Technical Letter Report on Evaluation of Long-term Aluminum Solubility in Borated Water Following a LOCA

Executive Summary

Long-term aluminum hydroxide [Al(OH)$_3$*] solubility tests were conducted in solutions containing 2500 parts per million (ppm) boron (B), and an aluminum concentration ranging from 40-98 ppm using aluminum nitrate or sodium aluminate as the aluminum (Al) source. The solution pH values were adjusted to achieve target pH ranging from 7.0 to 8.5. The solution temperature was cycled to obtain a temperature history more representative of Emergency Core Cooling System (ECCS) temperatures during operation in the recirculation mode after a loss-of-coolant accident (LOCA) in a pressurized water reactor (PWR). The observed Al solubility as a function of temperature and pH was close to predicted results for amorphous precipitates in a borated environment, which are higher than the solubility expected for crystalline forms of aluminum hydroxide. Precipitates were observed to form either as fine, cloudy suspensions, which showed very little tendency to settle, or under certain conditions, as flocculated precipitates, which were formed at the inner surface of the test flasks. The flocculated precipitates have an average diameter of 4-6 μm. Based on prior Argonne National Laboratory (ANL) head loss tests with surrogates[1], they would be expected to cause significant increases in head loss in glass fiber beds. Very fine precipitates associated with the cloudy solutions were less effective at causing head loss in the ANL vertical loop with a fiberglass-only bed, but could cause a different head loss response given different bed conditions, e.g., in cases where pore sizes are smaller than in fiberglass-only beds. The flocculation tendency of the precipitates can be qualitatively explained in terms of ionic strength or solution pH based on a colloidal stability theory referred as the Derjaguin, Landau, Verwey and Overbeek (DLVO) theory. The effect of solution pH is related to the zeta potential change on the particle surface according to this theory, and the effect of the total Al concentration is related the resulting increased particle concentration, which tends to increase flocculation.

Westinghouse report WCAP-16785-NP[2] also provides 30-day solubility data at 140 and 200°F for Al(OH)$_3$. The Westinghouse report estimated that Al hydroxide solubility at 140 and 200°F is 40 and 98 ppm, respectively, for pH=8.0. The Westinghouse tests considered limited temperature perturbations, but based on the results in this report from tests with increased thermal cycling, these limits still appear to be reasonable. WCAP-16785-NP also states that perturbations in pH down to 7.0 did not affect these results. However, those tests in which the pH was decreased to 7.0 appear to be very short term and were only performed at 200°F with 40 ppm Al. The current results suggest that the solubility limit at pH =7.0 is less than 40 ppm, even at 200°F.

Background

The Nuclear Regulatory Commission (NRC) requested technical assistance from ANL to investigate certain aspects of GSI-191 chemical effects in order to support the NRC staff review of licensee supplemental responses to Generic Letter 2004-02. A previous technical letter report[1] provided results from ANL head loss testing of chemical precipitates used in industry sump strainer head loss tests. The objective of the current work was to assess solubility of aluminum in borated water for a typical plant ECCS mission time, including the Al concentrations reported in WCAP-16785-NP. The solubility tests were performed with thermal cycling and with fiberglass insulation in the test flasks. The thermal cycling was not intended to exactly match the thermal cycles experienced in the ECCS during recirculation but to

---

*In this report, Al(OH)$_3$ is used a short–hand for a family of aluminum oxyhydroxides that could form in a variety of crystalline and amorphous forms.

evaluate the effect on solubility of thermal cycling superposed on a reasonably representative long-term cooling profile.

Test Procedures

The long-term aluminum solubility tests were conducted in 250-mL flasks without agitation. The test solutions contained 2500 ppm B and Al ranging from 40-98 ppm using aluminum nitrate or sodium aluminate as the Al source. The solution pH was adjusted to achieve target pH values ranging from 7.0 to 8.5. The solution temperature history was varied to obtain a time-temperature history reasonably representative of that seen by the reactor coolant in the case of LOCA. Thermal cycling of the solution was performed to simulate the transit of the coolant through the RHR heat exchanger (cool down) and reactor core (heat up). A representative history of the temperature is shown in Figure 1.

Figure 2 shows the long-term Al solubility test apparatus setup. In Figure 2, each combination of condenser, flask, beaker, and heater represents an independent long-term solubility test. The 2500 ppm boron test solution is prepared by adding 3.572 g boric acid [H_3BO_3, Fisher Scientific] to about 200 mL of deionized water. The solution pH is then adjusted using sodium hydroxide [NaOH, Fisher Scientific] to a target pH ranging from 7.0 to 8.5, and the final solution volume is adjusted to 250 mL. The final solution pH and conductivity are measured and recorded. The 250-mL pH-adjusted borated solution is transferred to a designated Pyrex flask. A condenser is mounted on each flask to minimize the water loss by evaporation at high temperature. A thermocouple is mounted on the flask. The flask is submerged in a 2000-mL Pyrex beaker filled with deionized water. The beaker is placed on an electric heater. As shown in Figure 2, the open top of the 2000-mL beaker is covered with plastic transparent film and aluminum thin foil to minimize evaporative loss of water from the beaker. The solution temperature is raised up to 200°F. Once the solution temperature reaches 200°F, sufficient concentrated aluminum nitrate or sodium aluminate solution is added to obtain a target Al concentration ranging from 40 to 98 ppm. For example, a concentrated 10,000 ppm Al solution would be prepared by adding 3.038 g of sodium aluminate powder [$NaAlO_2$, Alfa],to 100 mL high purity water. No precipitate should be present in this solution. To obtain a 98 ppm Al concentration test solution, 2.45 mL of the concentrated Al solution is added into the test flask. Twenty-four mg of NUKON glass fiber is added to maintain the same mass ratio between NUKON and total test solution as in the ANL loop tests. Final solution pH at 200°F is measured. The solution temperature is then cycled following a target schedule such as that shown in Figure 1. To achieve faster cooling, hot water is drained from the 2000-mL beaker and chilled water (2-3ºC) is added. As soon as the solution temperature reaches the target temperature, the chilled water is drained, hot water is added, and the electric heater is turned on again. The actual solution temperature variations during heating and cooling cycles are shown in Figure 3. Cooling to the target temperature was achieved within 10 minutes; bringing the temperature back to the original value took more time than cooling.

The solution is observed daily to determine if any precipitate has formed in the solution. After 30 days, pH and conductivity are measured and solution samples are taken for chemical composition analysis by Inductively Coupled Plasma/Optical Emission Spectroscopy (ICP/OES). The particle size distributions of the post-test solution were also measured using a laser granulometry technique (CILAS 1064 laser particle size analyzer). The minimum particle size that can be recognized by visual observation is around 100 nm based on previous and current bench top test experience.
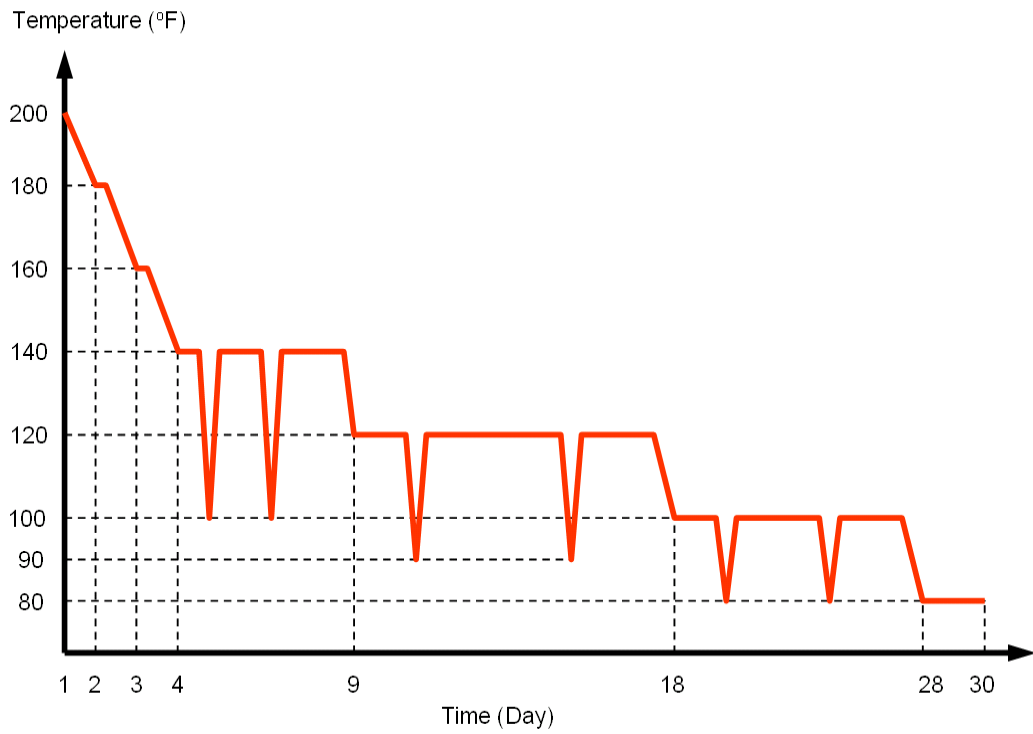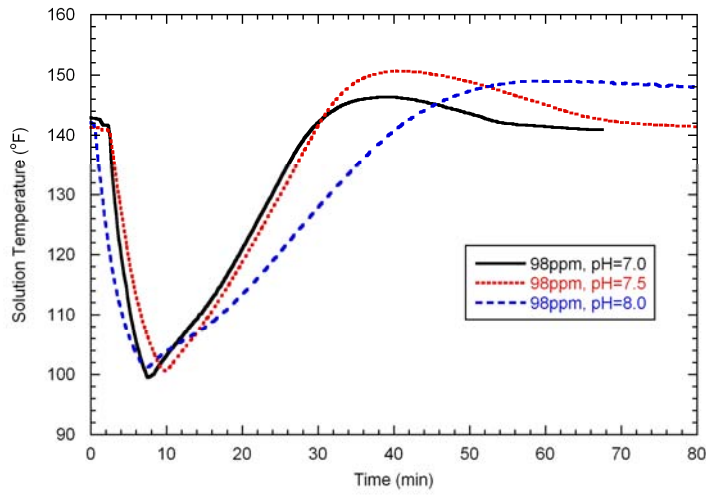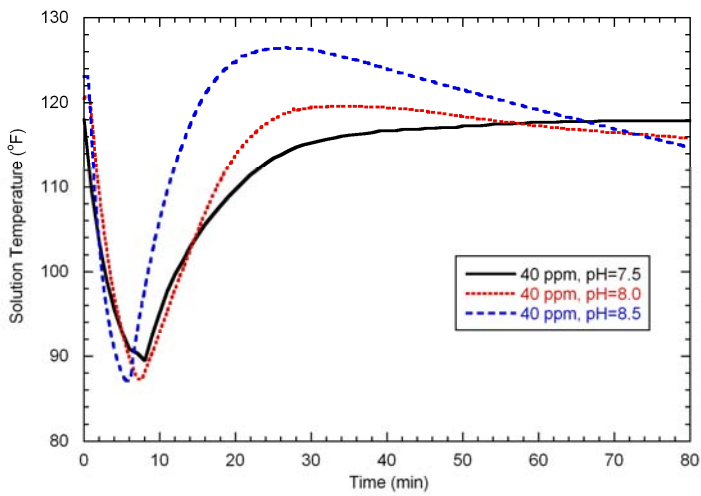
3

Figure 1.   Typical solution temperature history for the long-term 30 day Al solubility tests.
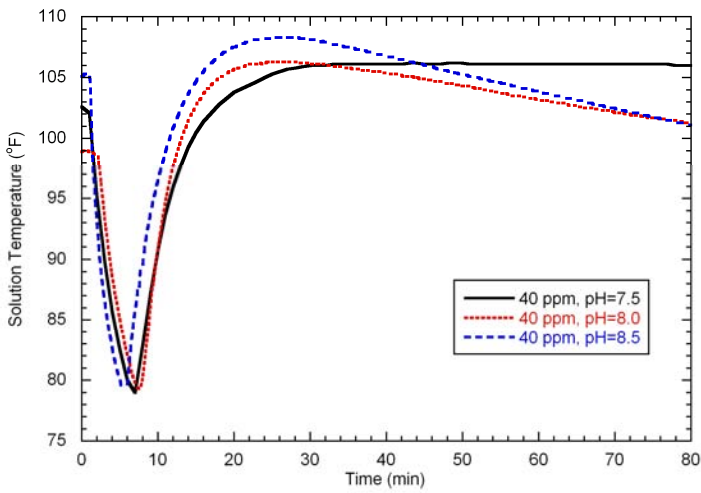

Figure 2.
The long-term Al
solubility test facility
setup.

4

(a) 140 to 100 F thermal cycling

(b) 120 to 90 F thermal cycling

(c) 100 to 80 F thermal cycling

Figure 3.   Examples of temperature history during thermal cycling for (a) 140 to 100 F, (b) 120 to 90 F, and (c) 100 to 80 F.

5

Test Results

Aluminum Nitrate Tests

Initially aluminum nitrate powder [Al(NO$_3$)-9H$_2$O, Alfa, 99.9%] was used as the Al source. Adding 40 ppm Al as concentrated aluminum nitrate solution caused the pH decrease of 0.2-0.3 in the initial pH before adding Al solution. However, adding 98 ppm Al caused much larger pH decrease: around 0.5 for pH=8.0 and 7.5 solutions and 3.3 for pH=7.0 solution. It appears that nitrate ions from 98 ppm Al solution cannot be adequately buffered with 2500 ppm B solution.♣ Because the pH control was inadequate, the bench top tests were terminated before the 30-day test schedule. Actual daily test observation results are summarized in Table A1-A3 for 98 ppm Al tests and Table A4-A6 for 40 ppm Al tests. The Westinghouse report WCAP-16785-NP also noted that adequate control of pH could not be maintained using aluminum nitrate.[2] Their subsequent tests used sodium aluminate as the Al source as was also done in the ANL tests. Since nitrate ions are not likely to be present in recirculating water of the ECCS, using sodium aluminate can exclude any non-representative effects that might be caused by nitrate ions.

Sodium Aluminate Tests

Figure A1 in Appendix A shows the long-term solubility test schedule in calendar form. Four different Al concentrations, 98, 70, 55, and 40 ppm, were explored over a pH range of 7.0-8.5. The results of the 30 day tests are summarized in Table 1. The daily test observation results are given in Table A7–A9 for 98 ppm, Table A13–A14 for 70 ppm, Table A15–A16 for 55 ppm, and Table A10–A12 for 40 ppm.

Figures 4-7 show long-term Al solubility test results as a function of temperature for 98, 70, 55, and 40 ppm Al solutions, respectively. In the figures, an open circle means no precipitate was observed during the observation time period at the conditions of interest and a solid circle indicates precipitates were formed during the observation time period regardless of whether the precipitates became flocculated or remained as a very fine suspension. In Figure 4, the solid curve represents the predicted contour line for an Al solubility of 98 ppm in the pH-temperature domain. The curve is based on a solubility equation for amorphous Al(OH)$_4^-$ as a function of temperature and pH taken from a previous ANL technical letter report.[3] In that report, a value for the solubility constant log K of -12.2 at 25 °C was estimated from literature and test data. An activation energy for the solubility constant was estimated from the work of Benezeth et al.[4] on crystalline boehmite (γ-AlOOH, aluminum oxyhydroxide or Al$_2$O$_3$-H$_2$O, alumina monohydrate)and assumed to be applicable to amorphous Al(OH)$_3$. The dashed lines represent predictions of contour lines for Al solubilities of 98, 40, and 10 ppm in the pH-temperature domain based directly on Benezth's data for the crystalline forms. Comparison with the test results following the prototypic temperature variation shown in Figure 1, shows the prediction based on the ANL technical letter report expression for K is more consistent with the observed results than the predictions based on Benezeth's results. This is not unexpected since Benezeth's experimental data are for crystalline boehmite (γ-AlOOH, aluminum oxyhydroxide or Al$_2$O$_3$-H$_2$O, alumina monohydrate) in an environment with no boron. Under the current experimental conditions the precipitates are expected to be amorphous aluminum hydroxide, which has a higher solubility than the crystalline forms. It should be noted that the

---

♣ Al(NO$_3$)$_3$ solutions are acidic because it is the salt of a weak base and a strong acid. Boric acid solutions can buffer against strong base additions as most of the boron is present as neutral boric acid. However it has very little acid buffering capacity.

dwell time above 140°F at each temperature was limited, because the test temperature profiles, however, were intended to be reasonably representative of post-LOCA conditions in a PWR. The precipitation of Al hydroxide from the supersaturated state typically has an incubation time of less than an hour at 50°C, while the phase transformation from amorphous or pseudo-boehmite to thermodynamically more stable form, bayerite occurs over a few weeks[5]. Thus, if the solutions were held at a higher temperature for longer times before cooling, we might obtain solubility results closer to Benezeth's results.

Westinghouse estimated that the 30-day Al solubilities in pH=8.0 borated solution at 200 and 140°F are 98 and 40 ppm, respectively. The estimated Al solubility at pH=8.0 and 200°F from the current tests appears to be higher than 98 ppm, as shown in Figure 4. The estimated Al solubility at pH=8.0 and 140°F appears to be between 98 and 40 ppm, as shown in Figures 4-7. WCAP-16785-NP also states that perturbations in pH down to 7.0 did not affect these results. However, those tests in which the pH was decreased to 7.0 appear to be very short term and were only performed at 200°F with 40 ppm Al. Even at 200°F, the current results suggest that the solubility limit at pH =7.0 is less than 40 ppm.

Table 1. Indication of precipitate formation as either a cloudy suspension or a flocculate in test solutions upon cooling solutions from 200 to 80 °F.

Total Al Concentration (ppm)	Initial Solution pH at Room Temperature Before Adding Al Solution			
	7.0	7.5	8.0	8.5
98	●[a]	●	△[b]	ND[c]
70	●	●	ND	ND
55	●	△	ND	ND
40	ND	△	△	△[d]

[a] Solid symbols indicate precipitates were flocculated.
[b] Open symbols indicate precipitate was a cloudy suspension and not flocculated.
[c] ND indicates no test data are available.
[d] Solution became cloudy after cooling down from 100 to 80 F.

Figure 4.
Long-term aluminum solubility test results for 98 ppm Al solution as a function of solution temperature compared with predicted Al solubility data (solid line: previous ANL's technical letter report[3], dashed lines: Benezeth's data).

Figure 5.
Long-term aluminum solubility test results for 70 ppm Al solution as a function of solution temperature compared with predicted Al solubility data (solid line: previous ANL's technical letter report[3], dashed lines: Benezeth's data).

8

Figure 6.
Long-term aluminum solubility test results for 55 ppm Al solution as a function of solution temperature compared with predicted Al solubility data (solid line: previous ANL's technical letter report[3], dashed lines: Benezeth's data).
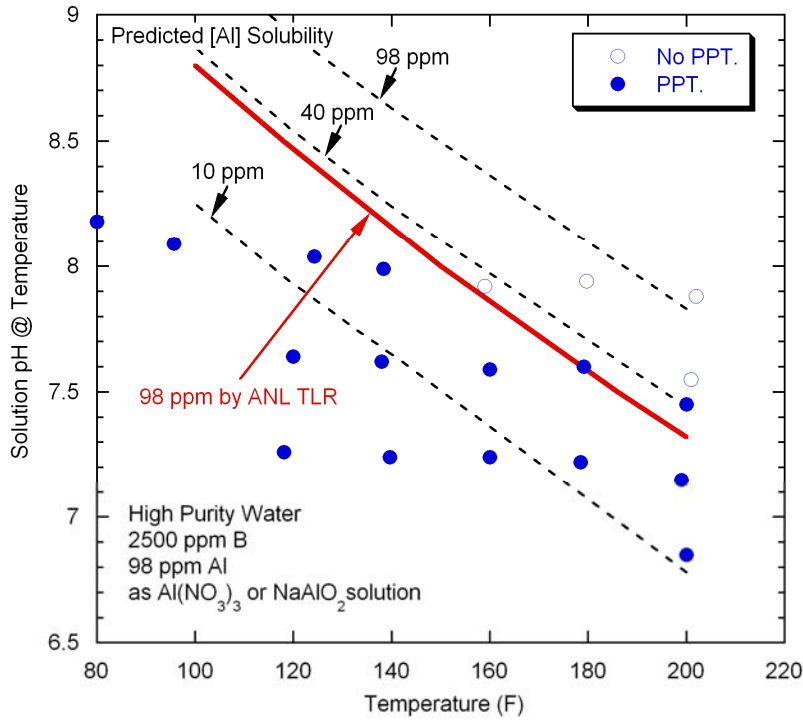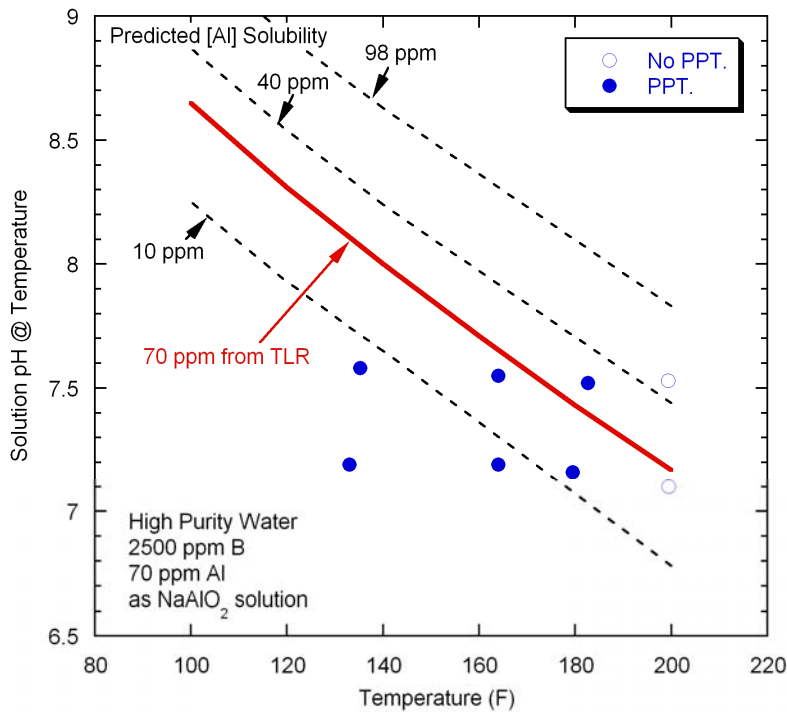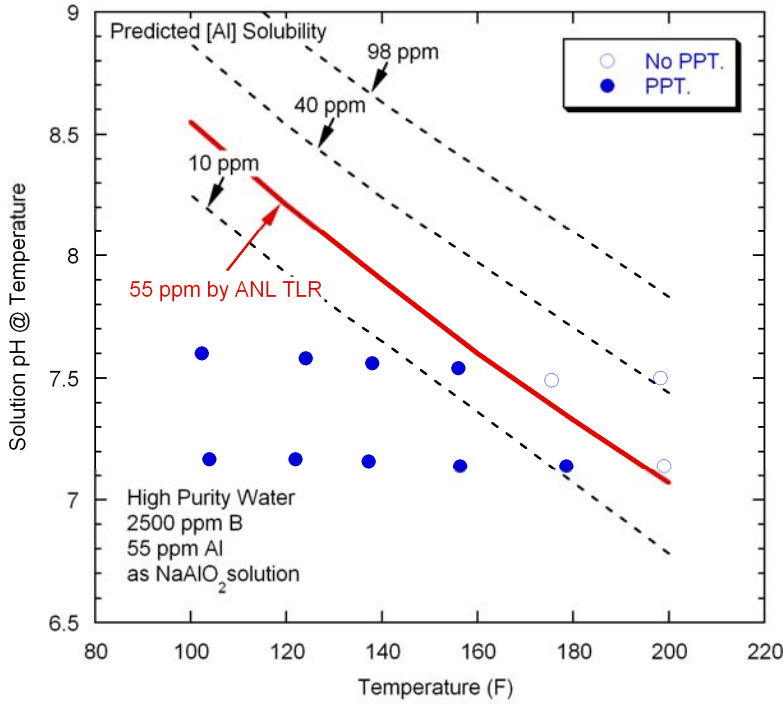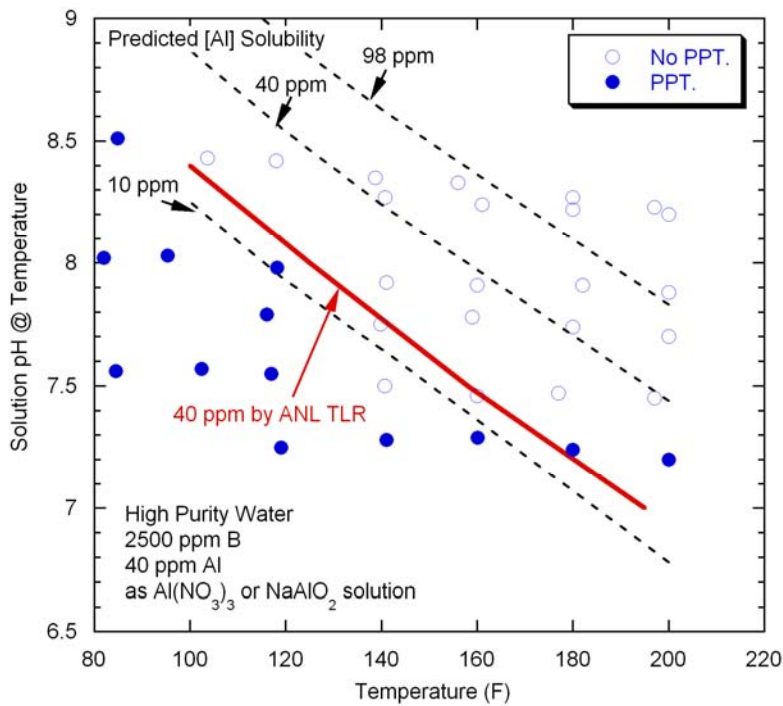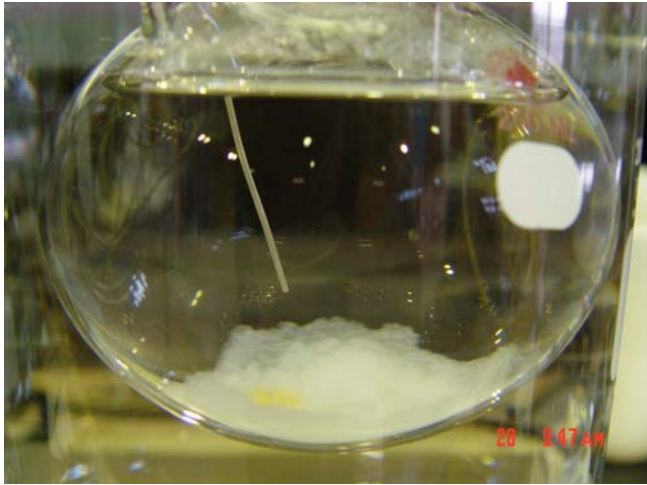

Figure 7.
Long-term aluminum solubility test results for 40 ppm Al solution as a function of solution temperature compared with predicted Al solubility data (solid line: previous ANL's technical letter report[3], dashed lines: Benezeth's data).

9

Two different types of precipitates were observed. In some cases, flocculated, cottony precipitates were formed on inner surface of the flask and on the surfaces of the thermocouple and glass fibers, and slid down and accumulated at the bottom of the flask; in other cases, the precipitates formed as a very fine suspension with a slightly milky appearance and settled extremely slowly. It should be noted that these tests were static and that shear stresses from flow could affect the flocculation tendency and settling behavior. Figure 8 shows photographs of the flasks after 30 days containing flocculated or very fine precipitates with 98 ppm Al contrasting the two different precipitates characteristics. The small yellow ball at the bottom of the flask is the 24-mg cluster of NUKON fibers. Figure 9 shows additional photographs of the flasks contrasting the different behaviors. The 40 ppm Al test solution shown in Figure 9d is slightly cloudy at 120°F. In the cases where a milky solution formed at a higher temperature, simply lowering the temperature was not sufficient to cause flocculation, although an increase in cloudiness could be observed. There was no case where the test solution kept clear until cooled down to 80 F. When the solution was clear at higher temperature, any sign of preferential precipitation on the glass fibers was not notified with naked eyes.

At constant Al concentration, the flocculation tendency of precipitates is dependent on the solution pH as shown in Table 1. This behavior may be explained by the variation in zeta potential with the solution pH. Table 1 also shows that at constant solution pH, the flocculation tendency of Al hydroxide precipitates is dependent on total Al concentration. A possible mechanism explaining both of these effects on flocculation tendency is discussed in a later section.

The particle size distributions of the post-test solutions were measured using a laser granulometry technique (CILAS 1064 laser particle size analyzer). Figure 10 shows the particle size distributions of 98 ppm Al test solutions. Figure 10a shows the effect of the solution pH. As shown in Figures 8a and 8b, the pH=7.0 test solution appeared to have slightly larger volume of precipitates than the pH=7.5 test solution, but the average particle size of pH=7.0 solution is slightly smaller than pH=7.5 solution. Since the maximum particle size of pH=7.0 is larger than that of pH=7.5 and flocculated precipitates are easily breakable as shown in Figure 10b, it can't be concluded that lower pH solutions have smaller Al hydroxide particle sizes. It is appropriate to conclude that Al hydroxide precipitates are flocculated at pH=7 and 7.5 with 98 ppm Al concentration and the particle size distribution measurements clearly indicate that there is no significant difference in these size distributions. The overall shape of size distributions is very similar to that of Al hydroxide precipitates reported in an earlier ANL study[1]. In that report, the average particle size of flocculated Al hydroxides in borated tap water was observed to be 7 μm.

(a) pH=7.0 with 98 ppm Al

(b) pH=7.5 with 98 ppm Al
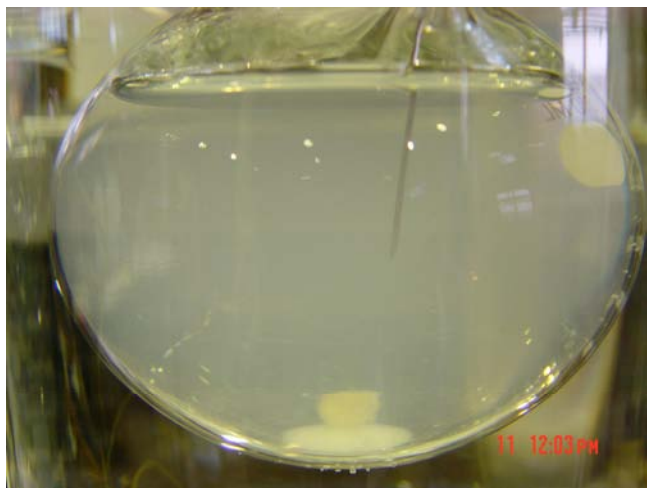
(c) pH=8.0 with 98 ppm Al

Figure 8.   Photographs of the test flasks after 30 days containing 2500 ppm B, 98 ppm Al, and three different NaOH concentrations to adjust the solution pH: (a) pH=7.0, (b) pH=7.5, and (c) pH=8.0. Note yellow fiberglass at the bottom of the flask.

(a) 98 ppm @ 120 F	(b) 70 ppm @ 140 F
(c) 55 ppm @ 120 F	(d) 40 ppm @ 120 F
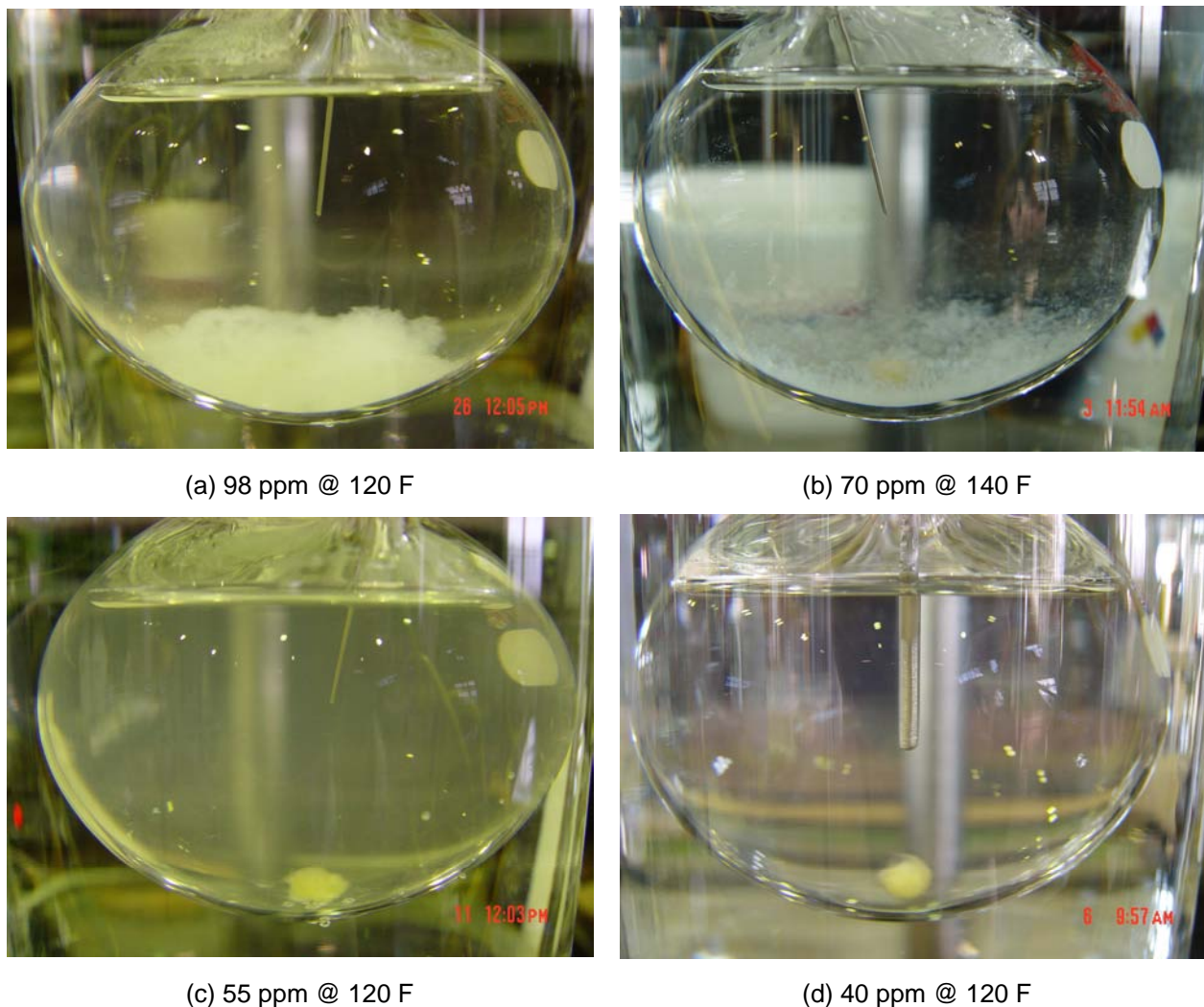
Figure 9.   Photographs of the flasks in which test solutions at constant pH of 7.5 contain 2500 ppm B and four different Al concentrations: (a) 98, (b) 70, (c) 55, and (d) 40 ppm. Note yellow fiberglass at the bottom of the flask.

Chemical composition analyses by ICP/OES were performed for the post-test solutions. The results for all solution samples are summarized in Table B1. The Al concentrations of the supernates of the flocculated solutions before and after filtering by 20-nm size filters are almost the same. For example, the supernate of '98 ppm Al and pH=7.0' test solution has only 2 ppm Al regardless of filtering. The unfiltered supernate samples of the cloudy solutions have much higher Al concentrations than those of the flocculated solutions.   However, the Al concentrations decrease drastically after filtering. For example, the sample of '40 ppm Al and pH=7.5' test solution had 33 ppm Al before filtering but after filtering by 20-nm size filter the concentration dropped down to 4 ppm Al. This suggests that most of the particles in the cloudy solutions are larger than 20 nm, which is consistent with earlier ANL's analysis[1], the particle sizes in cloudy Al solutions with boron ranges from 100 to 500 nm depending on the

turbidity. In most cases, the concentrations of Al in the supernates of both types of solutions after filtering are similar when compared at comparable pH values. This suggests the differences in the precipitate characteristics due to flocculation do not indicate an actual difference in solubility.
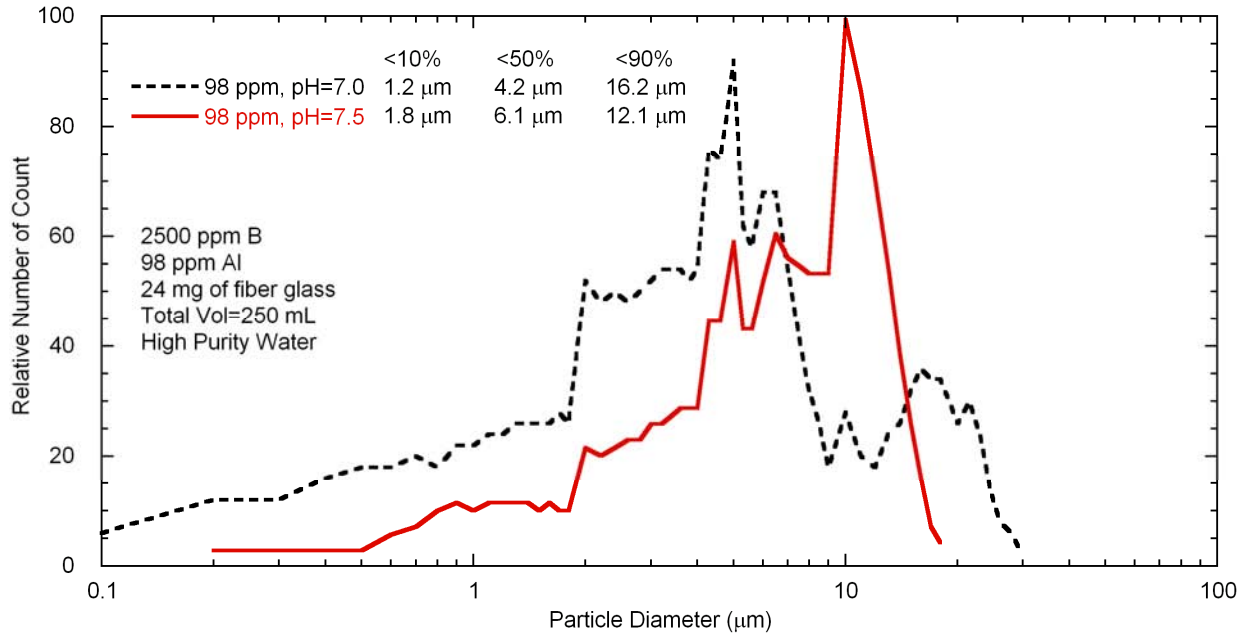
(a) 98 ppm Al solutions
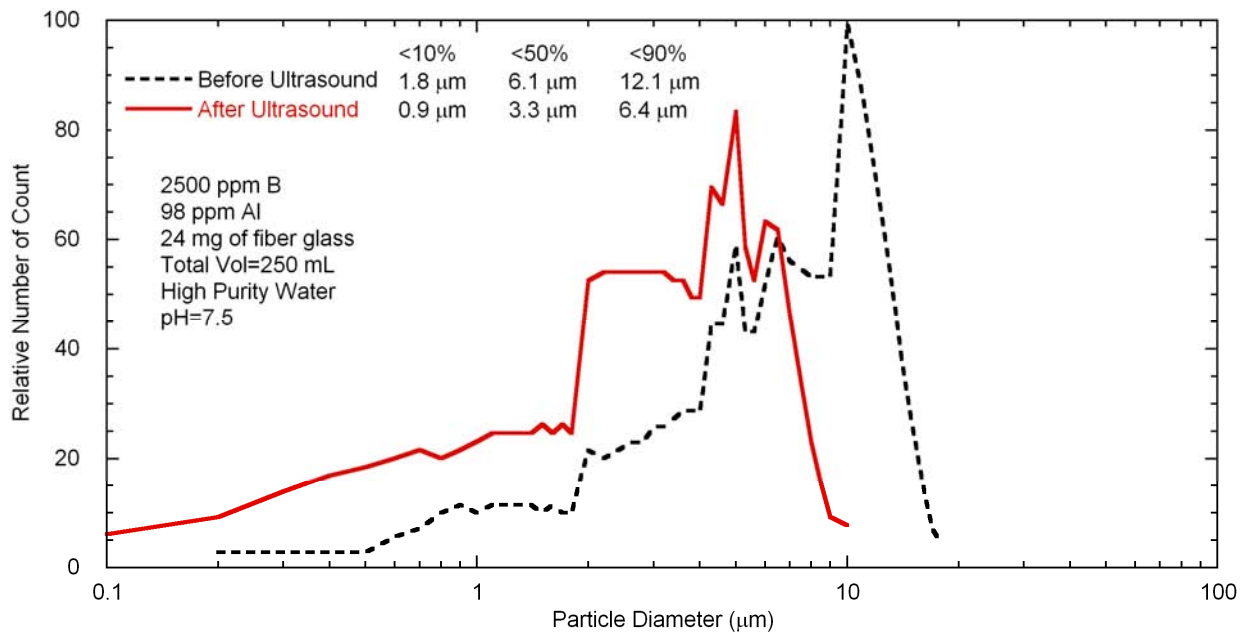
(b) Effect of ultrasonic deflocculation

Figure 10. Particle size distributions of 98 ppm Al post-test solutions showing the effect of (a) pH variation and (b) ultrasonic deflocculation.

13

Discussion

Colloid Stability: DLVO Theory

The discussion in this section is mainly taken from Ref. 6, a handbook of colloid chemistry. Colloidal particles in a dispersion medium are always subjected to Brownian motion with frequent collisions between them. Stability is thus determined by the nature of the interactions between the particles during such collisions. The first quantitative theory of interactions is referred to as the DLVO theory called after the names of the authors: Derjaguin, Landau, Verwey, and Overbeek. In this theory, the total interaction is assumed to be due to superposition of the van der Waals and double layer interactions. The total interaction energy is presented in the form:

$$V = V_{elec} + V_{vw}. \tag{1}$$

Typical curves of potential energy vs. inter-particle distance, as shown in Figure 11, exhibit an energy barrier against coagulation, and two potential minima, called the primary and secondary minimum. The primary minimum appears if strong short-range repulsive forces are present. If the particles cannot overcome the energy barrier, coagulation (flocculation) does not take place (See Figure 11a). This case usually occurs when the surface potential is high and/or ionic strength is low. With larger particles, the secondary minimum could be deep enough to cause flocculation (See Figure 11b). This case can happen when the surface potential is low and/or ionic strength is high. By addition of electrolyte or by decreasing the surface potential of the particles, we can suppress the electrostatic repulsion and thus decrease the height of the barrier and generate the secondary energy minimum that causes flocculation of colloidal particles. Interaction at this state is so weak that the flocculated state can be reversed if external energy is applied like a shear force due to flow.

Application of the DLVO Theory to the Test Results

The conventional DLVO theory discussed in the previous section was used to explain the flocculation tendency observed in the bench top tests. The electrostatic interaction energy is simply formulated in the case that two particles are identically charged particles of equal radius R and zeta potential ζ at a distance of separation d :[7]

$$V_{elec} = \frac{1}{2} \pi \varepsilon R \zeta^2 \ln \left[1 + e^{-\kappa d} \right]. \tag{2}$$

In Eq. (2) ε represents the electric permittivity of the surrounding medium, which is given by the product of relative and absolute permittivity. The relative permittivity of water is known as a function of temperature.[8] The zeta potential, which is the potential at the shear plane close to the Stern plane, can be experimentally measured and is often used as a measure of the surface potential. In Eq. (2) κ is known as the reciprocal Debye length, often referred to as the double layer thickness, which can be calculated from

$$\kappa^2 = \frac{2F^2}{\varepsilon R_o T} I \tag{3}$$

where F is Faraday's constant, R_o is universal gas constant, T is temperature in Kelvin scale, and I is ionic strength, which is defined by

14
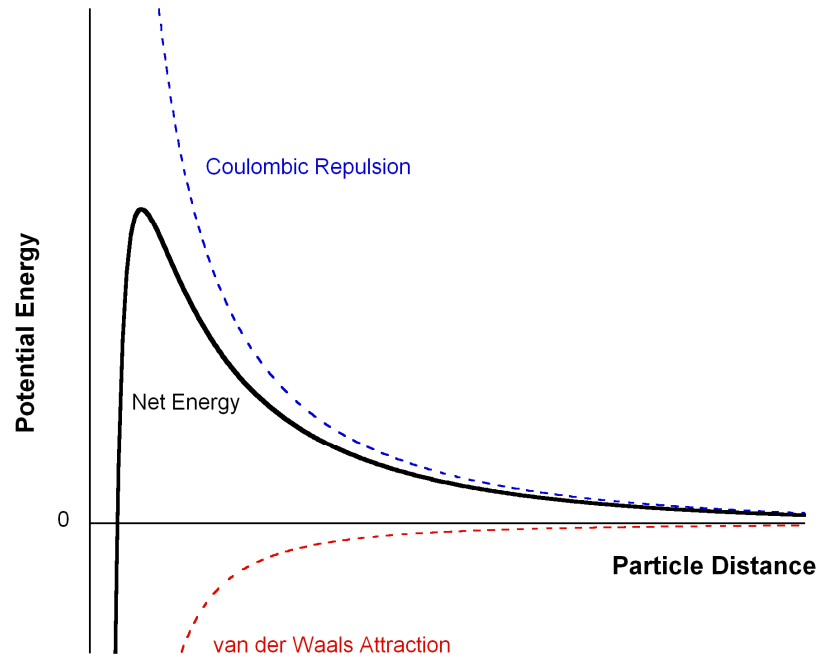
(a) Higher surface charge and/or lower ionic strength
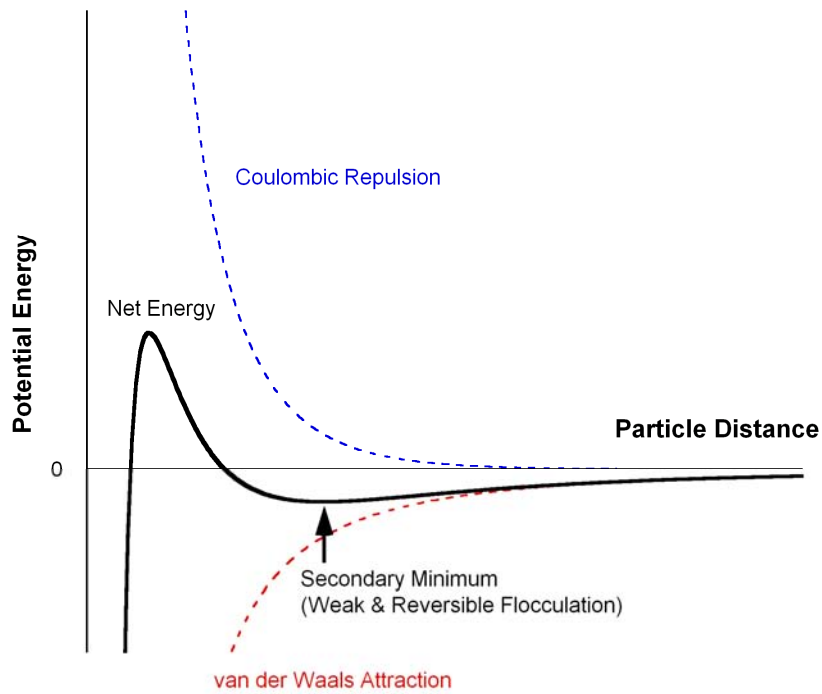
(b) Lower surface charge and/or higher ionic strength

Figure 11. Schematics of net potential energy between two colloidal particles for the cases of (a) higher surface charge and/or lower ionic strength and (b) lower surface charge and/or higher ionic strength.

$$I = \frac{1}{2}\sum_i z_i^2 c_i \; .$$ (4)

In Eq. (4) z_i and c_i represent the valency and molar concentration of the ith ion, respectively.

For two spherical particles suspended in a liquid medium, the potential energy due to the van der Waals interaction can be calculated approximately using the following equation:[7]

$$V_{vw} = -\frac{AR}{12d} \quad \text{for } d \ll R \, ,$$ (5)

where A is the combined Hamaker constant for the particle in the liquid; for alumina (Al_2O_3) particles this value is estimated as 3.9 k_BT in water,[7] and k_B is the Boltzmann constant. The Hamaker constant is dependent on the liquid medium and particle characteristic itself.

Figure 12 shows the effect of ionic strength on the total interaction energy between colloidal particles. As discussed in the previous ANL technical letter report,[1] tap water enhances the flocculation of aluminum hydroxide or any other surrogates as compared with high purity water. As shown in Figure 12, the total interaction energy profile, especially the depth of the secondary energy minimum is strongly dependent on the ionic strength. The van der Waals attractive force is not affected by the ionic strength change, but the electrostatic energy is drastically reduced as the ionic strength increases because the electric double layer around the particle is suppressed by the screening effect of dissolved ions. Figure 13 shows the effect of particle size on the total interaction energy. The electrostatic energy is not significantly affected by the increase of particle size, but the van der Waals attraction energy is significantly increased by the increase of the particle size. Once the particles start to grow, it is expected that the particle growth will be accelerated due to the deeper secondary potential minimum with bigger particle size. Figure 14 shows the effect of the zeta potential on the total interaction energy. Like the effect of ionic strength, the zeta potential does not affect the van der Waals force but the electrostatic force is increased by the increase of the zeta potential.

The zeta potential is affected by the ionic strength, but is primarily dependent on the solution pH. Typically the zeta potential is positive at acidic solution due to H^+ adsorption and becomes negative as the solution pH increases. At somewhere between acidic and alkaline pH the zeta potential is zero, which means the surface charge on a particle is neutral. The solution pH where the zeta potential is zero is called the point of zero charge (PZC). PZC can be varied by the presence of specific ions having strong adsorption tendency like SO_4^{2-} and $H_2PO_4^-$. Ions such as Cl^- and NO_3^- usually do not modify the PZC. Since the zeta potential becomes less as the solution pH approaches the PZC, it is expected that rapid coagulation would occur near the PZC.

In the bench top tests, since Al hydroxide is of interest, the PZC and the effect of specific ions on the PZC of Al hydroxide are needed to calculate the total interaction energy using DLVO theory. The PZC for pseudo-boehmite[9], gibbsite ($Al(OH)_3$, crystalline aluminum hydroxide or Al_2O_3-$3H_2O$)[10], and alumina[11] are available. All three Al oxide forms have very similar PZC value ranging from 8.9 to 9.2 in 0.01 M NaCl or $NaNO_3$ solution. Beyrouty et al.[12] reported that the PZC for amorphous $Al(OH)_3$ free of any specifically adsorbed anions was 9.72, which is slightly higher than other crystalline Al hydroxides. In these bench top tests, 2500 ppm boron as boric acid, less than 100 ppm of Al, and several hundreds ppm of Na were introduced in high purity water. There is no SO_4^{2-} or $H_2PO_4^-$ in solution, but any dissolved

ions from boric acid may affect the surface charge property of Al hydroxide particles. Alwitt[9] reported that the PZC of pseudo-boehmite is decreased to less than 6 in solutions with 0.3 M of boric acid. Goldberg[13] stated that boron adsorption on both crystalline and amorphous Al oxides has a maximum at pH 6 to 8. This specific adsorption of borate anions produces a shift in the PZC of Al oxides to a more acidic pH value. Interpolating the experimental data with 0.3 and 0.1 M boric acid by Alwitt and combining the PZC data by Beyrouty, we can estimate the PZC of amorphous Al hydroxide in solutions with 2500 ppm as 6.8. In addition to the PZC value, the zeta potential profile as a function of pH for Al hydroxide precipitates is needed. Since the zeta potential profile of amorphous Al hydroxide in 2500 ppm B solution is not available, the zeta potential profile of gibbsite in 0.01 M NaCl solution by Rowlands et al.[10] is introduced instead. However, the profile is shifted to more acidic direction and it has a new PZC value of 6.8 to reflect the effect of boron adsorption. The estimated zeta potentials at pH=7.0, 7.5, 8.0, and 8.5 in 2500 ppm B solution are –4, –8, –13, and –19 mV, respectively. The Na ion, which is used to adjust the solution pH, and borate ion ($B(OH)_4^-$, $B_3O_3(OH)_4^-$, $B_4O_5(OH)_4^{2-}$, etc.) are main elements to determine the ionic strength. The estimated ionic strength at pH=7.0, 7.5, 8.0, and 8.5 in 2500 ppm B are 0.015, 0.03, 0.045, and 0.06 M♣.

As shown in Table 1, the tendency for flocculation decreased with the increase of solution pH at 98 ppm Al. The calculated total interaction energy for three different pH test solutions of 98 ppm Al predicted that pH=8.0 test solution has higher secondary potential minimum than pH=7.0 or 7.5 solutions, which suggests that the pH=8.0 solution should have a greater tendency for flocculation, but this prediction is not consistent with the observed results. However, Wiese and Healy[11] experimentally showed that rapid coagulation of alumina particles, whether induced by pH or KNO_3 concentration changes, occurs at $|\zeta|$ < 14 mV. This result appears to be consistent with the observed results in the 98 ppm Al tests, because the estimated zeta potential of pH=7.5 solution is –8 mV and that of pH=8.0 solution is –13 mV, which is close to the critical zeta potential of 14 mV. Quantitative comparisons may be difficult, since Eq. (2) may overestimate the effect of ionic strength and/or underestimate the zeta potential effect. It is noted that Eq. (2) is the simplest form that can calculate the electrostatic energy between colloidal particles, and the conventional DLVO theory does not include other interaction forces like steric interaction, hydration interaction, polymeric bridging, and hydrophobic interaction.

In Table 1, at pH=7.5 the flocculation tendency of Al hydroxide depends on the total Al concentration; at 55 and 40 ppm Al the solution became cloudy but at 70 and 98 ppm Al the solution became flocculated. A change in Al concentration within the range in the tests does not significantly affect either the ionic strength change or zeta potential. Therefore, this specific observation cannot be explained by the conventional DLVO theory, but our results are consistent with the experimental results of Wiese and Healy[11] who showed that at the same pH value the stability of particles is a function of the particle concentration; higher particle concentration causes easier flocculation. It is expected that higher particle concentration has a higher frequency of inter-particle collision. The flocculation tendency difference at pH=7.5 may be attributed to the difference of particle concentration. In addition, at lower particle concentrations where the B/Al molar ratio becomes high, it may be more efficient for borate

---

♣ The ionic strength was estimated using ICP data for Na concentrations: 250, 500, 800, and 1250 ppm at pH=7.0, 7.5, 8.0, and 8.5, respectively. MULTEQ indicated that divalent polyborate ion became significant with 2500 ppm B so that the estimated ionic strength was adjusted. These estimated values should be used as only reference due to the uncertainty of divalent polyborate ion concentration.

ions to cover the surface of particles and increase the resistance to flocculation at the given total boron concentration.
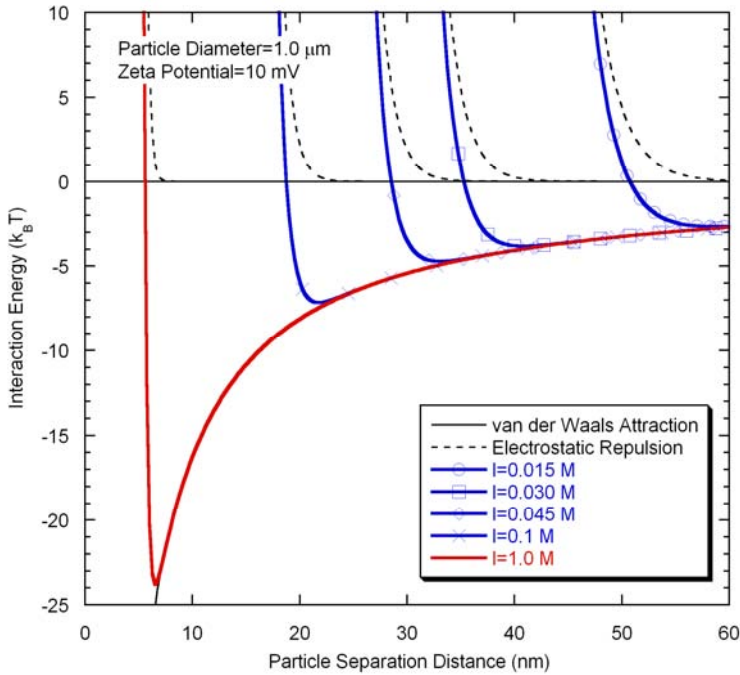

Figure 12.
Total interaction energy between colloidal particles calculated by DLVO theory as a function of ionic strength at constant particle size and zeta potential.
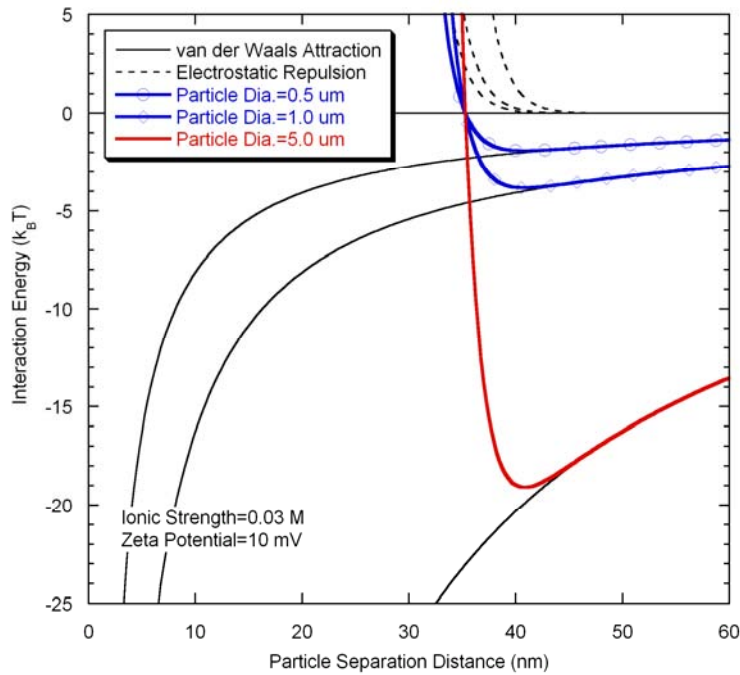
Figure 13.
Total interaction energy between colloidal particles calculated by DLVO theory as a function of particle size at constant ionic strength and zeta potential.
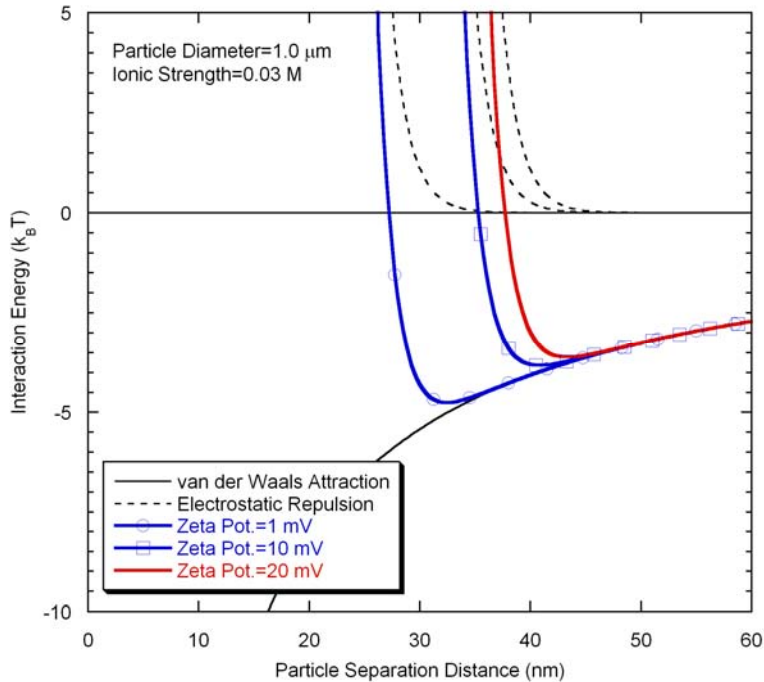
Figure 14.
Total interaction energy between colloidal particles calculated by DLVO theory as a function of zeta potential at constant particle size and ionic strength.

Interaction between Particles and Glass Fiber Bed

The potential increase of head loss in a glass fiber bed due to chemical effects is related to how colloidal particles and glass fibers interact with each other. Imdakm and Sahimi[14] investigated the reduction in permeability in a porous medium due to the transport of particles using Monte Carlo simulation. If the particle size is comparable to or larger than the pore size in the porous medium, "size exclusion," in which large particles block smaller pores, is the dominant mechanism of the reduction of permeability of the porous medium. This kind of phenomenon is called a percolation process. Their results show that with this mechanism, the permeability of the porous medium can drop down to zero even if unplugged pore volume ratio still remains higher than 0.9. Such results are consistent with results from ANL head loss tests[1] that show in some cases only a volume of precipitate equivalent to the removal of 1.5 ppm Al from solution is needed to completely plug the glass fiber bed. These tests suggest that if the average particle size is larger than about 10 μm, which is an estimated pore size of the fiberglass-only bed, a rapid increase of head loss occurred with particle additions. The rapid increase in these cases appears to be dominated by the size exclusion mechanism, but the relatively slow and continuous increase of head loss observed in other tests needs to be explained by another mechanism.

If the particle size is less than the pore size, size exclusion may not be sufficient to explain increases in head loss. Since the particle size is small enough to pass through the porous medium without restriction unless there is interaction between the particle and the porous medium, the interaction energy should be considered. Torkzaban et al.[15] simulated the colloidal particle attachment in porous media. They showed that colloidal attachment was only possible on regions of the collector where the torque from hydrodynamic shear acting on colloids adjacent to collector surfaces was less than the adhesive (DLVO) torque that resists detachment. The fraction of the porous media surface area on which attachment was possible increased with solution ionic strength, collector size, and decreasing flow velocity, as expected.

19

The flocculated Al hydroxide precipitates observed in these bench top tests in which the average particle size is 4-6 μ m are expected to result in head loss by both plugging mechanisms described above because their particle size is slightly less than the pore size of the fiberglass-only bed. The non-flocculated and very fine Al hydroxide precipitates will be dominated solely by the hydrodynamic and DLVO forces.

Major Findings and Summary

❖ The bench top Al solubility test results fit reasonably well, especially near room temperature, with the earlier ANL prediction results based on amorphous Al hydroxides in boron environment but differs from other literature data based on crystalline boehmite in environments without boron.

❖ These results did not contradict the 30-day solubility data at 140 and 200°F for $Al(OH)_3$ in WCAP-16785-NP[2] for solutions with pH 8.0 or greater. WCAP-16785-NP also states that perturbations in pH down to 7.0 did not affect these results. The test results reported here, that were based on longer term tests, show that the WCAP limits are not conservative for pH values as low as 7.0.

❖ Aluminum hydroxide precipitates showed two different types of behavior under the solubility limit depending on the solution pH and total Al concentration; the precipitates become flocculated (low pH and high Al concentration) or the solution becomes just cloudy (high pH and low Al concentration). Flocculated precipitates have the average particle diameter of 4-6 μ m (98 ppm Al at pH=7.0 and 7.5) and these flocculated precipitates will cause significant head loss in a fiberglass-only bed while solutions that were only visibly cloudy will not cause significant head loss, but may cause a different head loss response given different bed conditions where their pore sizes are smaller than the fiberglass-only bed.

❖ Most of the particles in the cloudy solutions are larger than 20 nm, which is consistent with earlier ANL results[1]. The differences in the precipitate characteristics due to flocculation do not indicate an actual difference in solubility.

❖ A colloidal stability theory called DLVO theory can qualitatively explain the ionic strength or solution pH effect on the flocculation tendency of the precipitates. In particular, solutions of high ionic strength will yield precipitates that are more likely to agglomerate and settle.

❖ At constant Al concentration, the flocculation tendency is dependent on the solution pH. This may be attributed to the zeta potential change according to the solution pH, which is consistent with earlier literature data.

❖ At constant pH, the flocculation tendency is dependent on total Al concentration; higher Al concentration tends to have a higher tendency of flocculation. Based on earlier literature data, it seems that particle concentration affects the flocculation tendency. It is expected that at a higher particle concentration, the frequency of inter-particle collision will be higher.

References

1. C. B. Bahn, K. E. Kasza, W. J. Shack, and K. Natesan, *Technical Letter Report on Evaluation of Chemical Effects: Studies on Precipitates Used in Strainer Head Loss Testing,* U.S. Nuclear Regulatory Commission, Washington D.C., January 30, 2008, AgencyWide Documents Access & Management System (ADAMS) Accession No. ML080600223.

2. Richard D. Reid, Kurtis R. Crytzer, and Ann E. Lane, *Evaluation of Additional Inputs to the WCAP-16530-NP Chemical Model,* WCAP-16785-NP, Revision 0, Westinghouse Electric Company LLC, May 2007.

3. W.J. Shack, *Technical Letter Report on WCAP-16530-NP, Evaluation of Post-Accident Chemical Effects in Containment Sump Fluids to Support GSI-191*, U.S. Nuclear Regulatory Commission, Washington, D.C., February 2007, ADAMS Accession No. ML080650350.

4. P. Benezeth, D. A. Palmer, and D. J. Wesolowski, "Aqueous high-temperature solubility studies. II. The solubility of boehmite at 0.03 m ionic strength as a function of temperature and pH as determined by in situ measurements," *Geochim. Cosmochim. Acta* **65** (2001) 2097-2111.

5. H. A. Van Straten and P. L. De Bruyn, "Precipitation from Supersaturated Aluminate Solutions," *J. Colloid Interface Sci.* **102** (1984) 260-277.

6. K. S. Birdi, Ed., "Handbook of surface and colloid chemistry," 2nd Ed., CRC Press: FL, USA, 2003, Ch. 5.

7. K. S. Birdi, Ed., "Handbook of surface and colloid chemistry," 2nd Ed., CRC Press: FL, USA, 2003, Ch. 7.

8. G. C. Akerlof and H. I. Oshry, *J. Am. Chem. Soc.,* 72 (1950) 2844.

9. R. S. Alwitt, "The Point of Zero Charge of Pseudoboehmite," *J. Colloid Interface Sci.* **40** (1972) 195-198.

10. W. N. Rowlands, R. W. O'Brien, R. J. Hunter, and V. Patrick, "Surface Properties of Aluminum Hydroxide at High Salt Concentration," *J. Colloid Interface Sci.* **188** (1997) 325-335.

11. G. R. Wiese and T. W. Healy, "Coagulation and Electrokinetic Behavior of TiO_2 and Al_2O_3 Colloidal Dispersions," *J. Colloid Interface Sci.* **51** (1975) 427-433.

12. C. A. Beyrouty, G. E. Van Scoyoc, and J. R. Feldkamp, "Evidence Supporting Specific Adsorption of Boron on Synthetic Aluminum Hydroxides," *Soil Sci. Soc. Am. J.* **48** (1984) 284-287.

13. S. Goldberg, "Reactions of boron with soils," *Plant and Soil* **193** (1997) 35-47.

14. A. O. Imdakm and Muhammad Sahimi, "Transport of large particles in flow through porous media," *Physical Review A,* **36** (1987) 5304-5309.

15. S. Torkzaban, S. A. Bradford, and S. L. Walker, "Resolving the Coupled Effects of Hydrodynamics and DLVO Forces on Colloid Attachment in Porous Media," *Langmuir,* **23** (2007) 9652-9660.

Appendix A: Full Test History (Daily Observation Results)

Figure A1. Long-term solubility test schedule from November 12 to December 19, 2007 ('AN' means aluminum nitrate and 'SA' means sodium aluminate as the Al source).

Table A1.   Daily observation history for '98 ppm & pH=7.0' test solution using aluminum nitrate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	3.65 @ 185 F	200->180 F	Solution was cloudy initially but becomes clear.
2	180			The test was terminated.

Table A2.   Daily observation history for '98 ppm & pH=7.5' test solution using aluminum nitrate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	6.85 @ 194 F	200->180 F	Precipitates were formed and flocculated.
2	180			The test was terminated.

Table A3.   Daily observation history for '98 ppm & pH=8.0' test solution using aluminum nitrate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.45 @ 194 F	200->180 F	Cloudy precipitates were not readily dissolved.
2	180			The test was terminated.

Table A4.   Daily observation history for '40 ppm & pH=7.5' test solution using aluminum nitrate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200		200->180 F	Solution was clear but small cloudy precipitates were present at the bottom.
2	180		180->160 F	Solution was clear but small cloudy precipitates were present at the bottom.
3	160	7.29 @ 160 F	160->140 F	Solution became slightly cloudy and small cloudy precipitates were still present at the bottom.
4	140		140->120 F	Solution became cloudier than yesterday.
5	120			Not observed
6	120			Not observed
7	120	7.25 @ 119 F		The test was terminated.

Table A5. Daily observation history for '40 ppm & pH=8.0' test solution using aluminum nitrate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200		200->180 F	Solution was clear.
2	180		180->160 F	Solution was clear.
3	160	7.78 @ 159 F	160->140 F	Solution was clear.
4	140		140->120 F	Solution was clear
5	120			Not observed
6	120			Not observed
7	120	7.79 @ 116 F		Solution became slightly cloudy. The test was terminated.

Table A6. Daily observation history for '40 ppm & pH=8.5' test solution using aluminum nitrate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	8.2 @ 194 F	200->180 F	Solution was clear.
2	180		180->160 F	Solution was clear.
3	160	8.24 @ 161 F	160->140 F	Solution was clear.
4	140		140->120 F	Solution was clear.
5	120			Not observed
6	120			Not observed
7	120			Solution was clear. The test was terminated.

Table A7. Daily observation history for '98 ppm & pH=7.0' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.15 @ 199 F	200->180 F	Solution was cloudy and small precipitates are at the bottom.
2	180	7.22 @ 179 F	180->160 F	Flocculation occurred at the inner surfaces of the flask. Precipitates slid down by slight agitation and accumulated at the bottom.
3	160	7.24 @ 160 F	160->140 F	Flocculated precipitates were at bottom.
4	140			Not observed
5	140			Not observed
6	140	7.24 @ 140 F		Flocculated precipitates were at bottom.
7	140		140->100->140 F	Flocculated precipitates were at bottom. It looked same as yesterday.
8	140		140->100->140 F 140->120 F	Flocculated precipitates were at bottom.
9	120			Not observed
10	120			Not observed

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
11	120			Not observed
12	120			Not observed
13	120	7.26 @ 118 F	120->90->120 F	Flocculated precipitates were at bottom.
14	120			Test was terminated.

Table A8.   Daily observation history for '98 ppm & pH=7.5' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.55 @ 201 F	200->180 F	Solution was clear.
2	180	7.60 @ 179 F	180->160 F	Flocculation occurred at the inner surfaces of the flask. Precipitates slid down by slight agitation and accumulated at the bottom.
3	160	7.59 @ 160 F	160->140 F	Flocculated precipitates were at the bottom.
4	140			Not observed
5	140			Not observed
6	140	7.62 @ 138 F		Flocculated precipitates were at the bottom.
7	140		140->100->140 F	Flocculated precipitates were at the bottom. It looked same as yesterday.
8	140		140->100->140 F 140->120 F	Flocculated precipitates were at the bottom.
9	120			Not observed
10	120			Not observed
11	120			Not observed
12	120			Not observed
13	120	7.64 @ 120 F	120->90->120 F	Flocculated precipitates were at the bottom.
14				Test was terminated.

Table A9.   Daily observation history for '98 ppm & pH=8.0' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.88 @ 202 F	200->180 F	Solution was clear.
2	180	7.94 @ 180 F	180->160 F	Solution was clear.
3	160	7.92 @ 159 F	160->140 F	Solution was clear.
4	140			Not observed
5	140			Not observed
6	140	7.99 @ 138 F		Solution became very slightly cloudy but no flocculation and did not settle down.
7	140		140->100->140 F	Solution became cloudier than yesterday.
8	140		140->100->140 F 140->120 F	Solution was cloudy.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
9	120			Not observed
10	120			Not observed
11	120			Not observed
12	120			Not observed
13	120	8.04 @ 124 F	120->90->120 F	Solution was cloudy.
14	120			Solution was cloudy.
15	120			Solution was cloudy.
16	120		120->90->120 F	Solution was cloudy.
17	120		120->100 F	Solution was cloudy.
18	100			Not observed
19	100			Not observed
20	100		100->80->100 F	Solution was cloudy but white fine precipitates settled at the bottom.
21	100	8.09 @ 96 F		Solution was cloudy and white precipitates were at the bottom.
22	100			Solution was cloudy and white precipitates were at the bottom.
23	100		100->80->100 F	Solution was cloudy and white precipitates were at the bottom.
24	100			Solution was cloudy and white precipitates were at the bottom.
25	100			Not observed
26	100			Not observed
27	100		100->80 F	Solution was cloudy and white precipitates were at the bottom.
28	80			Solution was cloudy and white precipitates were at the bottom.
29	80	8.18 @ 77 F		About 1/8" deep region from the top water level was clear. It appeared to start settling. Test was terminated.

Table A10. Daily observation history for '40 ppm & pH=7.5' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.45 @ 196 F	200->180 F	Solution was clear.
2	180	7.47 @ 177 F	180->160 F	Solution was clear.
3	160	7.46 @ 160 F	160->140 F	Solution was clear.
4	140			Not observed
5	140			Not observed
6	140			Not observed
7	140			Not observed
8	140	7.50 @ 141 F	140->100->140 F	Solution was clear.
9	120			Solution was clear.
10	120		140->120 F	Solution was clear.
11	120	7.55 @ 117 F	120->90->120 F	Solution became very slightly cloudy.
12	120			Solution was very slightly cloudy.
13	120			Not observed
14	120			Not observed
15	120		120->90->120 F	Solution was slightly cloudy.

16	120			Solution was slightly cloudy.
17	120			Solution was slightly cloudy.
18	100		120->90->120 F 120->100 F	Solution was slightly cloudy.
19	100	7.57 @ 102 F		Solution was slightly cloudy.
20	100			Not observed
21	100			Not observed
22	100		100->80->100 F	Solution became cloudier.
23	100			Solution was cloudy.
24	100			Solution was cloudy.
25	100		100->80->100 F	Solution was cloudy.
26	100		100->80 F	Solution was cloudy.
27	100			Not observed
28	80			Not observed
29	80			Solution was cloudy but cloudier than the solution of pH=8.0.
30	80	7.56 @ 84 F		Test was terminated.

Table A11. Daily observation history for '40 ppm & pH=8.0' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.88 @ 200 F	200->180 F	Solution was clear.
2	180	7.91 @ 182 F	180->160 F	Solution was clear.
3	160	7.91 @ 160 F	160->140 F	Solution was clear.
4	140			Not observed
5	140			Not observed
6	140			Not observed
7	140			Not observed
8	140	7.92 @ 141 F	140->100->140 F	Solution was clear.
9	120			Solution was clear.
10	120		140->120 F	Solution was clear.
11	120	7.98 @ 118 F	120->90->120 F	Solution was clear.
12	120			Solution was clear.
13	120			Not observed
14	120			Not observed
15	120		120->90->120 F	Solution became very slightly cloudy.
16	120			Solution was slightly cloudy.
17	120			Solution was slightly cloudy.
18	100		120->90->120 F 120->100 F	Solution was slightly cloudy.
19	100	8.03 @ 95 F		Solution was slightly cloudy.
20	100			Not observed
21	100			Not observed
22	100		100->80->100 F	Solution became cloudier.
23	100			Solution was cloudy.
24	100			Solution was cloudy.
25	100		100->80->100 F	Solution was cloudy.
26	100		100->80 F	Solution was cloudy.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
27	100			Not observed
28	80			Not observed
29	80			Solution was cloudy but less cloudy than the solution of pH=7.5.
30	80	8.02 @ 82 F		Test was terminated.

Table A12. Daily observation history for '40 ppm & pH=8.5' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	8.23 @ 197 F	200->180 F	Solution was clear.
2	180	8.29 @ 180 F	180->160 F	Solution was clear.
3	160	8.33 @ 156 F	160->140 F	Solution was clear.
4	140			Not observed
5	140			Not observed
6	140			Not observed
7	140			Not observed
8	140	8.35 @ 139 F	140->100->140 F	Solution was clear.
9	120			Solution was clear.
10	120		140->120 F	Solution was clear.
11	120	8.42 @ 118 F	120->90->120 F	Solution was clear.
12	120			Solution was clear.
13	120			Not observed
14	120			Not observed
15	120		120->90->120 F	Solution was clear.
16	120			Solution was clear.
17	120			Solution was clear.
18	100		120->90->120 F 120->100 F	Solution was clear.
19	100	8.43 @ 104 F		Solution was clear.
20	100			Not observed
21	100			Not observed
22	100		100->80->100 F	Solution was clear.
23	100			Solution was clear.
24	100			Solution was clear.
25	100		100->80->100 F	Solution was clear.
26	100		100->80 F	Solution was clear.
27	100			Not observed
28	80			Not observed
29	80			Solution became very slightly cloudy.
30	80	8.51 @ 85 F		Test was terminated.

Table A13. Daily observation history for '70 ppm & pH=7.0' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.10 @ 200 F	200->180 F	Solution was clear.

28

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
2	180	7.16 @ 180 F	180->160 F	Al precipitates flocculation occurred at the inner surface of the flask. They slid down by slight agitation and accumulated at the bottom.
3	160	7.18 @ 163 F	160->140 F	Flocculated precipitates were at the bottom.
4	140			Not observed
5	140			Not observed
6	140	7.19 @ 133 F		Flocculated precipitates were at the bottom. The test was terminated.

Table A14. Daily observation history for '70 ppm & pH=7.5' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.53 @ 199 F	200->180 F	Solution was clear.
2	180	7.52 @ 183 F	180->160 F	Al precipitates flocculation occurred at the inner surface of the flask. Precipitates slid down by slight agitation and accumulated at the bottom. Total volume looked less than pH=7.0.
3	160	7.55 @ 163 F	160->140 F	Flocculated precipitates were at the bottom.
4	140			Not observed
5	140			Not observed
6	140	7.58 @ 135 F		Flocculated precipitates were at the bottom. The test was terminated.

Table A15. Daily observation history for '55 ppm & pH=7.0' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.14 @ 199 F	200->180 F	Solution was clear.
2	180	7.14 @ 179 F	180->160 F	Al flocculation occurred at the inner surface of the flask. Precipitates slid down by slight agitation and accumulated at the bottom.
3	160	7.14 @ 156 F	160->140 F	Flocculated precipitates were at the bottom.
4	140	7.16 @ 137 F	140->100->140 F	Flocculated precipitates were at the bottom.
5	140			Not measured
6	140			Not measured
7	140		140->100->140 F 140->120 F	Flocculated precipitates were at the bottom.
8	120	7.17 @ 122 F	120->90->120 F	Flocculated precipitates were at the

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
				bottom.
9	120		120->90->120 F	Flocculated precipitates were at the bottom.
10	120		120->90->120 F 120->100 F	Flocculated precipitates were at the bottom.
11	100	7.17 @ 104 F	100->80->100 F	Flocculated precipitates were at the bottom.
12	100			Not measured
13	100			Not measured
14	100		100->80->100 F	Flocculated precipitates were at the bottom.
15	100		100->80 F	Flocculated precipitates were at the bottom.
16	80	7.22 @ 82 F		Test was terminated.

Table A16. Daily observation history for '55 ppm & pH=7.5' test solution using sodium aluminate.

Day	Nominal Solution Temp. (F)	Solution pH	Temperature Change or Cycling	Observation & Action Description
1	200	7.50 @ 198 F	200->180 F	Solution was clear.
2	180	7.49 @ 176 F	180->160 F	Solution was clear.
3	160	7.54 @ 156 F	160->140 F	Solution became slightly cloudy.
4	140	7.56 @ 138 F	140->100->140 F	Solution became cloudier than yesterday.
5	140			Not measured
6	140			Not measured
7	140		140->100->140 F 140->120 F	Solution was cloudy.
8	120	7.58 @ 124 F	120->90->120 F	Solution was cloudy.
9	120		120->90->120 F	Solution was cloudy.
10	120		120->90->120 F 120->100 F	Solution was cloudy.
11	100	7.60 @ 103 F	100->80->100 F	Solution was cloudy.
12	100			Not measured
13	100			Not measured
14	100		100->80->100 F	Solution was cloudy.
15	100		100->80 F	Solution was cloudy.
16	80	7.59 @ 85 F		Test was terminated.

Appendix B: Summary of ICP Analyses for the Post-test Solution Samples

Table B1.   Summary of ICP analyses for the post-test solution samples.

Sample No.	Sample Identification	Chemical Composition (ppm)				
		Al	B	Ca	Si	Na
SOL-01	98 ppm, pH=7.0 Meas. pH=7.30 @ 76.3 F Unfiltered	2.12	2390	0.62	2.51	287
SOL-02	98 ppm, pH=7.0 Meas. pH=7.30 @ 76.3 F 0.02 um Filtrate	2.19	2390	0.56	2.45	287
SOL-03	98 ppm, pH=7.5 Meas. pH=7.68 @ 74.6 F Unfiltered	8.63	2480	0.51	1.68	498
SOL-04	98 ppm, pH=7.5 Meas. pH=7.68 @ 74.6 F 0.02 um Filtrate	8.92	2470	<0.5	1.68	496
SOL-05	70 ppm, pH=7.0, unfiltered	4.46	2480	<0.5	0.99	271
SOL-06	70 ppm, pH=7.0, 0.02um filtrate	4.42	2440	<0.5	1.02	270
SOL-07	70 ppm,pH=7.5, unfiltered	19.1	2480	<0.5	1.26	476
SOL-08	70 ppm,pH=7.5, 0.02um filtrate	19.0	2500	<0.5	1.28	482
SOL-09	98ppm, pH=8.0, unfiltered	56.1	1930	0.89	2.24	630
SOL-10	98ppm, pH=8.0, 0.02um filtrate	6.29	1900	<0.5	0.72	616
SOL-11	40 ppm,pH=7.5, unfiltered	33.2	2540	0.70	1.87	451
SOL-12	40ppm, pH=7.5,0.02um filtrate	3.63	2540	<0.5	1.18	463
SOL-13	40ppm,pH=8.0, unfiltered	23.5	2590	0.92	3.40	778
SOL-14	40 ppm,pH=8.0, 0.02um filtrate	7.47	2560	<0.5	2.16	773
SOL-15	40ppm,pH=8.5, unfiltered	33.6	2470	0.60	3.42	1230
SOL-16	40ppm,pH=8.5,0.02um filtrate	20.4	2420	<0.5	2.42	1200
SOL-17	55ppm,pH=7.0, unfiltered	2.62	2480	<0.5	1.06	255
SOL-18	55ppm,pH=7.0, 0.02 um filtrate	2.94	2400	0.53	1.08	253
SOL-19	55ppm,pH=7.5, unfiltered	35.7	2530	0.71	1.68	473
SOL-20	55ppm, pH=7.5, 0.02um filtrate	4.44	2540	<0.5	1.04	462