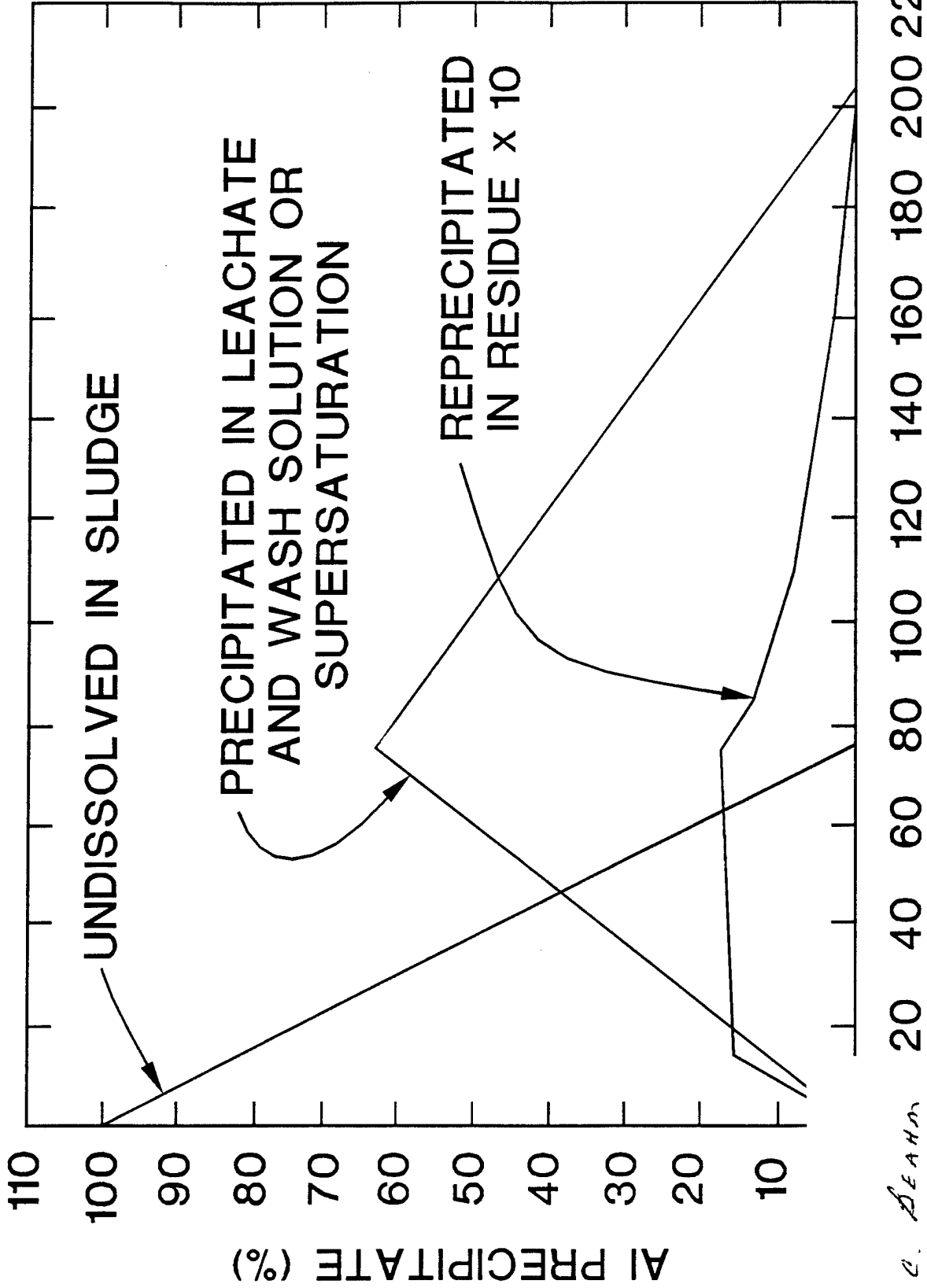
FIGURE 1.





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FIGURE 3.



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