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Preliminary Results from Plutonium/Americium Solubility Studies Using Simulated Savannah River Site Waste Solutions

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Summary

To address the accelerated disposition of the supernate and salt portions of Savannah River Site (SRS) high level waste (HLW), solubility experiments were performed to develop a predictive capability for plutonium (Pu) and americium (Am) solubility. Preliminary results from the experiments indicate hydroxide (OH^-), carbonate (CO_3^{2-}), nitrate (NO_3^-) and temperature are the predominant parameters affecting Pu and Am solubilities in alkaline salt solutions representative of those present in SRS waste tanks. This study used a statistically designed experimental matrix to evaluate the effects of six major anionic components (OH^-, aluminate ($Al(OH)_4^-$), sulfate (SO_4^{2-}), CO_3^{2-}, NO_3^-, and nitrite (NO_2^-)) and temperature (25 and 80 °C). This work extends previous data on Pu solubility to a wider range of solution compositions and is the first systematic evaluation of Am solubility in SRS waste solutions.

Analysis of the solubility data collected in nominal 1 month intervals over a 3-month period showed no time-dependence of the Pu and Am concentrations. However, the data scatter among the three data sets was sufficiently large to introduce considerable variance in model parameters. The data scatter likely indicates that not all solutions have obtained equilibrium after 3 months. We recommend that the testing continue to obtain an additional two sets of data as planned in the experimental design.

First-order models of the data indicate that the Pu solubility is a function of OH^- and CO_3^{2-} concentrations and that the Am solubility is a function of temperature and OH^-, CO_3^{2-}, and NO_3^- concentrations. These factors are statistically significant with at least 90% confidence. We also evaluated the Pu solubility data obtained in this study with previous data from the literature to develop a modified response surface model. All of the salt concentrations and temperature terms are statistically significant at the 90% confidence level. Upon completion of the solubility testing, we will incorporate the additional data into the response surface model and complete an evaluation of parameter sensitivity and validation of predictive performance.

Introduction

The SRS is currently working to accelerate the disposition of the supernate and salt portions of the HLW contained in the SRS tank farm system. In particular, projects are underway to disposition salt waste according to its radionuclide content. For each disposition pathway, the Pu content of the waste is a dominating factor in determining the acceptance of the waste at the SRS Saltstone Facility. Since the specific activity of americium-241 (Am) is nominally a factor of 100 greater than Pu, process conditions which increase its solubility in salt solutions are also a concern. The solubility of Pu and Am in acidic waste streams resulting from Pu purification activities is relatively well known. When waste solutions containing these elements are neutralized, coprecipitated with other metals, and discharged into the HLW system, the solubilities change with time and with conditions under which the waste is stored or evaporated.

The solubility of Pu in alkaline waste solutions representative of SRS HLW has only been measured for a narrow range of process conditions. Hobbs et al. [1] investigated the solubility of Pu and uranium in alkaline salt solutions comparable to the waste solutions to be processed by the In-Tank Precipitation Process. Plutonium solubility data from other studies have been evaluated and a predictive model developed as a function of the free hydroxide concentration; however, other constituents in the waste solution can change the Pu solubility by as much as three orders of magnitude for a given hydroxide concentration.[2] Americium solubility in SRS waste solutions has never been measured. Limited data are available for Hanford tank waste [3] and pure sodium hydroxide solutions.[4]

The objective of this work was to measure the solubility of Pu and Am in simulated SRS waste solutions. A statistically designed experiment was performed in which the composition and temperature of the waste solutions were varied over expected waste tank ranges. The statistically designed experiment also allowed modeling of the solubility data and development of a predictive capability given the composition and temperature of waste tank solutions. Constituents of the simulated waste solutions included: OH^-, $Al(OH)_4^-$, SO_4^{2-}, CO_3^{2-}, NO_3^-, and NO_2^-. Each anion was added to the waste solution in the sodium form. Solubility experiments were performed at 25 and 80°C to bound temperatures normally seen in SRS waste tanks. Once the simulated waste solutions were prepared, weapons grade Pu and Am were added as a nitrate solution. The Pu and Am solubility data presented in this report are limited to 3 data sets from the analysis of samples over a 3 month time period. The analysis of additional samples over a 2-3 month period is planned to ensure equilibrium concentrations were reached.

Experimental

Constraints on Concentration

The range of salt concentrations and temperature expected in the SRS waste tanks are summarized in Table 1. Each anion was assumed to be in the sodium (Na^+) form.

Table 1 Range of Salt Concentrations and Temperature in SRS Waste Tanks

Parameter	Low (M)	High (M)
OH^-	0.0001	15
$Al(OH)_4^-$	0.001	1
SO_4^{2-}	0.001	0.5
CO_3^{2-}	0.001	1
NO_3^-	0.1	6
NO_2^-	0.1	6
Temperature (°C)	25	80

From the range of concentrations, one might expect that the preparation of solutions containing combinations of OH^- and salt concentrations near the maximum levels are not possible due to the precipitation of one or more of the salts. To eliminate solution compositions that cannot be prepared, a series of constraints on the OH^- and salt concentrations was utilized in the experimental design. The constraints are given by equations 1-3.

$$\left[Al(OH)_4^-\right] \le \frac{1}{3}\left[NO_3^-\right] \quad (1)$$

$$\left[OH^-\right] \ge 0.4M + \left[Al(OH)_4^-\right] \quad (2)$$

$$\left[Na^+\right]_{Total} \le 9M \quad (3)$$

The constraint on $Al(OH)_4^-$ given by equation 1 is based on the stoichiometry of aluminum nitrate ($Al(NO_3)_3$), the primary source of Al in the tank farm. The second constraint on $Al(OH)_4^-$ given by equation 2 was based on Al solubility studies in strongly alkaline solutions and ensures that $Al(OH)_4^-$ remains soluble at the high end of its concentration range. The constraint on total Na^+ was empirically determined during this study when attempts to prepare solutions containing high salt concentrations failed even when the solution compositions were based on an experimental design utilizing constraints 1 and 2. These results and information were important inputs to the experimental design process, which is discussed in the next section.

Statistical Design

The goal of this study was to generate Pu and Am solubility data over the factor space defined by the seven factors (and their intervals of possible values) in Table 1. Historical data of interest to this study were available for Pu solubility but not for Am solubility. The data are provided in Table A.1 in Appendix A. The last column of this table provides an indicator for whether or not the historical data point satisfies constraints 1-3. Those data in Table A.1 that met the restrictions on the factor space imposed by constraints 1-3 (71 data points in all) served as the starting place for the development of the test matrix for this task. This was true even for data that extended slightly beyond the region defined by Table 1 (i.e. not all of the species were used

to prepare the salt solutions). Thus, the test matrix designed for this study was selected to complement the previous Pu solubility efforts while providing a good initial basis for the study of the relationship between Am solubility and the factors of Table 1.

The Pu model of interest in this study is a response surface model in the 7 factors of Table 1 except for the quadratic temperature term, which is not of interest. Thus, the model of interest is made up of an intercept term, 7 main effect terms, 6 quadratic (or squared) terms, and 21 two-way interaction terms for a total of 35 terms. For the initial investigation of Am solubility, the model form of interest was a first-order model in the 7 factors.

Statistical routines are available to assist with experimental design problems. One such routine is the D-optimal routine available in JMP Version 3.2.6.[5] This routine selects a design of a specified size (i.e., number of design points) from a set of candidate points that optimizes the fitting of a specified model form. The optimization is relative to efficiency measures of the fitted model. (The optimal design criterion used by JMP Version 3.2.6 is D-optimality (i.e., to maximize the D-efficiency), where D-efficiency is a measure of design efficiency that is related to the determinant of the variance-covariance matrix of the design. [6])

To use this routine, there was a need to generate a set of candidate design points that were within the factor space of Table 1 and that satisfied constraints 1-3. This was accomplished by generating an initial, space-filling design using a modified, orthogonal Latin hypercube (OLH) approach. An OLH design is a space filling design that provides an opportunity for independently estimating main effects.[7] However, due to the restrictions imposed by constraints 1-3, the OLH was modified to satisfy these constraints. Temperature was excluded from this process, and each of the OLH points generated for the other factors was used to define candidate points, one at 25 °C and one at 80 °C. Exhibit A.1 in Appendix A displays the correlations and corresponding scatterplot matrix of the candidate points generated using this approach. As seen in this exhibit, the restrictions imposed by constraints 1-3 lead to correlations in the candidate design points among the factors.

A set of special candidate design points was added to these OLH points. These points were added (at temperatures of 25 and 80 °C) to represent possible solution concentrations that may result from evaporator operations. These selective design points are provided in Table A.2 of Appendix A and together with the OLH points, they provided the set of candidate design points that was submitted to the D-Optimal routines from which 8 optimal points were selected to support the fitting of a first-order model relating Am solubility to the 7 factors of this study. Exhibit A.2 provides the information generated by the routine, which was conducted for 1000 iterations as indicated in the exhibit. Some correlations among these design points at or near 50% are observed in this exhibit; this is an artifact of the restrictions imposed by constraints 1-3. Table A.3 in Appendix A provides a listing of these 8 design points.

Next, the D-Optimal routine of JMP Versions 3.2.6 was used a second time. In this run of the routine, the set of candidate design points was formed by adding the 71 acceptable data points from Table A.1, the OLH points, and the special data points of Table A.2. The model of interest for this run was the modified response surface model of interest in this study of Pu solubilities. For this use of D-Optimal, which was run for 1000 iterations, the 71 points from Table A.1 and

the 8 points from Table A.3 were forced into the test matrix and the routine was asked to complement these 79 points with 12 additional, "optimal" points for a total of 91 design points. The results from this run of D-Optimality are provided in Exhibit A.3 of Appendix A, and the final test matrix is given in Table 2. The low value for the G-efficiency of this design and the high correlations among the model terms as provided in the exhibit suggest that it is an ambitious undertaking to push the currently available set of 71 data points to a set of 91 for the fitting of the model of interest.

Table 2 Design Points for Pu and Am Solubility Experiments

Solution	OH^- (M)	$Al(OH)_4^-$ (M)	SO_4^{2-} (M)	CO_3^{2-} (M)	NO_3^- (M)	NO_2^- (M)	Temp (°C)
1	15.00	0.0010	0.0010	0.0010	0.1000	0.1000	25
2	15.00	0.0010	0.0010	0.0010	0.1000	0.1000	85
3	5.512	0.0167	0.0158	0.9109	0.7523	0.8151	85
4	0.8722	0.1323	0.0104	0.8877	4.8795	1.2663	25
5	0.8722	0.1323	0.0104	0.8877	4.8795	1.2663	85
6	2.268	0.6631	0.2065	0.0591	2.4717	2.9888	85
7	1.779	0.1133	0.1209	0.3627	0.6260	5.4283	85
8	1.640	0.4130	0.3459	0.2389	1.5864	4.1019	25
9	3.384	0.6089	0.2157	0.4540	2.5616	0.8756	25
10	2.895	0.7561	0.4826	0.2217	2.9888	0.6283	85
11	5.837	0.1128	0.4091	0.2914	0.3547	0.9686	85
12	2.756	0.0482	0.0880	0.9342	0.1686	3.5988	25
13	1.221	0.6012	0.1112	0.0358	4.1477	1.8151	25
14	0.4536	0.0313	0.1074	0.0726	3.6446	3.4159	25
15	1.291	0.7406	0.1809	0.9497	2.5012	0.1686	25
16	1.291	0.7406	0.1809	0.9497	2.5012	0.1686	85
17	2.070	0.0500	0.4671	0.8258	0.2500	0.7942	25
18	0.8606	0.0672	0.1809	0.9497	0.4663	0.1105	85
19	0.6280	0.0067	0.4362	0.4463	0.5151	0.4174	85
20	2.628	0.0246	0.0842	0.0126	0.1593	0.1453	85

Preparation of Salt Solutions

The 20 salt solutions specified as design points for the Pu and Am solubility experiments were based on the target concentrations given in Table 2. The solutions were prepared using ACS certified reagents. The starting materials are shown in Table 3. The amount of each starting material was based on the preparation of 100 mL of salt solution.

Table 3 Starting Materials Used for Preparation of Salt Solutions

Chemical	Concentration (wt%)	Formula
Sodium Hydroxide	50	NaOH
Aluminum Nitrate Nonahydrate		$Al(NO_3)_3 \cdot 9H_2O$
Sodium Sulfate Decahydrate		$Na_2SO_4 \cdot 10H_2O$
Sodium Carbonate Monohydrate		$Na_2CO_3 \cdot H_2O$
Sodium Nitrate		$NaNO_3$
Sodium Nitrite		$NaNO_2$

The solutions were prepared by initially transferring the desired amount of 50 wt% NaOH to a 250 mL beaker. The target amount of $Al(NO_3)_3 \cdot 9H_2O$ was then dissolved in 4-5 mL of deionized water in a separate beaker and mixed with the NaOH until all solids dissolved. The beaker was rinsed with 2, 1-2 mL aliquots of deionized water. The 250 mL beaker was stirred using a magnetic stirring bar. The target masses of $Na_2SO_4 \cdot 10H_2O$, $Na_2CO_3 \cdot H_2O$, $NaNO_3$, and $NaNO_2$ were then added to the caustic solution in the order indicated. Each salt was dissolved before the next was added to the solution. The contents of the beaker were stirred and heated (at <80°C) when necessary to dissolve the salts. A watch glass containing water was placed on top of the beaker to reduce evaporation losses; however, periodically it was necessary to add deionized water to the beaker to replace evaporated water or promote the dissolution of one of the salts. After dissolving the salts, the solutions were transferred to a graduated cylinder. The beaker was rinsed with 3, 1-2 mL aliquots of deionized water. The contents of the graduated cylinder were then diluted to 100 mL. The salt solutions were transferred to plastic bottles and then back into the graduated cylinder to mix the solution and ensure the residual volume remaining in the cylinder was the same composition as the solution transferred to the bottle. This procedure was repeated 3 times. A summary of the target and actual masses of each starting material used to prepare the salt solutions is provided in Appendix B.

During the preparation of the salt solutions, it was necessary to reduce the SO_4^{2-} or CO_3^{2-} concentrations of several solutions to completely dissolve the $Na_2SO_4 \cdot 10H_2O$ and $Na_2CO_3 \cdot H_2O$. The reduction in the target mass of the starting material is noted in Appendix B (see Table B.1) for these solutions. In addition, the final volume of several solutions was slightly greater than 100 mL. During solution preparation, deionized water was added to aid in the dissolution of the salts. After the dissolution of the starting materials, the final volume of the solutions could not be evaporated below 100 mL without the precipitation of solids. The required adjustments in volume are noted in Appendix B, Table B.2. The actual concentrations of the salt solutions and equilibration temperature are given in Table 4.

Table 4 Actual Concentration of Salt Solutions

Solution	OH^- (M)	$Al(OH)_4^-$ (M)	SO_4^{2-} (M)	CO_3^{2-} (M)	NO_3^- (M)	NO_2^- (M)	Temp (°C)
1A	15.00	0.0010	0	0.0010	0.1001	0.1000	25
2A	15.00	0.0010	0	0.0010	0.1001	0.1000	80
3A	5.512	0.0167	0.0159	0.6001	0.7523	0.8154	80
4	0.8730	0.1323	0.0105	0.8878	4.880	1.266	25
5	0.8723	0.1323	0.0104	0.8877	4.880	1.266	80
6	2.268	0.6631	0.2066	0.0591	2.472	2.989	80
7	1.780	0.1133	0.1209	0.3630	0.6262	5.428	80
8	1.478	0.3721	0.3117	0.2152	1.429	3.695	25
9A	3.350	0.6029	0.2136	0.2476	2.536	0.8670	25
10A	2.895	0.7561	0.2500	0.2217	2.989	0.6284	80
11A	5.639	0.1090	0.2416	0.2816	0.3429	0.9360	80
12	2.610	0.0457	0.0833	0.8847	0.1597	3.408	25
13	1.221	0.6012	0.1113	0.0358	4.148	1.815	25
14	0.4537	0.0313	0.1075	0.0726	3.645	3.416	25
15	1.291	0.7406	0.1809	0.9498	2.501	0.1687	25
16	1.291	0.7406	0.1809	0.9498	2.501	0.1687	80
17	2.070	0.0501	0.4671	0.8259	0.2503	0.7944	25
18	0.8611	0.0672	0.1810	0.9498	0.4665	0.1106	80
19	0.6281	0.0067	0.4362	0.4463	0.5151	0.4176	80
20	2.628	0.0246	0.0843	0.0126	0.1596	0.1455	80

Once the salt solutions were prepared, nominally 10 mL of each solution were transferred to 5, 15 mL plastic bottles. The intent of preparing 5 sample bottles for each solution was to allow the removal of a separate bottle each time the solutions were sampled. This procedure eliminated the need to remove the entire solution from a temperature-controlled environment during sampling and allowed the solutions to be maintained at the desired temperature using equipment which was not radioactively contaminated.

The weapons grade Pu added to the salt solutions was initially purified and concentrated by anion exchange. Likewise, Am recovered from the raffinate of a Pu anion exchange column run was purified and concentrated using a chelating resin. The Pu^{4+} and Am^{3+} nitrate solutions were used to prepare a stock solution containing 1000 µg/mL each of Pu and Am in nominally 1M nitric acid (HNO_3). Each of the salt solution sample bottles was spiked with 50 µL of the Pu/Am stock solution. During the Pu/Am additions, the sample bottles were handled in a way to prevent the outside from becoming contaminated. The bottles were surveyed for contamination and placed in 250 mL plastic bottles which provided secondary containment for the solutions. One to 3 sample bottles were placed in each of the bottles. The 250 mL bottles were secured in New Brunswick Scientific Innova 4080 Incubator Shakers to maintain the solutions at the desired temperature. The temperature of the incubator shakers were continuously monitored using calibrated thermistors. Variations in temperature were held to less than ±1°C.

Sampling and Analysis of Salt Solutions

Three sets of the salt solution samples were analyzed at nominally 1 month intervals. To prepare the samples for analysis, 5-8 mL of solution were removed from a sample bottle using a 10 mL disposable syringe with a piece of plastic tubing attached to facilitate reaching into the sample bottle. The solution was expelled through an Acrodisc® 0.45 µm Versapor® membrane disk filter into a clean glass vial. A 1 mL aliquot of the filtered solution was slowly transferred (to reduce the rate of gas evolution) by pipette into a glass sample vial containing 2 or 4 mL of 5M HNO_3. The 4 mL aliquot of acid was used to neutralize salt solution samples containing 15M NaOH; a 2 ml aliquot of acid was used to neutralize the remainder of the samples. The resulting acid concentrations in the samples were 1-3M. The acidified samples were analyzed for Pu (^{238}Pu, ^{239}Pu, and ^{240}Pu) by thenolytrifluoroacetone (TTA) extraction and alpha pulse height analysis (APHA). The Am concentration was measured by gamma pulse height analysis (GPHA).

Since 11 of the salt solutions were equilibrated at 80°C, care was taken during the sampling procedure to ensure that the solutions stayed at or near temperature to prevent the precipitation of Pu and Am as the solutions cooled. To maintain the salt solutions at this temperature, a drying oven was used to preheat to 80°C the syringes, tubing, filter disks, glass vials, and pipette tips used in the sampling procedure. When the sample bottles were removed from the incubator shaker, the bottle(s) not being sampled were placed in the drying oven to maintain the solution(s) at 80°C.

To check the consistency of the Pu and Am analyses, blank and standard Pu/Am solutions were analyzed with each set of salt solutions. The blank solutions were randomly selected samples of the 20 salt solutions which did not contain Pu and Am. A standard solution containing 0.5 µg/mL of Pu and Am was prepared by diluting a 50 µL aliquot of the Pu/Am stock solution with 10 mL of 2M HNO_3 followed by the dilution of a 1 mL aliquot of the resulting solution with 9 mL of 2M HNO_3.

Results and Discussion

The Pu and Am solubilities measured in each of the salt solutions as a function of the equilibration time are presented in Appendix C. The Pu and Am analyses for the blank and standard solutions are also included for each data set. In this section these solubility data are reviewed and their relationship to the 7 factors of this study are explored and modeled. The statistical analyses supporting this review were conducted using JMP Version 5 from SAS Institute, Inc.[8]

Exhibit D.1 in Appendix D provides plots of the Pu and Am data by sample number. As seen in these plots, the behavior of these solubility data over time is somewhat erratic, which brings into question the equilibrium of the Pu and Am in the solution and raises the question of how to best represent the solubilities for each of these experimental trials. The first question is being addressed by additional samples being taken at two times in the future to provide more data to assess the stability of these solutions. The second question is addressed in the preliminary

analysis by using each of the individual 3 samples and their average to represent the solubility data for each of the 20 trials during the statistical modeling process.

Exhibit D.2 in Appendix D provides plots of the Pu and Am data by each factor for each of the 3 samples and their average. As discussed above, a first-order model in the 7 factors was of interest for exploring the relationship between the factors and Am solubility. For completeness, the Pu solubility data was modeled in the same manner. Exhibit D.3 in Appendix D provides the (summary) results generated by the JMP software used to conduct these fits. As seen in Exhibit D.3, the R^2 values for these fits ran from a low of 60% to a high of 84%. Table 5 was prepared to allow for easy comparisons across these results. The estimated effect of each of the factors for each of the first-order models of Exhibit D.3 is provided in this table, where shaded entries are statistically significant with at least 90% confidence.

Table 5 Summary of First-Order Models Fit to the Solubility Data from This Study

First-Order Model	Intercept	Temp (°C)	OH^- (M)	$Al(OH)_4^-$ (M)	SO_4^{2-} (M)	CO_3^{2-} (M)	NO_3^- (M)	NO_2^- (M)
Sample 1 Pu	-3.79E-7	-1.68E-8	1.30E-6	-1.74E-7	1.21E-7	5.20E-6	4.25E-8	4.08E-7
Sample 2 Pu	1.40E-6	-3.59E-8	1.20E-6	1.20E-6	-7.23E-7	5.00E-6	-1.78E-7	3.38E-7
Sample 3 Pu	-2.00E-6	-1.23E-8	1.20E-6	3.00E-6	3.50E-6	5.90E-6	7.69E-7	1.48E-7
Average Pu	-2.05E-7	-2.16E-8	1.30E-6	-5.06E-7	9.64E-7	5.40E-6	2.11E-7	2.98E-7
Sample 1 Am	3.65E-7	-1.76E-8	4.56E-7	2.03E-7	9.57E-7	1.00E-6	3.57E-9	1.55E-7
Sample 2 Am	1.80E-6	-2.87E-8	2.58E-7	2.00E-6	-3.19E-7	1.70E-6	-1.46E-7	1.68E-7
Sample 3 Am	-1.00E-6	-6.40E-9	4.82E-7	-2.00E-6	2.20E-6	2.60E-6	9.52E-7	-1.21E-8
Average Am	3.09E-7	-1.76E-8	3.98E-7	1.70E-7	9.41E-7	1.80E-6	2.70E-7	1.03E-7

Note: Shaded entries are statistically significant with at least 90% confidence.

Since a first-order model was of primary interest for the Am solubility data, one additional fit was conducted for the Am data using JMP. In this fit, the average Am solubility data were used to represent the response, and this response was modeled using Temperature, OH^-, CO_3^{2-}, and NO_3^-, all of the terms that showed as significant for a least one of the models in Exhibit D.3. The results from this fit are provided in Exhibit D.4 of Appendix D. The R^2 value of this fit is ~82%, all of the terms of the model are statistically significant with at least 90% confidence, and the fitted model for Am solubility is given by equation 4.

$$\mathrm{Am(M)} = 1.10\text{E-}6 - 1.91\text{E-}8\,\mathrm{Temp} + 3.54\text{E-}7\left[\mathrm{OH^-}\right] + 1.60\text{E-}6\left[\mathrm{CO_3^{2-}}\right] + 2.26\text{E-}7\left[\mathrm{NO_3^-}\right] \quad (4)$$

Thus, the model given by equation 4 provides the starting place for understanding, through this data-driven approach, the relationship of Am solubility to the factors of Table 1. After the remaining two samples are taken from the 20 experimental trials for this study, the data from those samples will be combined with the sample data presented in this report and the models fitting process will be repeated.

Once again for completeness, a similar first-order fit was conducted for the Pu data using JMP. In this fit, the average Pu solubility data were used to represent the response, and this response was modeled using only OH^- and CO_3^{2-}, the only terms that showed as significant for a least one of the models in Exhibit D.3. The results from this fit are provided in Exhibit D.5 of

Appendix D. The R^2 value of this fit is ~75%, both factors are statistically significant with at least 90% confidence, and the fitted model for Pu solubility is given by equation 5.

$$\text{Pu (M)} = \text{-7.48E-8} + \text{1.20E-6}\left[OH^-\right] + \text{4.90E-6}\left[CO_3^{2-}\right] \quad (5)$$

Although equation 5 provides what is felt to be the best first-order model for the Pu solubility data from this study, the primary goal for the modeling of Pu solubility was to combine available historical data with the current results to allow for the study of a more complex model form (i.e., a modified response surface model, as described above). The plots of Exhibit D.6 in Appendix D are presented as a prelude to this modeling effort. These plots show the Pu results from this study (all 3 samples and the sample average) versus the 7 factors of interest. Superimposed on these plots are the historical Pu solubility data. Different colors and symbols are used to represent the historical data sets as well as the current results. A dramatic feature of these plots is seen in the behavior of one of the previous data sets relative to the other plotted points. Points from this data set are represented by orange triangles in these plots, and the structure of the relationship between Pu solubility and the factors under study as revealed in these plots is often very different from that of the other data. These data were from a study by Delegard [3] and it is believed that the apparent differences in these data relative to the other data are due to other complexants being involved in Delegard's study. As a consequence, these data were excluded from the modeling efforts of this study.

Exhibit D.7 in Appendix D provides the results from the fit of a modified response surface model in the 7 factors (with the quadratic temperature term removed) to historical data complemented in turn by the Pu solubility data for each of the 3 samples and by the averages for the 20 trials. The R^2 values for these fitted models are all over 99.5%. Such high R^2 values may be due in part to the large number of candidate terms available to account for the variation in the solubility data. Table 6 was prepared to facilitate a closer look at the results for each of the fitted models. It provides a listing of the candidate terms and their estimated coefficients for each of the fits. A shaded entry indicates a term in a model that is statistically significant with at least 90% confidence. Even with the sample-to-sample variation in the Pu solubility measurements, there is some consistency in the terms that show up as statistically significant across these models.

Table 6 Summary of Modified Response Surface Models Fit to the Historical and Current Pu Solubility Data

Estimated Coefficient	Sample 1	Sample 2	Sample 3	Average
Intercept	1.00E-06	6.07E-07	4.00E-07	7.15E-07
$Al(OH)_4^-$	8.00E-06	1.90E-05	2.00E-06	9.00E-06
CO_3^{2-}	-1.00E-05	-8.96E-06	-2.00E-05	-1.00E-05
NO_2^-	-8.82E-07	-7.48E-07	-2.08E-07	-6.13E-07
NO_3^-	-6.88E-07	-7.34E-07	-2.97E-07	-5.73E-07
OH^-	2.94E-07	-6.57E-07	-2.39E-09	-1.22E-07
SO_4^{2-}	1.00E-06	-2.00E-05	5.00E-06	-3.37E-06
Temperature	-3.43E-08	8.46E-09	-1.48E-08	-1.35E-08
$CO_3^{2-}*Al(OH)_4^-$	-3.19E-06	-9.89E-06	-5.29E-06	-6.12E-06
$NO_2^-*Al(OH)_4^-$	3.72E-07	-1.92E-06	5.84E-07	-3.21E-07
$NO_3^-*Al(OH)_4^-$	-3.98E-06	-7.31E-06	-2.07E-06	-4.45E-06
$OH^-*Al(OH)_4^-$	-1.77E-06	-3.75E-06	1.00E-06	-1.36E-06
$SO_4^{2-}*Al(OH)_4^-$	6.00E-06	4.20E-05	6.00E-06	1.80E-05
Temperature$*Al(OH)_4^-$	-1.04E-07	-1.14E-07	-1.52E-07	-1.23E-07
$NO_2^-*CO_3^{2-}$	2.00E-06	1.00E-06	5.13E-07	1.00E-06
$NO_3^-*CO_3^{2-}$	5.84E-07	3.19E-07	3.00E-06	1.00E-06
$OH^-*CO_3^{2-}$	4.00E-06	4.00E-06	5.00E-06	4.00E-06
$SO_4^{2-}*CO_3^{2-}$	1.20E-05	1.90E-05	1.80E-05	1.60E-05
Temperature$*CO_3^{2-}$	2.82E-08	7.32E-09	3.22E-08	2.26E-08
$NO_3^-*NO_2^-$	1.62E-07	-1.41E-07	1.08E-07	4.30E-08
$OH^-*NO_2^-$	4.35E-08	8.03E-07	3.59E-07	4.02E-07
$SO_4^{2-}*NO_2^-$	1.00E-06	6.00E-06	-6.42E-07	2.00E-06
Temperature$*NO_2^-$	3.38E-09	-5.62E-09	-1.02E-08	-4.13E-09
$OH^-*NO_3^-$	2.68E-07	2.32E-07	2.16E-07	2.39E-07
$SO_4^{2-}*NO_3^-$	-1.34E-06	-1.17E-06	-1.71E-06	-1.41E-06
Temperature$*NO_3^-$	-9.40E-10	-1.95E-08	8.93E-09	-3.84E-09
$SO_4^{2-}*OH^-$	7.12E-07	5.49E-07	5.18E-08	4.38E-07
Temperature$*OH^-$	8.00E-09	5.87E-09	5.91E-09	6.60E-09
Temperature$*SO_4^{2-}$	8.71E-08	1.39E-07	1.35E-08	7.98E-08
$Al(OH)_4^-*Al(OH)_4^-$	1.70E-05	2.10E-05	1.00E-05	1.60E-05
$CO_3^{2-}*CO_3^{2-}$	8.00E-06	3.00E-06	1.00E-05	7.00E-06
$NO_2^-*NO_2^-$	-7.29E-09	-1.85E-07	7.09E-08	-4.05E-08
OH^-*OH^-	2.97E-08	8.20E-08	3.91E-08	5.03E-08
$SO_4^{2-}*SO_4^{2-}$	-1.00E-05	-2.94E-06	-5.37E-06	-6.14E-06
$NO_3^-*NO_3^-$	2.98E-07	7.25E-07	4.27E-08	3.55E-07

Note: Shaded entries are statistically significant with at least 90% confidence.

To gain a better understanding of which of the terms in Table 6 are important for modeling the relationship of interest in this study, a data-driven approach was used. JMP's stepwise regression procedure was used to select an "optimal" model for the historical data complemented by the average Pu data from this study, where all of the second order terms were significant to at

least the 10% level. Exhibit D.7 in Appendix D provides the results from the fitting of this model to the historical data complemented by the average Pu data from this study. The exhibit also presents the results of the fitting of this same model to the historical data complemented in turn by the Pu solubility data from this study for each of the 3 samples. Even though the R^2 values for all of these fits are above 97.8%, there is an indication of a statistically significant lack of fit for each of the models. This is indicated by the small p-values for the "Lack of Fit" portion of the exhibit for each of the models. Table 7 is presented to facilitate comparisons among these fitted models. Shading is used to highlight model terms that are statistically significant with at least 90% confidence. Note a main effect for a factor is shaded if the factor is involved in statistically significant interactions or quadratic effects. Since the average Pu data from this study were used to drive the stepwise regression model selection, all of the factor terms for this model are significant (shaded) in Table 7. There is, however, some consistency in the terms deemed statistically significant across these four models.

Table 7 Summary of "Optimal" Stepwise Regression Model Fit to the Historical and Current Pu Solubility Data

Estimated Coefficient	Sample 1	Sample 2	Sample 3	Average
Intercept	-4.93E-07	5.31E-07	-3.43E-07	-1.02E-07
$Al(OH)_4^-$	6.05E-07	1.98E-07	5.10E-06	2.00E-06
CO_3^{2-}	-1.50E-05	-9.00E-06	-2.10E-05	-1.50E-05
NO_2^-	-1.56E-07	-4.02E-07	-1.53E-07	-2.37E-07
NO_3^-	7.71E-07	7.86E-07	7.16E-07	7.58E-07
OH^-	8.37E-07	6.51E-07	7.14E-07	7.34E-07
SO_4^{2-}	7.89E-07	-4.00E-06	-2.00E-06	-2.00E-06
Temperature	-2.72E-08	-5.75E-08	-2.14E-08	-3.54E-08
CO_3^{2-}*$Al(OH)_4^-$	-2.00E-06	-6.28E-07	-9.00E-06	-4.00E-06
NO_2^-*$Al(OH)_4^-$	1.00E-06	2.00E-06	4.28E-07	1.10E-06
NO_3^-*$Al(OH)_4^-$	-2.00E-06	-1.00E-06	-3.00E-06	-2.00E-06
SO_4^{2-}*$Al(OH)_4^-$	4.60E-06	1.92E-05	1.49E-05	1.29E-05
Temperature*$Al(OH)_4^-$	-1.02E-07	-1.08E-07	-1.39E-07	-1.16E-07
NO_2^-*CO_3^{2-}	1.70E-06	1.50E-06	1.30E-06	1.50E-06
NO_3^-*CO_3^{2-}	9.18E-07	5.52E-07	2.50E-06	1.30E-06
OH^-*CO_3^{2-}	3.30E-06	3.20E-06	4.60E-06	3.70E-06
SO_4^{2-}*CO_3^{2-}	1.18E-05	5.00E-06	2.01E-05	1.23E-05
Temperature*CO_3^{2-}	2.68E-08	-3.39E-09	4.45E-08	2.26E-08
SO_4^{2-}*NO_3^-	-1.00E-06	-1.00E-06	-1.00E-06	-1.00E-06
Temperature*OH^-	7.96E-09	1.05E-08	5.93E-09	8.14E-09
Temperature*SO_4^{2-}	4.98E-08	1.66E-07	6.08E-08	9.22E-08
$Al(OH)_4^-$*$Al(OH)_4^-$	1.29E-05	6.60E-06	1.28E-05	1.08E-05
CO_3^{2-}*CO_3^{2-}	9.10E-06	7.30E-06	1.04E-05	8.90E-06

Note: Shaded entries are statistically significant with at least 90% confidence.

Between the extremes of the simple first-order model given by equation 5 and the more complex models provided in Table 7, these results suggest the potential for a data driven approach to gain insight into the relationship of Pu solubility to the factors defined in Table 1. After the remaining two samples are taken from the 20 experimental trials for this study, the data from those samples will be combined with the sample data presented in this report and the models fitting process will be repeated.

Conclusions and Recommendations

Preliminary results from a series of small-scale experiments were used to model Pu and Am solubilities with salt concentrations and temperatures representative of the ranges expected in SRS waste tanks. A statistically designed experiment was used to prepare 20 solutions with varying salt concentrations (OH^-, $Al(OH)_4^-$, SO_4^{2-}, CO_3^{2-}, NO_3^-, and NO_2^-) which were equilibrated at either 25 or 80°C. Three sets of salt solutions were analyzed at nominally 1 month intervals. Analysis of the solubility data showed no time-dependence of the Pu and Am concentrations; however, the data scatter was large enough that one cannot conclude that equilibrium concentrations were reached. For this reason, the analysis of 2 additional sets of solutions prepared as part of this study is recommended.

The 3 data sets were initially used to develop first-order models for both Pu and Am solubility. Since the equilibrium concentrations may not have been reached, preliminary models were developed using the analyzed concentrations for each of the individual samples and their average to represent the solubility of each element. In this analysis, Pu solubility was found to be only a function of OH^- and CO_3^{2-} concentrations; Am solubility was found to be a function of OH^-, CO_3^{2-}, and NO_3^- concentrations and temperature. These factors were statistically significant with at least 90% confidence. A comparison of the coefficients for the multiple Pu and Am models showed small changes in the values; however, the trends (sign and relative magnitude) were generally the same.

The solubility data obtained for Pu in this study were combined with historical data from the literature to develop a modified response surface model. As with the first-order model for Pu, response surface models were developed using the analyzed concentrations for each of the individual samples and their average to represent the solubility of Pu obtained in this study. Even with the sample-to-sample variation in the measurements, there was some consistency in the terms that were statistically significant across the models. Statistical methods were then used to reduce the number of model terms from 35 (the quadratic temperature term was not allowed) to 23 which were significant with at least 90% confidence. All of the salt concentrations and temperature terms were statistically significant. Even with the reduction in terms, the correlation coefficient (R^2) was still very good (97.8%). However, with this number of terms and a limited amount of data, it is difficult to distinguish whether a term is truly significant or is overly sensitive to variations in the solubility data. With better estimates of the Pu solubility for the 20 design points, additional analysis can be used to re-evaluate the significance of the model terms and complete an evaluation of parameter sensitivity and validate the predictive performance.

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Appendix A Tables and Exhibits Supporting the Development of the Test Matrix

Table A1. Historical Pu Solubility Data Based Upon Previous Studies

Data Set	$Al(OH)_4^-$	CO_3^{2-}	NO_2^-	NO_3^-	OH^-	SO_4^{2-}	Temperature	Pu Solubility	Constraints Met
	(M)	(M)	(M)	(M)	(M)	(M)	(M)	(M)	
1[1]	0	0	0	4	4	0	22	1.26E-06	yes
1	0	0	0	1	5	0	22	1.34E-06	yes
1	0	0	0	1.5	5	0	22	1.58E-06	yes
1	0	0	0	2	5	0	22	1.68E-06	yes
1	0	0	0	3	5	0	22	2.48E-06	yes
1	0	0	0	3	5	0	22	1.92E-06	yes
1	0	0	0	4	5	0	22	3.00E-06	yes
1	0	0	0	2	6	0	22	4.74E-06	yes
1	0	0	0	1	7	0	22	7.93E-06	yes
1	0	0	4	0	4	0	22	4.00E-07	yes
1	0	0	1	0	5	0	22	1.17E-06	yes
1	0	0	1.5	0	5	0	22	9.20E-07	yes
1	0	0	2	0	5	0	22	5.50E-07	yes
1	0	0	3	0	5	0	22	1.12E-06	yes
1	0	0	3	0	5	0	22	7.00E-07	yes
1	0	0	4	0	5	0	22	7.80E-07	yes
1	0	0	2	0	6	0	22	5.54E-06	yes
1	0	0	1	0	7	0	22	5.09E-06	yes
1	0	0	1	1	1	0	22	3.00E-08	yes
1	0	0	1	1	2	0	22	1.20E-07	yes
1	0	0	1	1	3	0	22	3.20E-07	yes
1	0	0	1	1	4	0	22	7.70E-07	yes
1	0	0	0.25	0.25	5	0	22	2.01E-06	yes
1	0	0	0.5	0.5	5	0	22	2.85E-06	yes
1	0	0	1	1	5	0	22	3.86E-06	yes
1	0	0	1	1	5	0	22	2.09E-06	yes
1	0	0	1.5	1.5	5	0	22	6.49E-06	yes
1	0	0	2	2	5	0	22	8.71E-06	yes
1	0	0	2.5	2.5	5	0	22	8.47E-06	yes
1	0	0	3	3	5	0	22	1.48E-05	yes
1	0	0	3	3	5	0	22	1.48E-05	yes
1	0	0	1	1	6	0	22	4.60E-06	yes
1	0	0	2.5	2.5	6	0	22	3.82E-05	yes
1	0	0	1	1	7	0	22	1.03E-05	yes
1	0	0	2	2	7	0	22	9.41E-06	yes
1	0	0	1	1	8	0	22	2.99E-05	yes
1	0	0	1.5	1.5	8	0	22	8.44E-05	yes
1	0	0	1	1	9	0	22	1.07E-04	yes
1	0	0	1	1	9	0	22	6.67E-05	yes
1	0	0	0.5	0.5	10	0	22	2.69E-04	yes
1	0	0	1	1	10	0	22	1.45E-04	yes
1	0	0	0.25	0.25	10.5	0	22	1.84E-04	yes

[1] Dataset number 1 is from reference [9].

Table A1. Historical Pu Solubility Data Based Upon Previous Studies (continued)

Data Set	$Al(OH)_4^-$	CO_3^{2-}	NO_2^-	NO_3^-	OH^-	SO_4^{2-}	Temperature	Pu Solubility	Constraints Met
	(M)	(M)	(M)	(M)	(M)	(M)	(°C)	(M)	
2[2]	0.00005	0.025	1	2	4	0.005	22	4.58E-06	yes
2	0.00005	0.025	1	2	1	0.005	22	1.04E-06	yes
2	0.00005	0.025	1	2E-08	4	0.005	22	2.07E-06	no
2	0.00005	0.025	1	2E-08	1	0.005	22	4.80E-07	no
2	0.5	0.025	1	2	2	0.005	22	3.39E-06	yes
2	5.00E-09	0.025	1	2	2	0.005	22	1.29E-06	yes
2	0.5	0.025	1	2E-08	2	0.005	22	1.24E-06	no
2	5.00E-09	0.025	1	2E-08	2	0.005	22	5.70E-07	yes
2	0.5	0.025	1	0.0002	4	0.005	22	3.31E-06	no
2	5.00E-09	0.025	1	0.0002	4	0.005	22	2.34E-06	yes
2	0.5	0.025	1	0.0002	1	0.005	22	1.00E-06	no
2	5.00E-09	0.025	1	0.0002	1	0.005	22	3.70E-07	yes
2	0.00005	0.025	1	0.0002	2	0.005	22	5.70E-07	yes
2	0.00005	0.025	1	0.0002	2	0.005	22	5.10E-07	yes
2	0.00005	0.025	1	0.0002	2	0.005	22	7.40E-07	yes
3[3]	0.05	0.02	2	1	2	0.4	60	1.58E-06	yes
3	0.19	0.16	1.05	2.5	1.25	0.21	60	2.70E-07	yes
3	0.19	0.16	1.05	2.5	1.25	0.21	60	2.90E-07	yes
3	0.19	0.16	1.05	2.5	1.25	0.21	25	5.00E-07	yes
3	0.33	0.3	0.1	4	0.5	0.02	25	1.22E-06	no
3	0.363	0.02	2	4.96	0.75	0.02	25	3.50E-07	no
3	0.33	0.02	0.1	1	2	0.4	25	3.07E-06	yes
3	0.33	0.3	2	1	2	0.02	25	1.30E-06	yes
3	0.05	0.3	0.1	1	2	0.02	60	5.30E-07	yes
3	0.33	0.02	2	4	0.5	0.4	25	2.60E-07	no
3	0.33	0.02	0.1	4	2	0.02	60	6.70E-07	yes
3	0.05	0.02	0.1	1	0.5	0.02	25	7.00E-08	yes
3	0.19	0.16	1.05	2.5	1.25	0.21	25	4.30E-07	yes
3	0.23	0.21	1.4	2.8	1.4	0.28	60	6.70E-07	yes
3	0.05	0.3	2	4	0.5	0.02	60	6.10E-07	yes
3	0.05	0.02	0.1	4	0.5	0.4	60	8.00E-08	yes
3	0.33	0.02	2	1	0.5	0.02	60	7.00E-08	no
3	0.5	0.3	2	1	0.5	0.4	25	1.80E-07	no
3	0.33	0.3	0.1	1	0.5	0.4	60	5.30E-07	no
3	0.05	0.3	0.1	4	2	0.4	25	4.09E-06	yes
3	0.33	0.3	0.1	1	0.5	0.4	22	1.20E-07	no
4[4]	0	0	0	1	4.1E-07	0	29.6	1.30E-07	no
4	0	0	0	1	7.2E-07	0	29.6	1.40E-07	no
4	0	0	0	1	1.05E-06	0	29.6	5.00E-07	no
4	0	0	0	1	6.17E-06	0	29.6	1.00E-07	no
4	0	0	0	1	1.23E-05	0	29.6	8.00E-08	no
4	0	0	0	1	1.74E-05	0	29.6	1.40E-07	no
4	0	0	0	1	1.74E-05	0	29.6	1.10E-07	no
4	0	0	0	1	2.51E-05	0	29.6	7.00E-08	no
4	0	0	0	1	0.000038	0	29.6	4.00E-07	no

2 Dataset number 2 is from reference [3].

3 Dataset number 3 is from reference [1].

4 Dataset number 4 is from reference [10].

Table A1. Historical Pu Solubility Data Based Upon Previous Studies (continued)

Data Set	$Al(OH)_4^-$	CO_3^{2-}	NO_2^-	NO_3^-	OH^-	SO_4^{2-}	Temperature	Pu Solubility	Constraints Met
	(M)	(M)	(M)	(M)	(M)	(M)	(°C)	(M)	
4	0	0	0	1	3.98E-05	0	29.6	1.70E-07	no
4	0	0	0	1	5.37E-05	0	29.6	4.00E-08	no
4	0	0	0	1	7.94E-05	0	29.6	3.00E-07	no
4	0	0	0	1	0.000148	0	29.6	1.90E-07	no
4	0	0	0	1	0.000339	0	29.6	2.20E-07	no
4	0	0	0	1	0.000347	0	29.6	3.00E-08	no
4	0	0	0	1	0.000468	0	29.6	4.00E-08	no
4	0	0	0	1	0.000617	0	29.6	6.00E-08	no
4	0	0	0	1	0.000741	0	29.6	2.00E-08	no
4	0	0	0	1	0.00275	0	29.6	8.00E-08	no
4	0	0	0	1	0.00724	0	29.6	3.70E-07	no
4	0	0	0	1	0.00741	0	29.6	7.00E-08	no
4	0	0	0	1	0.0102	0	29.6	1.50E-07	no
4	0	0	0	1	0.0132	0	29.6	1.40E-07	no
4	0	0	0	1	0.0132	0	29.6	1.60E-07	no
4	0	0	0	1	0.0427	0	29.6	2.00E-07	no
4	0	0	0	1	0.0759	0	29.6	7.00E-07	no
4	0	0	0	1	0.0871	0	29.6	1.50E-07	no
4	0	0	0	1	0.0891	0	29.6	1.00E-07	no
4	0	0	0	1	0.102	0	29.6	2.00E-08	no
4	0	0	0	1	0.11	0	29.6	3.30E-07	no
4	0	0	0	1	0.288	0	29.6	7.00E-08	no
4	0	0	0	1	0.479	0	29.6	3.00E-08	yes
4	0	0	0	1	0.501	0	29.6	1.00E-07	yes
4	0	0	0	1	0.575	0	29.6	4.00E-08	yes
4	0	0	0	1	0.589	0	29.6	1.90E-07	yes
4	0	0	0	1	0.646	0	29.6	5.00E-08	yes
5[5]	0	0	0.125	0.875	2.6E-07	0	30.4	4.00E-08	no
5	0	0	0.125	0.875	5E-07	0	30.4	9.30E-09	no
5	0	0	0.125	0.875	2.69E-06	0	30.4	6.10E-09	no
5	0	0	0.125	0.875	0.00724	0	30.4	4.50E-09	no
5	0	0	0.125	0.875	0.117	0	30.4	5.20E-09	no
5	0	0	0.125	0.875	0.224	0	30.4	5.00E-08	no
5	0	0	0.5	3.5	2E-07	0	30.4	3.90E-09	no
5	0	0	0.5	3.5	8.91E-06	0	30.4	3.00E-08	no
5	0	0	0.5	3.5	0.000741	0	30.4	3.70E-09	no
5	0	0	0.5	3.5	0.0117	0	30.4	1.80E-08	no
5	0	0	0.5	3.5	0.166	0	30.4	2.00E-08	no
6[6]	0.33	0.3	0.1	1	0.051	0.4	22	1.24E-06	no
6	0.33	0.3	0.1	1	0.051	0.4	22	1.67E-06	no

5 Dataset number 5 is from reference [11].

6 Dataset number 6 is from reference [1].

Table A.2 Additional Set of Selective Candidate Points

$Al(OH)_4^-$ (M)	CO_3^{2-} (M)	NO_2^- (M)	NO_3^- (M)	OH^- (M)	SO_4^{2-} (M)	Temperature (°C)
0.001	0.001	0.1	0.1	15	0.001	25
0.001	0.001	0.1	0.1	15	0.001	80
0.001	0.001	0.1	0.1	12	0.01	25
0.001	0.01	0.1	0.1	12	0.001	25
0.01	0.001	0.1	0.1	12	0.001	25
0.001	0.01	0.1	0.1	12	0.01	25
0.01	0.001	0.1	0.1	12	0.01	25
0.01	0.01	0.1	0.1	12	0.001	25
0.01	0.01	0.1	0.1	12	0.01	25
0.001	0.001	0.1	0.1	12	0.01	80
0.001	0.01	0.1	0.1	12	0.001	80
0.01	0.001	0.1	0.1	12	0.001	80
0.001	0.01	0.1	0.1	12	0.01	80
0.01	0.001	0.1	0.1	12	0.01	80
0.01	0.01	0.1	0.1	12	0.001	80
0.01	0.01	0.1	0.1	12	0.01	80
0.1	0.01	0.1	0.1	9	0.01	25
0.01	0.1	0.1	0.1	9	0.01	25
0.1	0.1	0.1	0.1	9	0.01	25
0.1	0.01	0.1	0.1	9	0.01	80
0.01	0.1	0.1	0.1	9	0.01	80
0.1	0.1	0.1	0.1	9	0.01	80

Table A.3 Initial Set of 8 Design Points

Type of Point	$Al(OH)_4^-$ (M)	CO_3^{2-} (M)	NO_2^- (M)	NO_3^- (M)	OH^- (M)	SO_4^{2-} (M)	Temperature (°C)
Modified OLH	0.74057	0.949663	0.168605	2.501163	1.290783	0.180872	25
Modified OLH	0.031278	0.072562	3.415891	3.644574	0.453583	0.107376	25
Modified OLH	0.049969	0.825756	0.794186	0.25	2.069833	0.46712	25
Modified OLH	0.132317	0.887709	1.266279	4.879457	0.872183	0.010369	85
Modified OLH	0.756058	0.221709	0.628295	2.98876	2.895417	0.482593	85
Modified OLH	0.113337	0.362678	5.428295	0.625969	1.77915	0.120932	85
Modified OLH	0.024584	0.012616	0.145349	0.159302	2.627963	0.084167	85
Special Design Point	0.001	0.001	0.1	0.1	15	0.001	25

Exhibit A.1 Correlations and Scatterplot Matrix for OLH Candidate Design Points

Multivariate
Correlations

	$Al(OH)_4^-$	CO_3^{2-}	NO_2^-	NO_3^-	OH^-	SO_4^{2-}	Temperature
$Al(OH)_4^-$	1.0000	-0.0740	0.1591	0.5231	-0.1779	-0.0340	-0.0000
CO_3^{2-}	-0.0740	1.0000	-0.2093	-0.1408	0.0311	-0.0041	-0.0000
NO_2^-	0.1591	-0.2093	1.0000	0.3091	-0.2249	-0.1020	-0.0000
NO_3^-	0.5231	-0.1408	0.3091	1.0000	-0.4179	-0.0485	0.0000
OH^-	-0.1779	0.0311	-0.2249	-0.4179	1.0000	-0.1424	0.0000
SO_4^{2-}	-0.0340	-0.0041	-0.1020	-0.0485	-0.1424	1.0000	-0.0000
Temperature	-0.0000	-0.0000	-0.0000	0.0000	0.0000	-0.0000	1.0000

Scatterplot Matrix

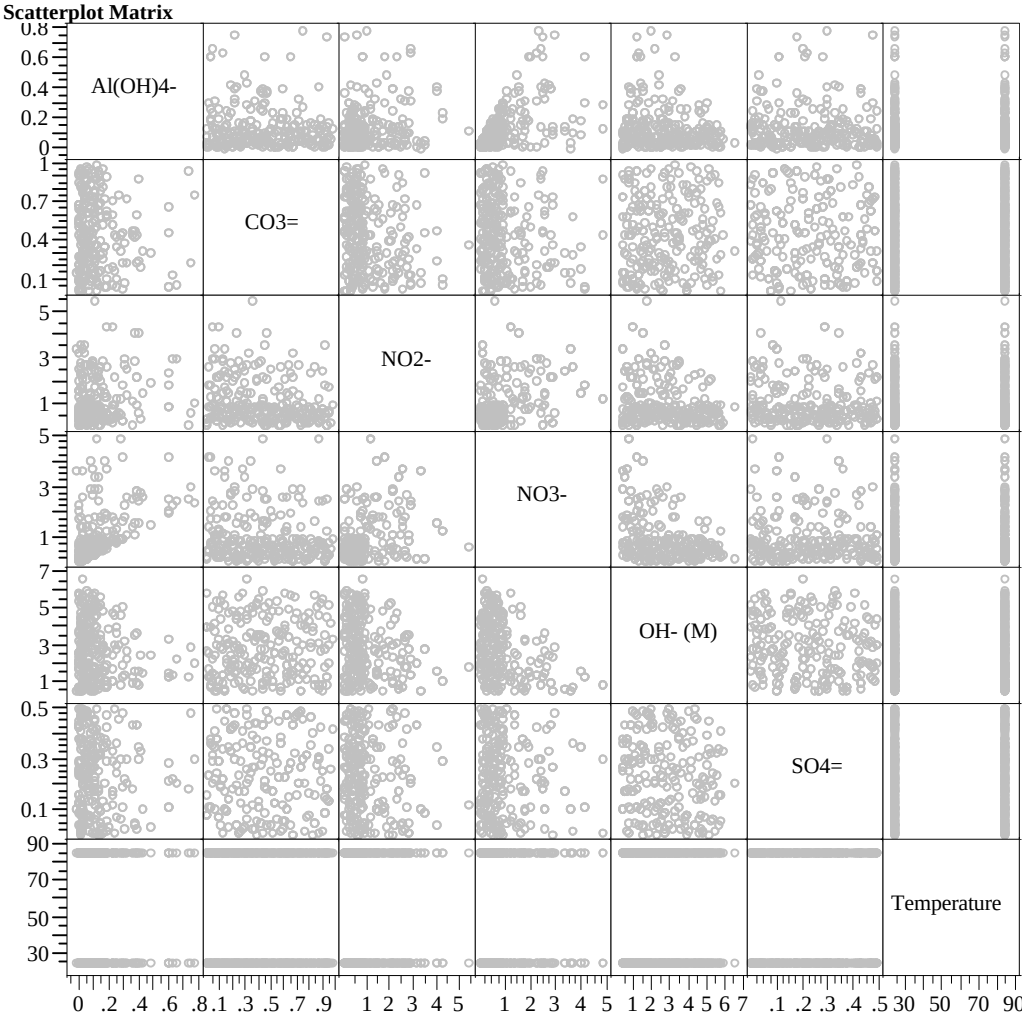


Exhibit A.2 Information Generated by the D-Optimality Routine in Selecting 8 Design Points for Am Solubility Investigation

N Desired	8	
N Random	5	
K Value	3	
Trips	1000	
N	8	------ Ready ------
Trips	1000	

Best Design

D-efficiency	46.4267
A-efficiency	25.1801
G-efficiency	73.8521
AvgPredSE	0.9047
N	8.0000

Correlations

Corr	Intercept	$Al(OH)_4^-$	CO_3^{2-}	NO_2^-	NO_3^-	OH^-	SO_4^{2-}	Temperature
Intercept	1	0.1032	0.3756	0.6695	0.378	0.8462	0.3972	0.1782
$Al(OH)_4^-$	0.1032	1	-0.1446	0.2242	-0.4084	-0.1067	-0.5036	-0.1666
CO_3^{2-}	0.3756	-0.1446	1	0.2425	-0.0578	0.4478	0.0538	0.2962
NO_2^-	0.6695	0.2242	0.2425	1	0.0459	0.4493	0.1429	-0.0289
NO_3^-	0.378	-0.4084	-0.0578	0.0459	1	0.4285	0.4506	0.0412
OH^-	0.8462	-0.1067	0.4478	0.4493	0.4285	1	0.4103	0.3316
SO_4^{2-}	0.3972	-0.5036	0.0538	0.1429	0.4506	0.4103	1	0.1649
Temperature	0.1782	-0.1666	0.2962	-0.0289	0.0412	0.3316	0.1649	1

Exhibit A.3 Information Generated by the D-Optimality Routine in Selecting Full Set of Design Points for Pu Solubility Investigation

Optimal Design Controls		
N Desired	91	
N Random	18	
K Value	3	
Trips	1000	
N	91	------ Ready ------
Trips	1000	

Best Design	
D-efficiency	4.3781
A-efficiency	0.4286
G-efficiency	24.3801
AvgPredSE	1.1516
N	91.0000

Exhibit A.3 Information Generated by the D-Optimality Routine in Selecting Full Set of Design Points for Pu Solubility Investigation (continued)

Correlations of Model Terms

	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)	(12)	(13)	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	(24)	(25)	(26)	(27)	(28)	(29)	(30)	(31)	(32)	(33)	(34)	(35)
(1) Intercept	1.000	0.725	0.376	0.395	0.305	0.921	-0.115	0.143	-0.499	0.532	0.320	0.347	0.138	-0.050	0.356	-0.084	0.224	0.068	0.749	0.114	0.151	0.030	0.148	-0.041	0.088	-0.107	0.103	-0.178	0.141	0.095	0.189	0.162	-0.130	0.006	0.005
(2)$Al(OH)_4^-$	0.725	1.000	0.042	0.163	-0.038	0.667	-0.418	-0.045	-0.483	0.705	0.330	0.554	0.065	-0.194	0.491	-0.394	0.003	-0.092	0.955	-0.253	-0.080	-0.255	-0.066	0.007	-0.228	-0.327	-0.125	-0.279	0.415	0.180	0.210	0.128	-0.332	-0.091	-0.287
(3) CO_3^{2-}	0.376	0.042	1.000	0.084	0.175	0.211	0.024	-0.014	-0.352	0.221	0.429	0.224	0.410	-0.146	-0.061	0.501	0.033	0.066	-0.019	0.829	-0.114	0.042	0.164	0.329	0.699	-0.262	0.106	-0.282	-0.039	-0.148	0.084	-0.059	-0.208	-0.262	0.280
(4) NO_2^-	0.395	0.163	0.084	1.000	-0.116	0.300	-0.002	0.337	-0.014	0.137	-0.154	0.051	0.388	0.157	-0.039	-0.187	0.431	0.300	0.131	-0.084	0.849	-0.351	-0.072	-0.030	0.009	0.326	-0.040	-0.243	0.021	0.073	0.310	0.311	-0.076	0.272	0.083
(5) NO_3^-	0.305	-0.038	0.175	-0.116	1.000	0.281	0.076	0.057	-0.306	0.155	0.162	0.105	-0.133	0.232	0.479	0.202	0.368	-0.032	0.006	0.120	-0.320	0.815	0.239	-0.191	-0.003	-0.179	0.391	0.134	0.067	-0.063	-0.168	-0.117	0.274	0.056	0.138
(6) OH^-	0.921	0.667	0.211	0.300	0.281	1.000	-0.118	0.057	-0.292	0.356	0.252	0.204	-0.027	0.052	0.283	-0.075	0.142	0.079	0.731	0.098	0.174	0.115	0.339	-0.188	-0.051	-0.024	0.016	-0.079	0.048	0.112	0.110	0.110	-0.019	-0.006	-0.035
(7) SO_4^{2-}	-0.115	-0.418	0.024	-0.002	0.076	-0.118	1.000	-0.203	0.067	-0.234	0.233	-0.254	0.067	-0.196	-0.243	0.132	-0.083	0.013	-0.374	-0.049	-0.002	0.082	0.193	0.187	0.378	0.338	0.574	0.693	0.049	-0.382	0.051	0.012	-0.078	-0.288	-0.014
(8) Temp.	0.143	-0.045	-0.014	0.337	0.057	0.057	-0.203	1.000	-0.071	-0.033	-0.116	-0.150	-0.028	0.117	-0.082	-0.108	0.277	0.244	0.018	-0.040	0.359	-0.035	-0.216	-0.080	0.046	0.197	-0.051	-0.382	-0.207	0.146	0.218	0.534	0.329	0.798	0.495
(9) $Al(OH)_4^-$*$Al(OH)_4^-$	-0.499	-0.483	-0.352	-0.014	-0.306	-0.292	0.067	-0.071	1.000	-0.781	-0.527	-0.537	0.059	0.198	-0.563	0.296	-0.150	0.059	-0.424	-0.040	0.247	-0.024	-0.010	-0.370	-0.046	0.422	-0.242	0.134	-0.584	-0.149	0.048	-0.233	0.358	0.191	-0.032
(10) CO_3^{2-}*$Al(OH)_4^-$	0.532	0.705	0.221	0.137	0.155	0.356	-0.234	-0.033	-0.781	1.000	0.329	0.732	0.078	-0.279	0.650	-0.521	0.098	-0.045	0.563	-0.126	-0.190	-0.109	-0.019	0.240	-0.231	-0.513	-0.006	-0.099	0.644	0.143	0.087	0.192	-0.352	-0.191	-0.232
(11) CO_3^{2-}*CO_3^{2-}	0.320	0.330	0.429	-0.154	0.162	0.252	0.233	-0.116	-0.527	0.329	1.000	0.310	0.086	-0.279	0.138	0.120	-0.066	-0.172	0.323	0.158	-0.381	0.028	0.074	0.276	0.371	-0.305	0.357	0.060	0.271	-0.245	-0.024	0.093	-0.296	-0.340	0.035
(12) NO_2^-*$Al(OH)_4^-$	0.347	0.554	0.224	0.051	0.105	0.204	-0.254	-0.150	-0.537	0.732	0.310	1.000	0.017	-0.122	0.527	-0.269	-0.108	-0.256	0.329	-0.037	-0.279	-0.056	0.015	0.279	-0.148	-0.772	-0.129	0.066	0.391	-0.011	0.013	-0.052	-0.156	-0.169	-0.133
(13) NO_2^-*CO_3^{2-}	0.138	0.065	0.410	0.388	-0.133	-0.027	0.067	-0.028	0.059	0.078	0.086	0.017	1.000	-0.320	-0.130	0.230	0.038	0.061	-0.002	0.058	0.212	-0.198	-0.067	0.064	0.378	-0.054	0.052	-0.115	0.013	-0.089	0.527	-0.093	-0.364	-0.115	-0.162
(14) NO_2^-*NO_2^-	-0.050	-0.194	-0.146	0.157	0.232	0.052	-0.196	0.117	0.198	-0.279	-0.279	-0.122	-0.320	1.000	0.013	0.129	0.412	-0.055	-0.134	0.098	0.125	0.125	-0.165	-0.326	-0.199	0.035	-0.196	-0.003	-0.313	0.105	-0.239	-0.299	0.544	0.360	0.337
(15) NO_3^-*$Al(OH)_4^-$	0.356	0.491	-0.061	-0.039	0.479	0.283	-0.243	-0.082	-0.563	0.650	0.138	0.527	-0.130	0.013	1.000	-0.356	0.042	-0.383	0.438	-0.253	-0.274	0.125	0.007	-0.022	-0.425	-0.430	-0.075	0.040	0.475	0.226	-0.012	0.034	-0.134	-0.049	-0.258
(16) NO_3^-*CO_3^{2-}	-0.084	-0.394	0.501	-0.187	0.202	-0.075	0.132	-0.108	0.296	-0.521	0.120	-0.269	0.230	0.129	-0.356	1.000	-0.134	-0.156	-0.332	0.579	-0.149	0.293	0.126	-0.069	0.602	0.041	0.085	-0.066	-0.460	-0.185	-0.013	-0.364	0.063	-0.089	0.245
(17) NO_3^-*NO_2^-	0.224	0.003	0.033	0.431	0.368	0.142	-0.083	0.277	-0.150	0.098	-0.066	-0.108	0.038	0.412	0.042	-0.134	1.000	0.303	0.045	0.000	0.251	0.090	-0.233	-0.176	-0.082	0.093	0.175	-0.143	0.049	0.118	-0.055	0.141	0.164	0.240	0.195
(18) NO_3^-*NO_3^-	0.068	-0.092	0.066	0.300	-0.032	0.079	0.013	0.244	0.059	-0.045	-0.172	-0.256	0.061	-0.055	-0.383	-0.156	0.303	1.000	-0.090	0.149	0.381	0.071	0.048	0.104	0.108	0.347	0.016	-0.251	-0.194	0.047	0.085	0.273	-0.019	0.117	0.166
(19) OH^-*$Al(OH)_4^-$	0.749	0.955	-0.019	0.131	0.006	0.731	-0.374	0.018	-0.424	0.563	0.323	0.329	-0.002	-0.134	0.438	-0.332	0.045	-0.090	1.000	-0.270	-0.051	-0.219	-0.075	-0.110	-0.204	-0.144	-0.054	-0.308	0.330	0.195	0.187	0.147	-0.269	-0.026	-0.230
(20) OH^-*CO_3^{2-}	0.114	-0.253	0.829	-0.084	0.120	0.098	-0.049	-0.040	-0.040	-0.126	0.158	-0.037	0.058	0.098	-0.253	0.579	0.000	0.149	-0.270	1.000	-0.081	0.139	0.238	0.201	0.547	-0.091	-0.082	-0.269	-0.300	-0.093	-0.166	-0.139	0.062	-0.192	0.379
(21) OH^-*NO_2^-	0.151	-0.080	-0.114	0.849	-0.320	0.174	-0.002	0.359	0.247	-0.190	-0.381	-0.279	0.212	0.125	-0.274	-0.149	0.251	0.381	-0.051	-0.081	1.000	-0.380	-0.006	-0.038	-0.029	0.530	-0.161	-0.248	-0.215	0.112	0.208	0.358	0.009	0.333	0.096
(22) OH^-*NO_3^-	0.030	-0.255	0.042	-0.351	0.815	0.115	0.082	-0.035	-0.024	-0.109	0.028	-0.056	-0.198	0.125	0.125	0.293	0.090	0.071	-0.219	0.139	-0.380	1.000	0.429	-0.206	-0.030	-0.112	0.276	0.204	-0.110	-0.097	-0.237	-0.216	0.348	0.010	0.104
(23) OH^-*OH^-	0.148	-0.066	0.164	-0.072	0.239	0.339	0.193	-0.216	-0.010	-0.019	0.074	0.015	-0.067	-0.165	0.007	0.126	-0.233	0.048	-0.075	0.238	-0.006	0.429	1.000	0.039	0.044	-0.032	0.058	0.225	-0.017	-0.031	-0.022	-0.010	-0.008	-0.305	-0.010
(24) SO_4^{2-}*$Al(OH)_4^-$	-0.041	0.007	0.329	-0.030	-0.191	-0.188	0.187	-0.080	-0.370	0.240	0.276	0.279	0.064	-0.326	-0.022	-0.069	-0.176	0.104	-0.110	0.201	-0.038	-0.206	0.039	1.000	0.337	-0.261	-0.049	-0.086	0.040	-0.326	-0.019	0.225	-0.242	-0.228	0.071
(25) SO_4^{2-}*CO_3^{2-}	0.088	-0.228	0.699	0.009	-0.003	-0.051	0.378	0.046	-0.046	-0.231	0.371	-0.148	0.378	-0.199	-0.425	0.602	-0.082	0.108	-0.204	0.547	-0.029	-0.030	0.044	0.337	1.000	0.059	0.310	-0.116	-0.267	-0.368	0.124	-0.035	-0.133	-0.177	0.295
(26) SO_4^{2-}*NO_2^-	-0.107	-0.327	-0.262	0.326	-0.179	-0.024	0.338	0.197	0.422	-0.513	-0.305	-0.772	-0.054	0.035	-0.430	0.041	0.093	0.347	-0.144	-0.091	0.530	-0.112	-0.032	-0.261	0.059	1.000	0.168	-0.118	-0.224	0.040	0.063	0.152	0.069	0.197	0.112
(27) SO_4^{2-}*NO_3^-	0.103	-0.125	0.106	-0.040	0.391	0.016	0.574	-0.051	-0.242	-0.006	0.357	-0.129	0.052	-0.196	-0.075	0.085	0.175	0.016	-0.054	-0.082	-0.161	0.276	0.058	-0.049	0.310	0.168	1.000	0.292	0.289	-0.213	-0.016	0.078	-0.201	-0.216	0.034
(28) SO_4^{2-}*OH^-	-0.178	-0.279	-0.282	-0.243	0.134	-0.079	0.693	-0.382	0.134	-0.099	0.060	0.066	-0.115	-0.003	0.040	-0.066	-0.143	-0.251	-0.308	-0.269	-0.248	0.204	0.225	-0.086	-0.116	-0.118	0.292	1.000	0.076	-0.198	-0.092	-0.212	0.098	-0.290	-0.236
(29) SO_4^{2-}*SO_4^{2-}	0.141	0.415	-0.039	0.021	0.067	0.048	0.049	-0.207	-0.584	0.644	0.271	0.391	0.013	-0.313	0.475	-0.460	0.049	-0.194	0.330	-0.300	-0.215	-0.110	-0.017	0.040	-0.267	-0.224	0.289	0.076	1.000	0.134	0.025	0.165	-0.493	-0.343	-0.402
(30) Temp.*$Al(OH)_4^-$	0.095	0.180	-0.148	0.073	-0.063	0.112	-0.382	0.146	-0.149	0.143	-0.245	-0.011	-0.089	0.105	0.226	-0.185	0.118	0.047	0.195	-0.093	0.112	-0.097	-0.031	-0.326	-0.368	0.040	-0.213	-0.198	0.134	1.000	-0.045	-0.010	-0.251	0.099	-0.232
(31) Temp.*CO_3^{2-}	0.189	0.210	0.084	0.310	-0.168	0.110	0.051	0.218	0.048	0.087	-0.024	0.013	0.527	-0.239	-0.012	-0.013	-0.055	0.085	0.187	-0.166	0.208	-0.237	-0.022	-0.019	0.124	0.063	-0.016	-0.092	0.025	-0.045	1.000	0.107	-0.262	0.155	-0.148
(32) Temp.*NO_2^-	0.162	0.128	-0.059	0.311	-0.117	0.110	0.012	0.534	-0.233	0.192	0.093	-0.052	-0.093	-0.299	0.034	-0.364	0.141	0.273	0.147	-0.139	0.358	-0.216	-0.010	0.225	-0.035	0.152	0.078	-0.212	0.165	-0.010	0.107	1.000	-0.140	0.206	0.075
(33) Temp.*NO_3^-	-0.130	-0.332	-0.208	-0.076	0.274	-0.019	-0.078	0.329	0.358	-0.352	-0.296	-0.156	-0.364	0.544	-0.134	0.063	0.164	-0.019	-0.269	0.062	0.009	0.348	-0.008	-0.242	-0.133	0.069	-0.201	0.098	-0.493	-0.251	-0.262	-0.140	1.000	0.505	0.403
(34) Temp.*OH^-	0.006	-0.091	-0.262	0.272	0.056	-0.006	-0.288	0.798	0.191	-0.191	-0.340	-0.169	-0.115	0.360	-0.049	-0.089	0.240	0.117	-0.026	-0.192	0.333	0.010	-0.305	-0.228	-0.177	0.197	-0.216	-0.290	-0.343	0.099	0.155	0.206	0.505	1.000	0.389
(35) Temp.*SO_4^{2-}	0.005	-0.287	0.280	0.083	0.138	-0.035	-0.014	0.495	-0.032	-0.232	0.035	-0.133	-0.162	0.337	-0.258	0.245	0.195	0.166	-0.230	0.379	0.096	0.104	-0.010	0.071	0.295	0.112	0.034	-0.236	-0.402	-0.232	-0.148	0.075	0.403	0.389	1.000

Appendix B Preparation of Salt Solutions

The target mass of each starting material for the preparation of the salt solution is shown in Table B.1.

Table B.1 Target Mass of Starting Materials

Solution	NaOH 50 wt% (g)	$Al(NO_3)_3 \cdot 9H_2O$ (g)	$Na_2SO_4 \cdot 10H_2O$ (g)	$Na_2CO_3 \cdot H_2O$ (g)	$NaNO_3$ (g)	$NaNO_2$ (g)	Notes
1	120.023	0.038	0.032	0.012	0.824	0.690	
1A	120.023	0.038	0	0.012	0.824	0.690	1
2	120.023	0.038	0.032	0.012	0.824	0.690	
2A	120.023	0.038	0	0.012	0.824	0.690	1
3	44.624	0.626	0.509	11.296	5.968	5.624	
3A	44.624	0.626	0.509	7.440	5.968	5.624	2
4	11.210	4.963	0.335	11.008	38.100	8.737	
5	11.210	4.963	0.335	11.008	38.100	8.737	
6	39.356	24.875	6.653	0.733	4.100	20.621	
7	17.858	4.250	3.895	4.498	2.432	37.453	
8	26.331	15.493	11.145	2.962	2.953	28.301	
9	46.552	22.842	6.950	5.630	6.246	6.041	
9A	46.552	22.842	6.950	3.100	6.246	6.041	3
10	47.355	28.364	15.549	2.749	6.124	4.335	
10A	47.355	28.364	8.055	2.749	6.124	4.335	4
11	50.304	4.232	13.181	3.613	0.139	6.683	
11A	50.304	4.232	8.055	3.613	0.139	6.683	5
12	23.588	1.808	2.835	11.584	0.204	24.830	
13	29.004	22.553	3.583	0.444	19.924	12.523	
14	4.630	1.174	3.460	0.900	30.179	23.568	
15	34.023	27.782	5.828	11.777	2.375	1.163	
16	34.023	27.782	5.828	11.777	2.375	1.163	
17	18.157	1.876	15.049	10.240	0.850	5.480	
18	9.035	2.521	5.828	11.777	2.250	0.762	
19	5.238	0.251	14.054	5.534	4.207	2.880	
20	21.810	0.923	2.713	0.156	0.727	1.003	

(1) SO_4^{2-} was reduced from 0.001M to 0M
(2) CO_3^{2-} was reduced from 0.9109M to 0.60M
(3) CO_3^{2-} was reduced from 0.4540M to 0.25M
(4) SO_4^{2-} was reduced from 0.4826M to 0.25M
(5) SO_4^{2-} was reduced from 0.4091M to 0.25M

The actual mass of each starting material used to prepare the salt solutions is shown in Table B.2.

Table B.2 Actual Mass of Starting Materials

Solution	NaOH 50 wt% (g)	$Al(NO_3)_3 \cdot 9H_2O$ (g)	$Na_2SO_4 \cdot 10H_2O$ (g)	$Na_2CO_3 \cdot H_2O$ (g)	$NaNO_3$ (g)	$NaNO_2$ (g)	Notes
1A	120.0200	0.0389	0	0.0118	0.8247	0.6900	
2A	120.0200	0.0393	0	0.0122	0.8242	0.6902	
3A	44.6297	0.6264	0.5112	7.4419	5.9688	5.6262	
4	11.2167	4.9631	0.3385	11.0091	38.1005	8.7380	
5	11.2116	4.9632	0.3366	11.0081	38.1001	8.7370	
6	39.3582	24.8751	6.6552	0.7334	4.1005	20.6211	
7	17.8643	4.2510	3.8953	4.5008	2.4328	37.4536	
8	26.3388	15.4930	11.1485	2.9622	2.9534	28.3016	1
9A	46.5500	22.8423	6.9500	3.1009	6.2468	6.0419	2
10A	47.3553	28.3642	8.0552	2.7494	6.1251	4.3358	
11A	50.3000	4.2324	8.0556	3.6141	0.1398	6.6838	3
12	23.5943	1.8086	2.8352	11.5847	0.2044	24.8309	4
13	29.0065	22.5527	3.5850	0.4442	19.9242	12.5239	
14	4.6312	1.1742	3.4643	0.9007	30.1795	23.5685	
15	34.0226	27.7822	5.8287	11.7775	2.3764	1.1639	
16	34.0248	27.7829	5.8290	11.7779	2.3759	1.1638	
17	18.1584	1.8784	15.0494	10.2415	0.8509	5.4810	
18	9.0392	2.5214	5.8302	11.7777	2.2511	0.7629	
19	5.2395	0.2519	14.0541	5.5344	4.2072	2.8812	
20	21.8108	0.9234	2.7154	0.1561	0.7287	1.0039	

(1) Final volume was 111.0 mL
(2) Final volume was 101.0 mL
(3) Final volume was 103.5 mL
(4) Final volume was 105.5 mL

Appendix C Pu and Am Solubilities in Simulated Salt Solutions

The Pu and Am solubilities measured in the salt solutions as a function of the equilibration time are given in Table C.1 – C.3. The Pu and Am concentrations measured in the blank and standard solutions are also included for each data set. For convenience, the solubilities are given in both µg/mL and moles/L.

Table C.1 Pu and Am Solubility in Salt Solutions – Sample 1

Solution	Equilibration Time (days)	Pu Solubility (µg/mL)	Pu Solubility (moles/L)	Am Solubility (µg/mL)	Am Solubility (moles/L)
1A	40	3.5	1.5E-05	1.7	7.2E-06
2A	41	4.8	2.0E-05	1.5	6.2E-06
3A	41	3.3	1.4E-05	0.19	7.7E-07
4	40	1.3	5.4E-06	0.19	7.8E-07
5	41	1.0	4.3E-06	0.27	1.1E-06
6	41	0.66	2.8E-06	0.27	1.1E-06
7	41	0.63	2.6E-06	0.34	1.4E-06
8	40	0.86	3.6E-06	0.19	7.8E-07
9A	40	1.6	6.6E-06	0.21	8.8E-07
10A	41	0.68	2.8E-06	0.36	1.5E-06
11A	41	2.6	1.1E-05	0.05	2.1E-07
12	40	2.4	1.0E-05	0.78	3.2E-06
13	40	0.68	2.9E-06	0.41	1.7E-06
14	40	0.35	1.5E-06	0.18	7.4E-07
15	40	1.6	6.5E-06	0.48	2.0E-06
16	41	0.78	3.3E-06	0.21	8.8E-07
17	40	1.8	7.7E-06	0.85	3.5E-06
18	41	0.20	8.6E-07	0.04	1.5E-07
19	41	0.13	5.3E-07	0.05	2.2E-07
20	41	0.32	1.3E-06	0.25	1.0E-06
Blank	NA				
Standard	NA	0.53	2.2E-06	0.58	2.4E-06

Table C.2 Pu and Am Solubility in Salt Solutions – Sample 2

Solution	Equilibration Time (days)	Pu Solubility (µg/mL)	Pu Solubility (moles/L)	Am Solubility (µg/mL)	Am Solubility (moles/L)
1A	68	3.1	1.3E-05	0.73	3.0E-06
2A	69	4.5	1.9E-05	1.2	4.8E-06
3A	69	3.2	1.3E-05	0.85	3.5E-06
4	68	1.9	7.8E-06	0.90	3.7E-06
5	69	0.43	1.8E-06	0.13	5.4E-07
6	69	0.55	2.3E-06	0.32	1.3E-06
7	69	0.25	1.0E-06	0.13	5.4E-07
8	68	1.6	6.5E-06	1.17	4.8E-06
9A	68	1.7	7.0E-06	0.65	2.7E-06
10A	69	0.81	3.4E-06	0.55	2.3E-06
11A	69	2.6	1.1E-05	0.14	5.9E-07
12	68	2.6	1.1E-05	1.0	4.3E-06
13	68	0.80	3.3E-06	0.62	2.6E-06
14	68	0.08	3.4E-07	0.0044	1.8E-08
15	68	1.8	7.4E-06	1.0	4.2E-06
16	69	0.66	2.8E-06	0.34	1.4E-06
17	68	1.6	6.5E-06	0.79	3.3E-06
18	69	0.21	8.8E-07	0.048	2.0E-07
19	69	0.11	4.5E-07	0.058	2.4E-07
20	69	0.20	8.2E-07	0.15	6.2E-07
Blank	NA	negligible	negligible	negligible	negligible
Standard	NA	0.51	2.1E-06	0.57	2.4E-06

Table C.3 Pu and Am Solubility in Salt Solutions – Sample 3

Solution	Equilibration Time (days)	Pu Solubility (µg/mL)	Pu Solubility (moles/L)	Am Solubility (µg/mL)	Am Solubility (moles/L)
1A	96	2.8	1.2E-05	1.1	4.6E-06
2A	97	3.9	1.6E-05	1.3	5.6E-06
3A	97	3.6	1.5E-05	0.96	4.0E-06
4	96	1.7	7.2E-06	1.3	5.4E-06
5	97	2.1	8.8E-06	1.8	7.4E-06
6	97	0.40	1.7E-06	0.19	7.8E-07
7	97	0.17	7.1E-07	0.082	3.4E-07
8	96	0.94	3.9E-06	0.41	1.7E-06
9A	96	2.1	9.0E-06	1.5	6.0E-06
10A	97	0.77	3.2E-06	0.57	2.4E-06
11A	97	2.5	1.0E-05	0.72	3.0E-06
12	96	2.0	8.6E-06	0.79	3.3E-06
13	96	0.43	1.8E-06	0.28	1.2E-06
14	96	0.26	1.1E-06	0.23	9.4E-07
15	96	1.40	5.9E-06	0.71	3.0E-06
16	97	0.54	2.3E-06	0.27	1.1E-06
17	96	2.1	8.8E-06	0.77	3.2E-06
18	97	0.15	6.3E-07	0.046	1.9E-07
19	97	0.13	5.5E-07	0.12	4.8E-07
20	97	0.24	9.9E-07	0.18	7.6E-07
Blank	NA	negligible	negligible	negligible	negligible
Standard	NA	0.48	2.0E-06	0.55	2.3E-06

The blank solutions analyzed with each sample set showed negligible amounts of both Pu and Am. The analysis of the standard Pu and Am solutions are compared with the known concentration in Table C.4.

Table C.4 Comparison of Analyzed Pu and Am Concentrations with Prepared Values

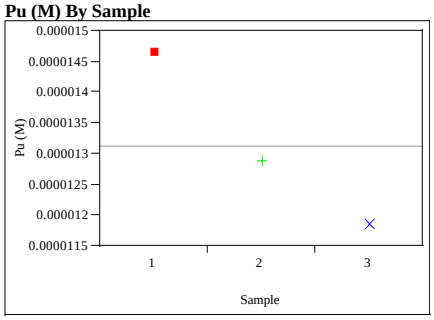
Data Set	Pu Prepared Conc	Pu Measured Conc	Percent Difference	Am Prepared Conc	Am Measured Conc	Percent Difference
	(µ/g/mL)	(µ/g/mL)	(%)	(µ/g/mL)	(µ/g/mL)	(%)
1	0.50	0.53	6.0	0.50	0.58	16
2	0.50	0.51	2.0	0.50	0.57	14
3	0.50	0.48	-4.0	0.50	0.55	10

The measured Pu concentrations are within nominally ±5 % of the prepared concentration which is typically the accuracy of the TTA/APHA method. The measured Am concentrations appear biased high based on the prepared concentration; although, they are reasonably consistent. The bias can likely be attributed to a concentration in the standard which was slightly higher than 1000 µg/mL. This could be attributed to a higher than measured concentration in the purified Am solution used to prepare the standard or the multiple transfers of the purified Am solution with a variable volume pipette in a radioactive glovebox which were required to prepare the standard.

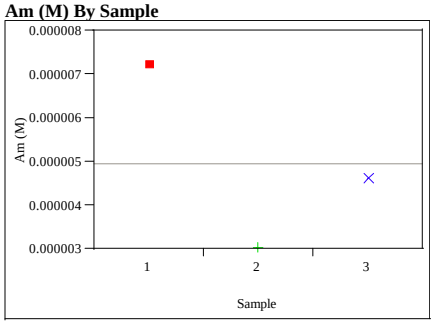
Appendix D Statistical Exhibits

Exhibit D.1 Plots of Pu and Am Solubility Data by Sample Number

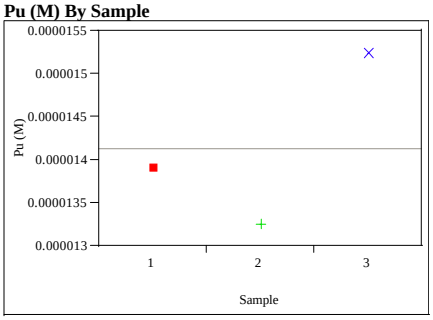
Trial ID=1A
Pu (M) By Sample



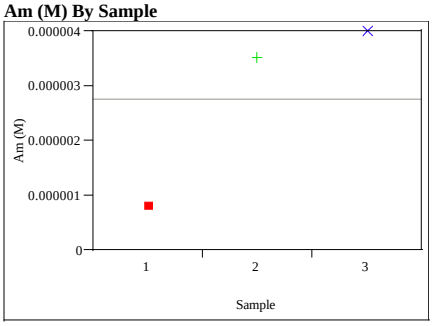
Am (M) By Sample



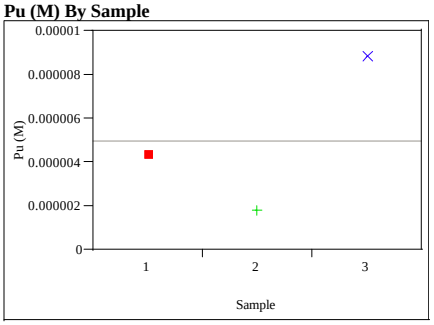
Trial ID=3A
Pu (M) By Sample



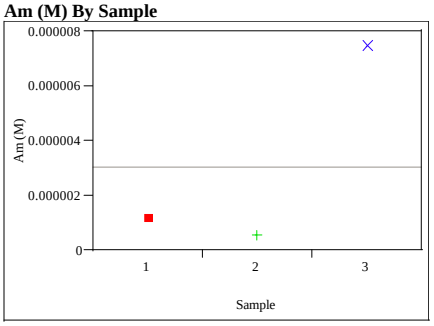
Am (M) By Sample



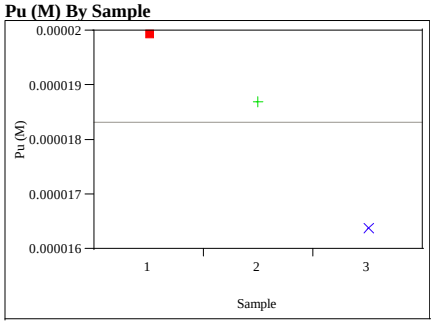
Trial ID=5
Pu (M) By Sample



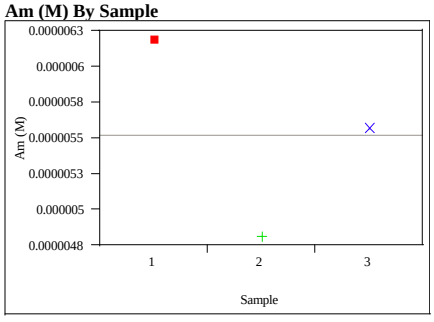
Am (M) By Sample



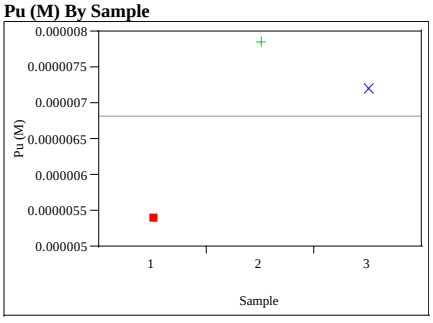
Trial ID=2A
Pu (M) By Sample



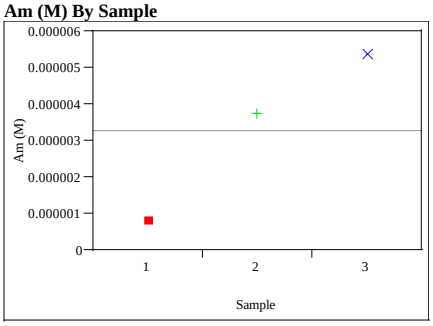
Am (M) By Sample



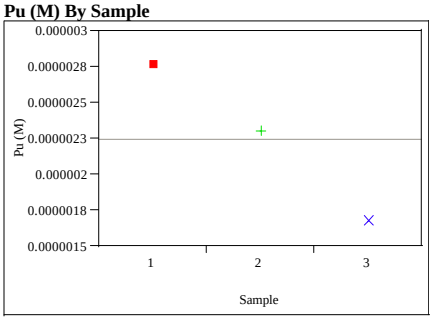
Trial ID=4
Pu (M) By Sample



Am (M) By Sample



Trial ID=6
Pu (M) By Sample



Am (M) By Sample

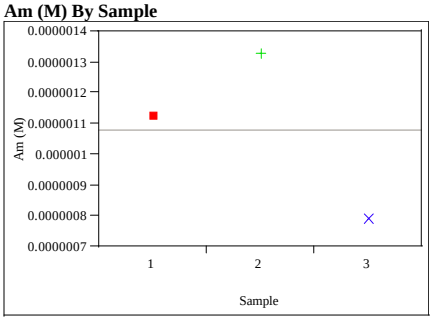
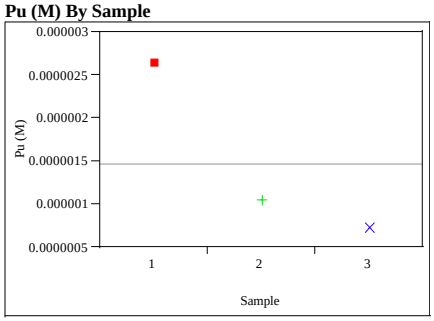
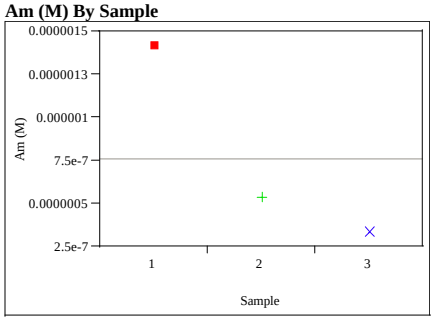


Exhibit D.1 Plots of Pu and Am Solubility Data by Sample Number (continued)

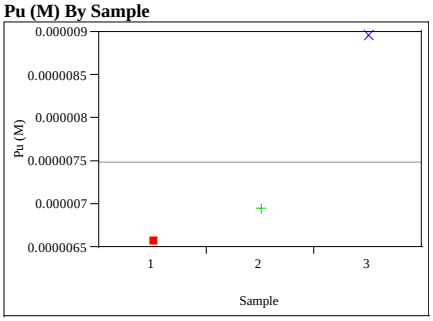
Trial ID=7
Pu (M) By Sample



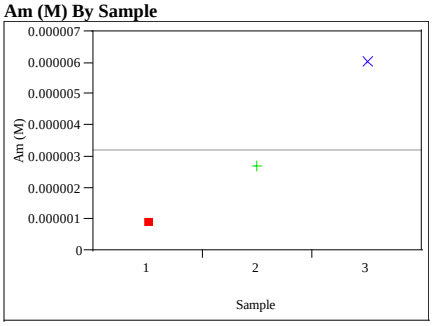
Am (M) By Sample



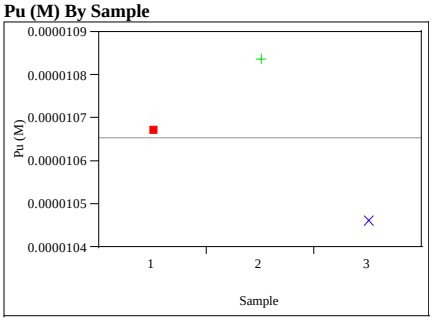
Trial ID=9A
Pu (M) By Sample



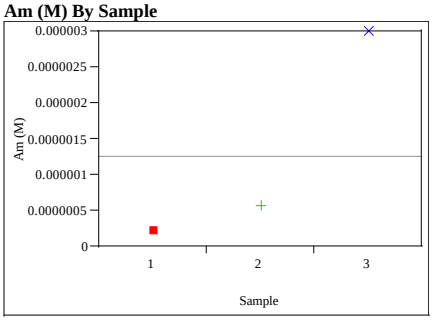
Am (M) By Sample



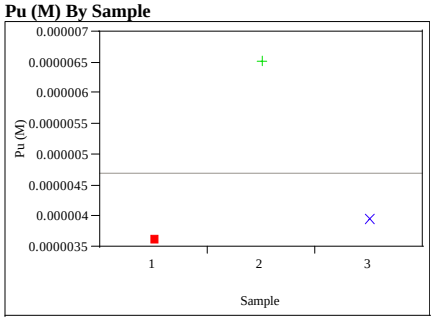
Trial ID=11A
Pu (M) By Sample



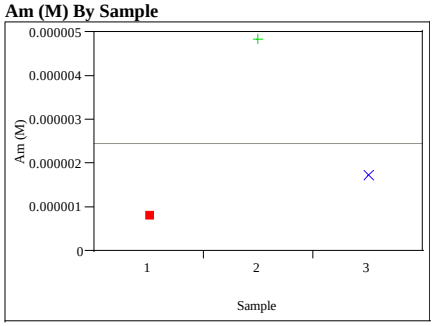
Am (M) By Sample



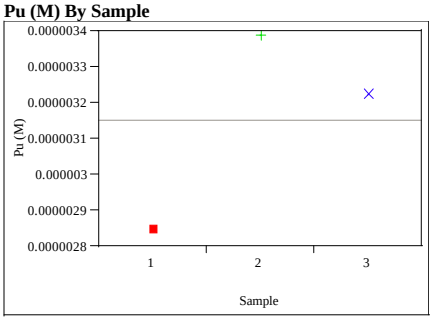
Trial ID=8
Pu (M) By Sample



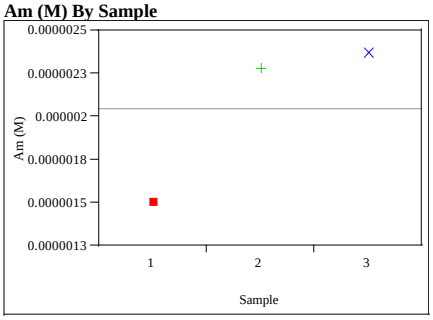
Am (M) By Sample



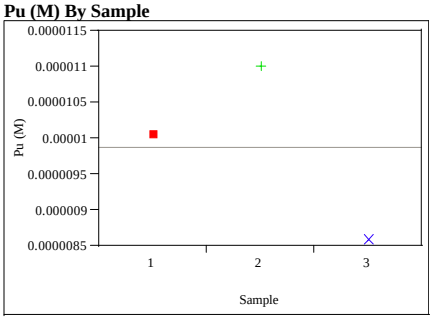
Trial ID=10A
Pu (M) By Sample



Am (M) By Sample



Trial ID=12
Pu (M) By Sample



Am (M) By Sample

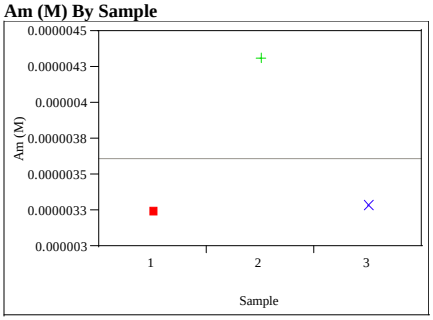
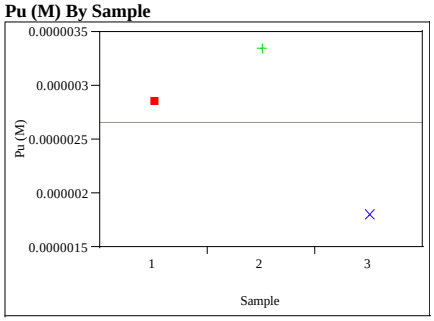
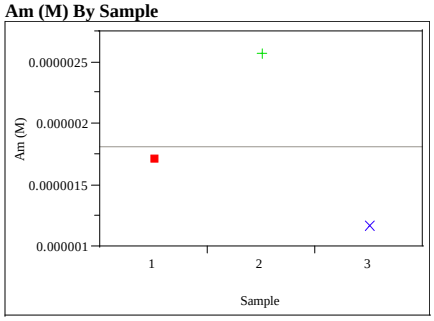


Exhibit D.1 Plots of Pu and Am Solubility Data by Sample Number (continued)

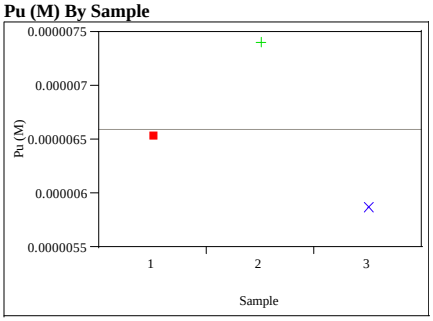
Trial ID=13
Pu (M) By Sample



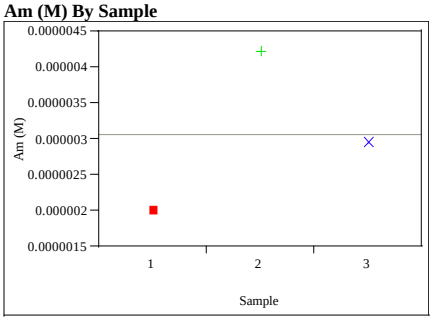
Am (M) By Sample



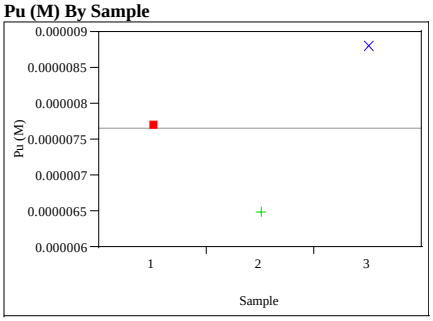
Trial ID=15
Pu (M) By Sample



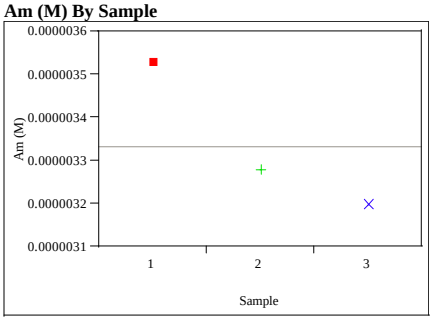
Am (M) By Sample



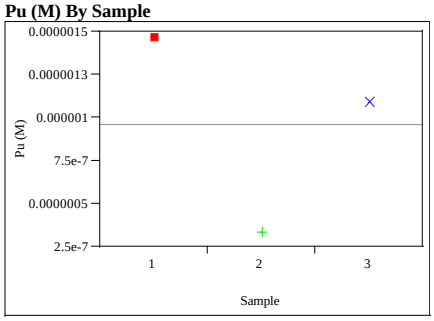
Trial ID=17
Pu (M) By Sample



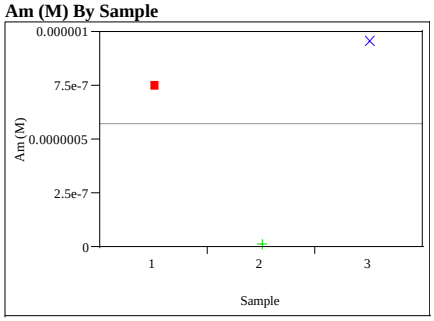
Am (M) By Sample



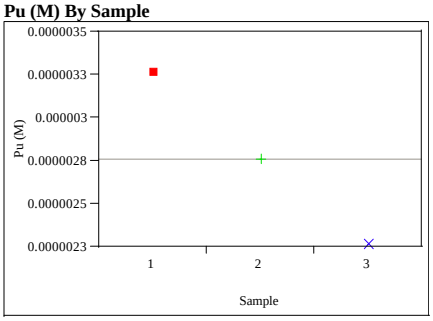
Trial ID=14
Pu (M) By Sample



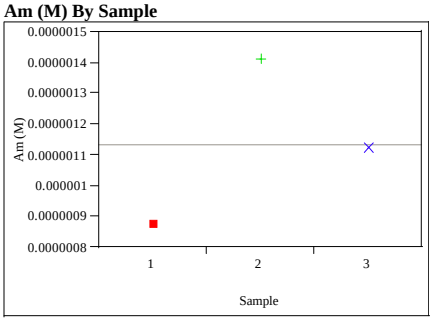
Am (M) By Sample



Trial ID=16
Pu (M) By Sample

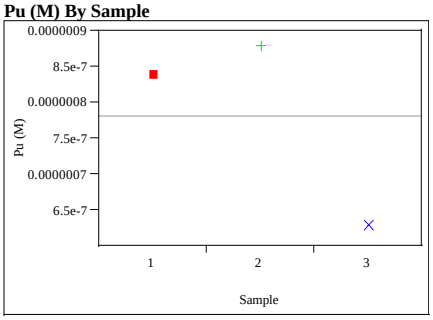


Am (M) By Sample



Trial ID=18

Pu (M) By Sample



Am (M) By Sample

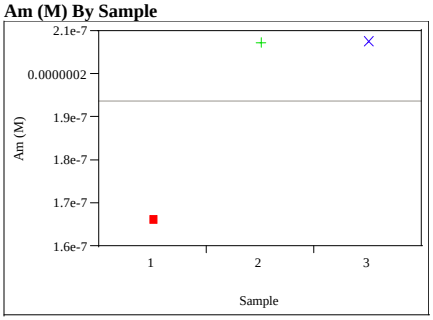
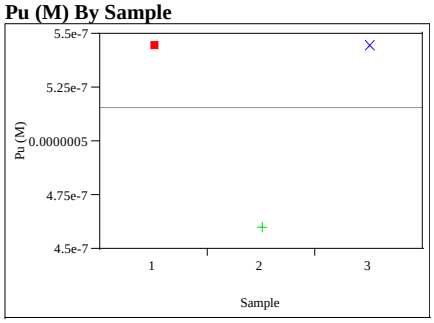
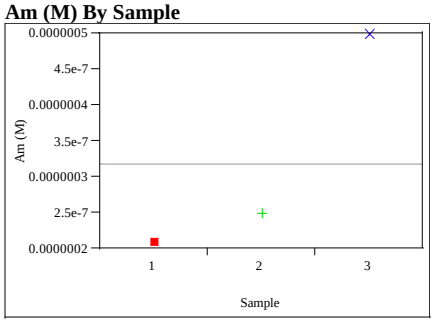


Exhibit D.1 Plots of Pu and Am Solubility Data by Sample Number (continued)

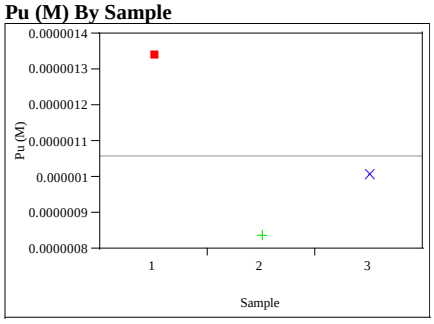
Trial ID=19
Pu (M) By Sample



Am (M) By Sample



Trial ID=20
Pu (M) By Sample



Am (M) By Sample

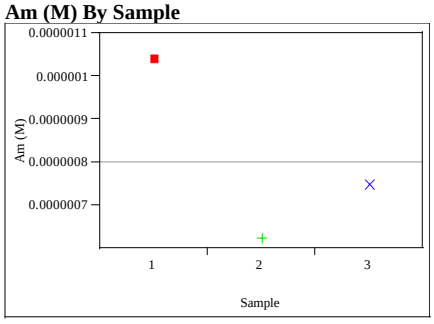
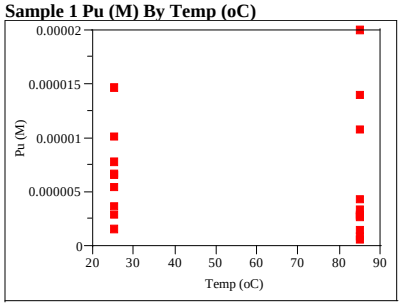
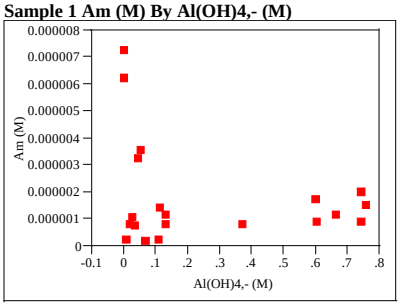


Exhibit D.2 Plots of Pu and Am Solubility Data (Each Sample and Average) by Each Factor

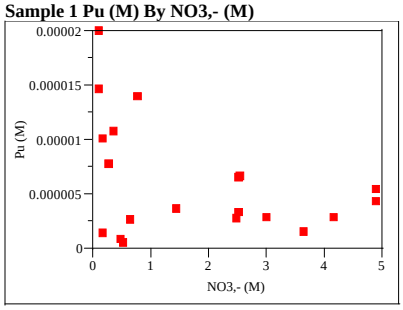
Sample 1 Pu (M) By Temp (oC)



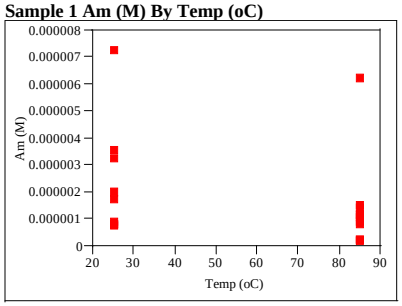
Sample 1 Am (M) By Al(OH)4,- (M)



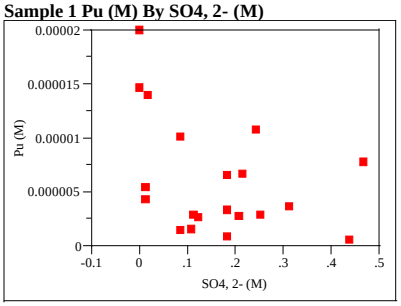
Sample 1 Pu (M) By NO3,- (M)



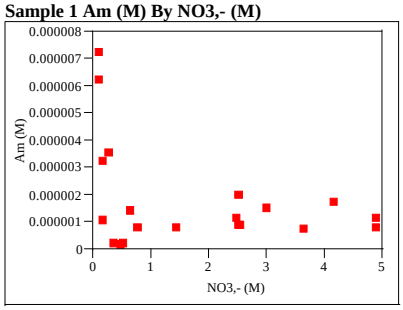
Sample 1 Am (M) By Temp (oC)



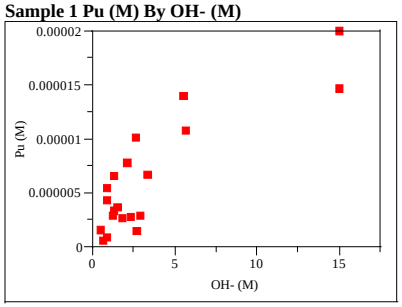
Sample 1 Pu (M) By SO4, 2- (M)



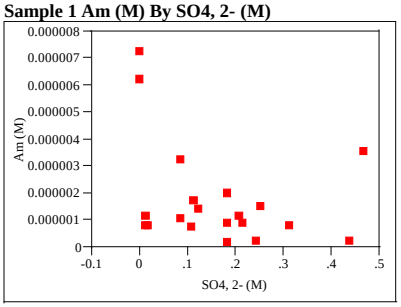
Sample 1 Am (M) By NO3,- (M)



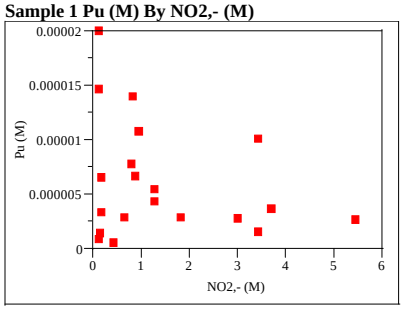
Sample 1 Pu (M) By OH- (M)



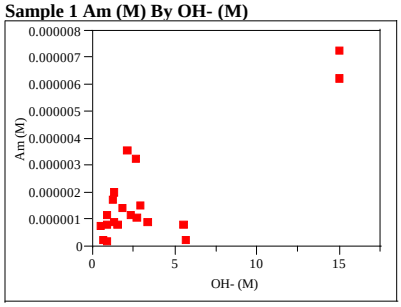
Sample 1 Am (M) By SO4, 2- (M)



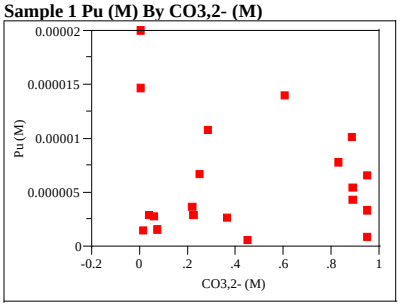
Sample 1 Pu (M) By NO2,- (M)



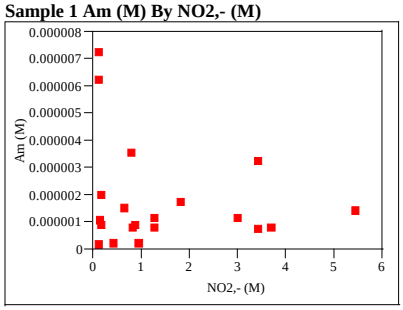
Sample 1 Am (M) By OH- (M)



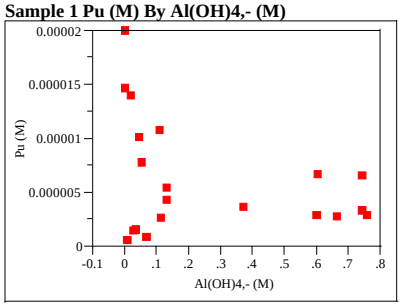
Sample 1 Pu (M) By CO3,2- (M)



Sample 1 Am (M) By NO2,- (M)



Sample 1 Pu (M) By Al(OH)4,- (M)



Sample 1 Am (M) By CO3,2- (M)

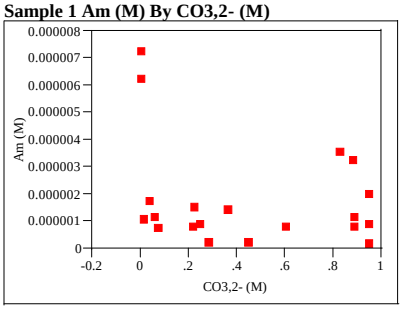
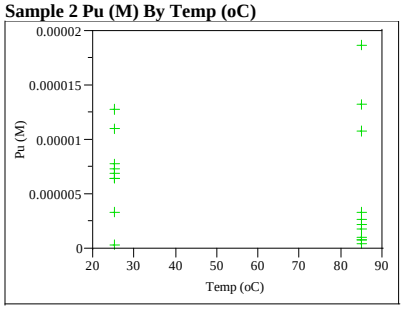
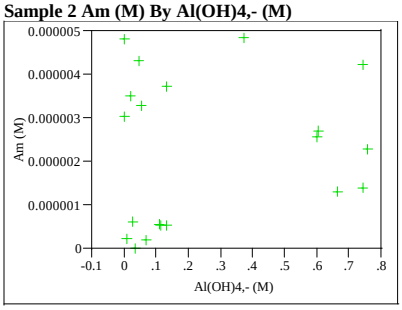


Exhibit D.2 Plots of Pu and Am Solubility Data (Each Sample and Average) by Each Factor (continued)

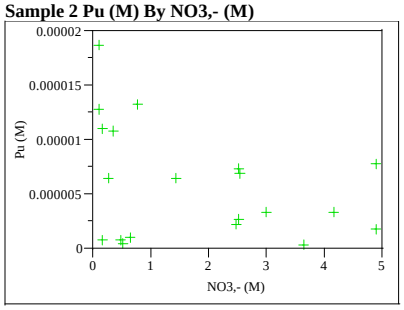
Sample 2 Pu (M) By Temp (oC)



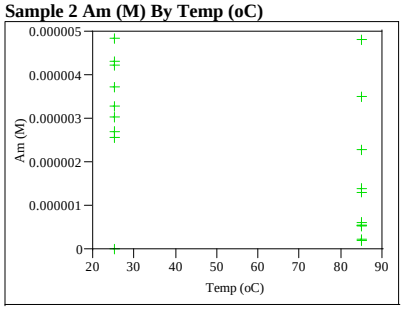
Sample 2 Am (M) By Al(OH)4,- (M)



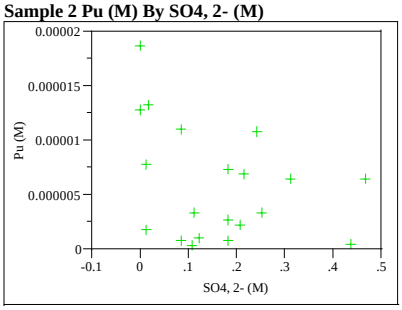
Sample 2 Pu (M) By NO3,- (M)



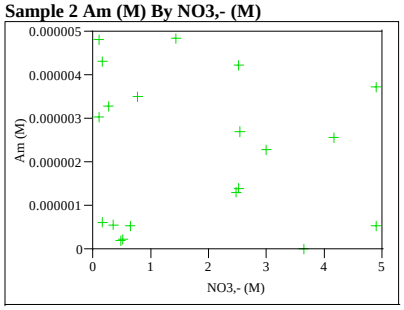
Sample 2 Am (M) By Temp (oC)



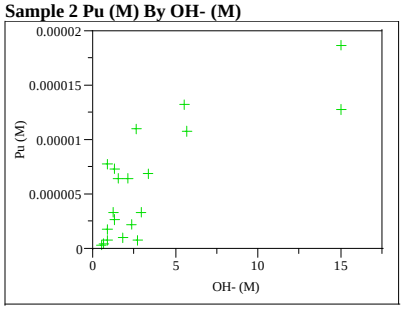
Sample 2 Pu (M) By SO4, 2- (M)



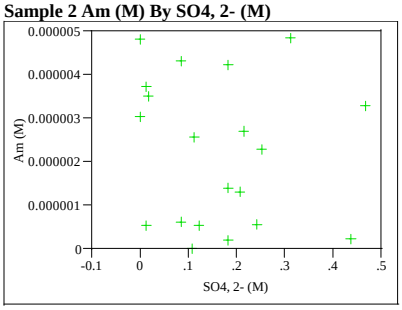
Sample 2 Am (M) By NO3,- (M)



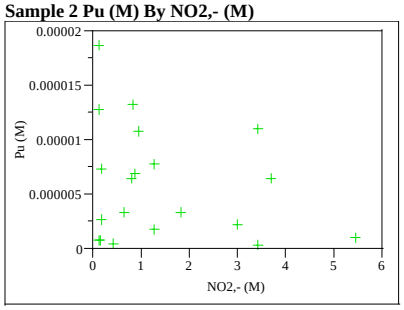
Sample 2 Pu (M) By OH- (M)



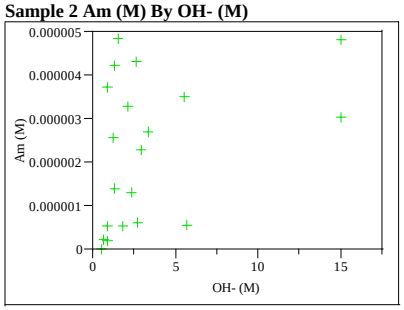
Sample 2 Am (M) By SO4, 2- (M)



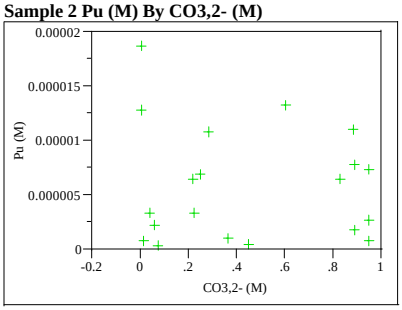
Sample 2 Pu (M) By NO2,- (M)



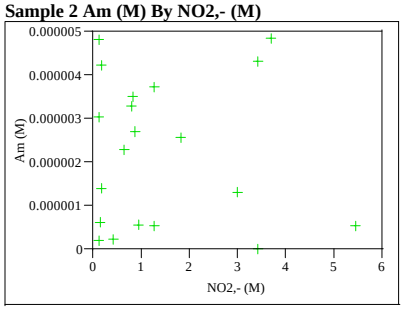
Sample 2 Am (M) By OH- (M)



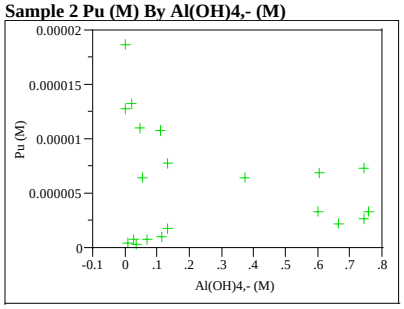
Sample 2 Pu (M) By CO3,2- (M)



Sample 2 Am (M) By NO2,- (M)



Sample 2 Pu (M) By Al(OH)4,- (M)



Sample 2 Am (M) By CO3,2- (M)

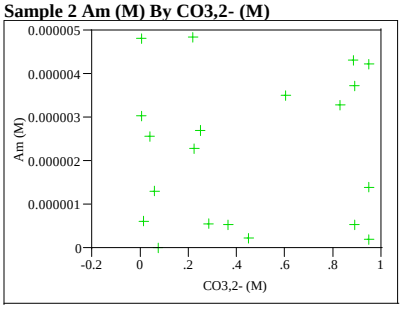
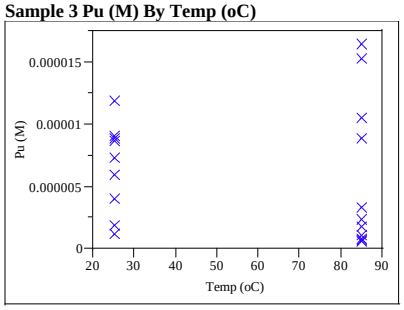
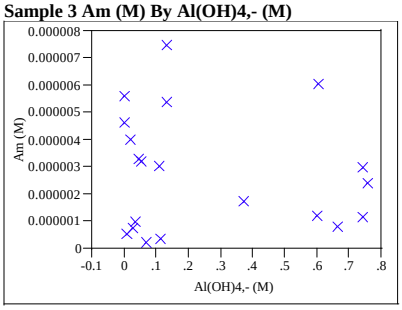


Exhibit D.2 Plots of Pu and Am Solubility Data (Each Sample and Average) by Each Factor (continued)

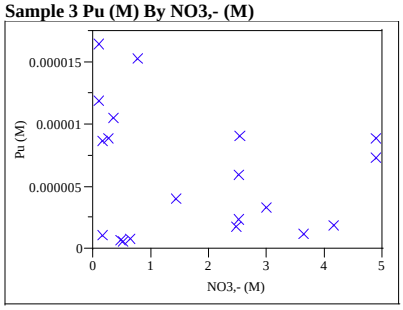
Sample 3 Pu (M) By Temp (oC)



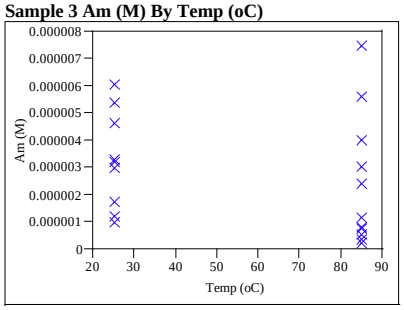
Sample 3 Am (M) By Al(OH)4,- (M)



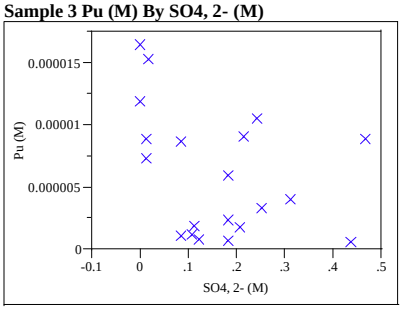
Sample 3 Pu (M) By NO3,- (M)



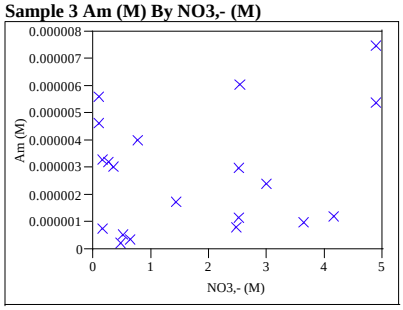
Sample 3 Am (M) By Temp (oC)



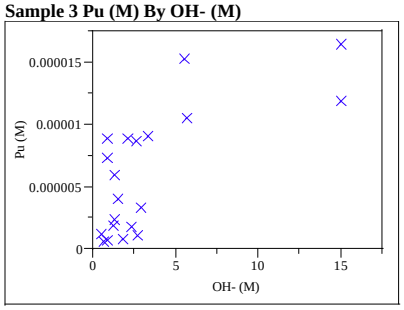
Sample 3 Pu (M) By SO4, 2- (M)



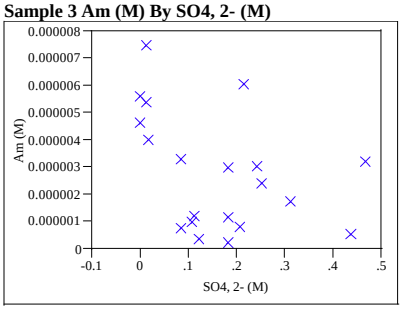
Sample 3 Am (M) By NO3,- (M)



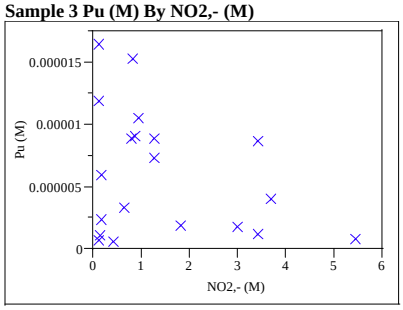
Sample 3 Pu (M) By OH- (M)



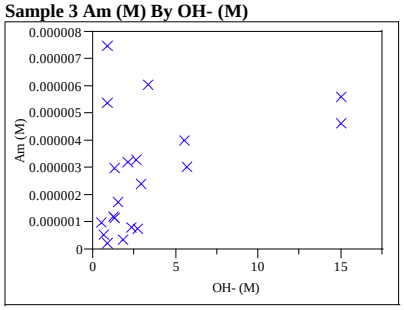
Sample 3 Am (M) By SO4, 2- (M)



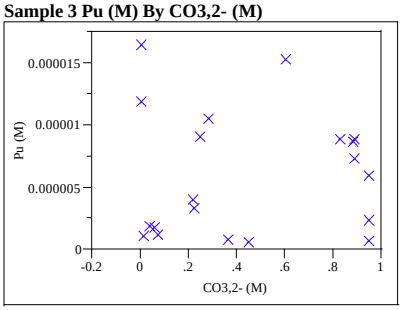
Sample 3 Pu (M) By NO2,- (M)



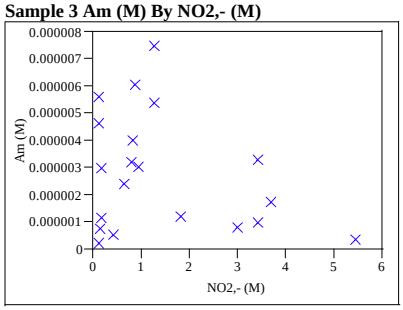
Sample 3 Am (M) By OH- (M)



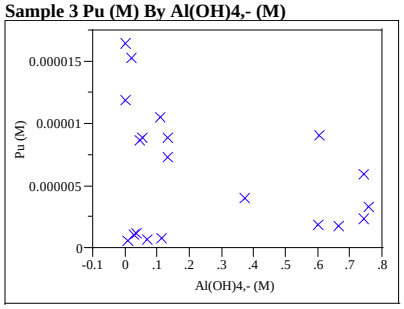
Sample 3 Pu (M) By CO3,2- (M)



Sample 3 Am (M) By NO2,- (M)



Sample 3 Pu (M) By Al(OH)4,- (M)



Sample 3 Am (M) By CO3,2- (M)

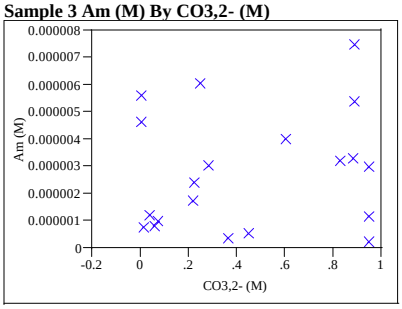
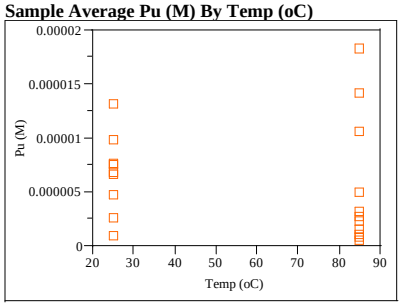
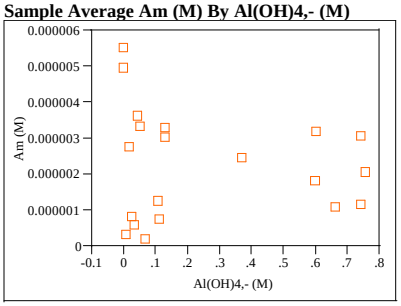


Exhibit D.2 Plots of Pu and Am Solubility Data (Each Sample and Average) by Each Factor (continued)

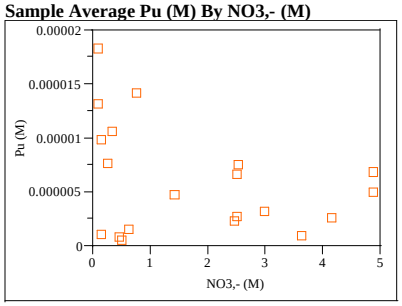
Sample Average Pu (M) By Temp (oC)



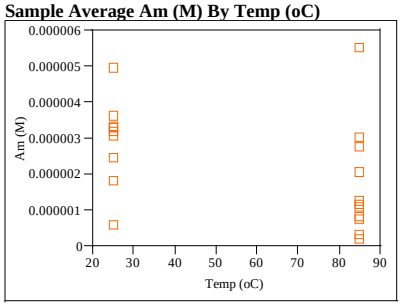
Sample Average Am (M) By Al(OH)4,- (M)



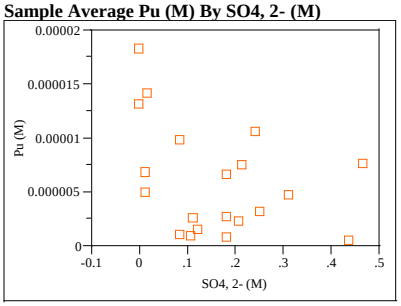
Sample Average Pu (M) By NO3,- (M)



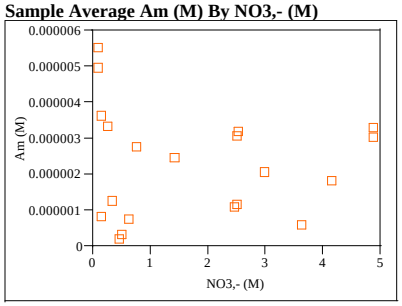
Sample Average Am (M) By Temp (oC)



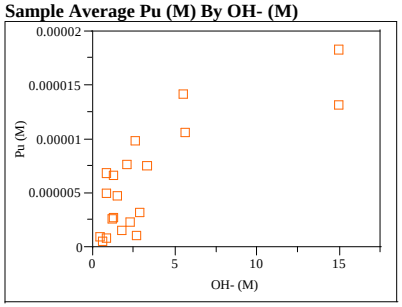
Sample Average Pu (M) By SO4, 2- (M)



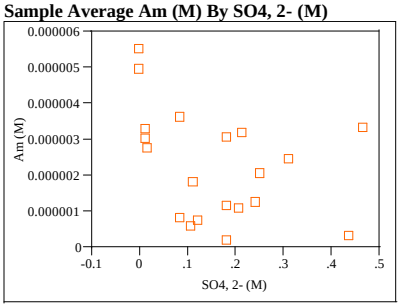
Sample Average Am (M) By NO3,- (M)



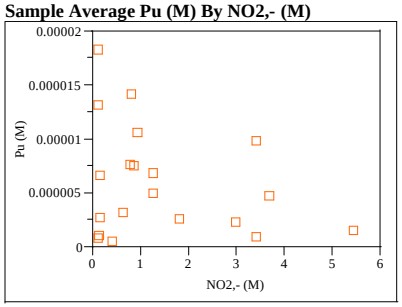
Sample Average Pu (M) By OH- (M)



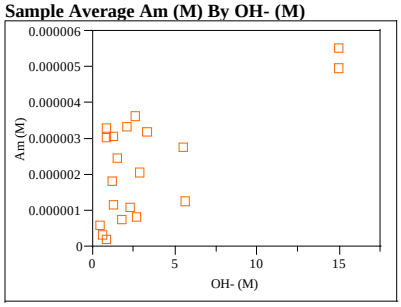
Sample Average Am (M) By SO4, 2- (M)



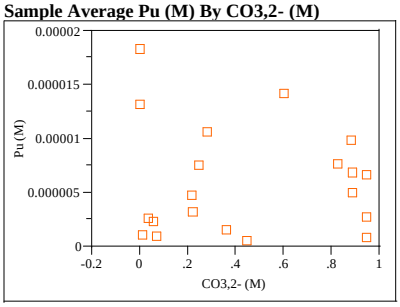
Sample Average Pu (M) By NO2,- (M)



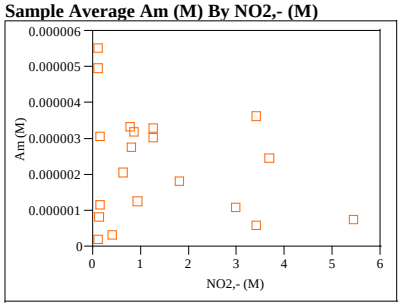
Sample Average Am (M) By OH- (M)



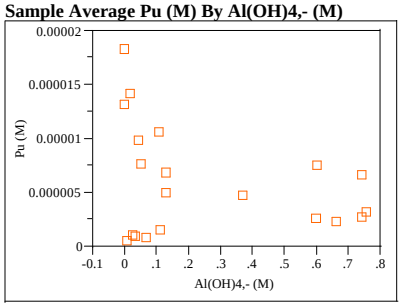
Sample Average Pu (M) By CO3,2- (M)



Sample Average Am (M) By NO2,- (M)



Sample Average Pu (M) By Al(OH)4,- (M)



Sample Average Am (M) By CO3,2- (M)

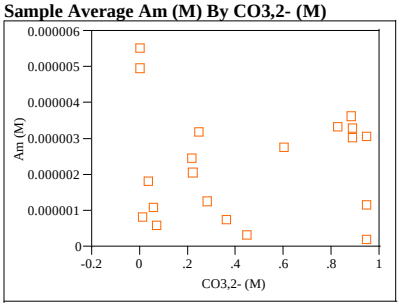
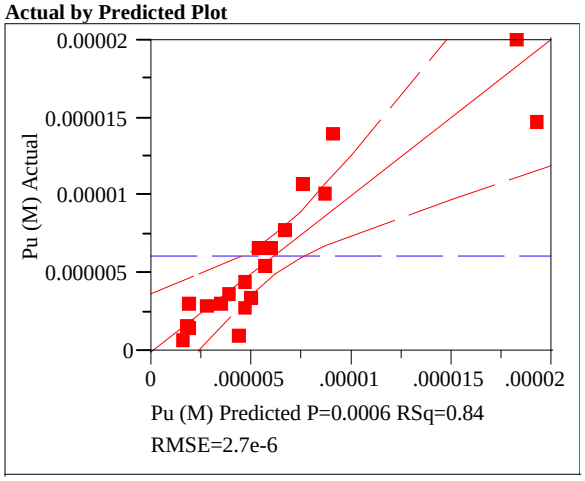


Exhibit D.3 First-Order Fits of the Am and Pu Solubility Data for Each Sample and the Average

Response Sample 1 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.84023
RSquare Adj	0.747031
Root Mean Square Error	0.000003
Mean of Response	0.000006
Observations (or Sum Wgts)	20

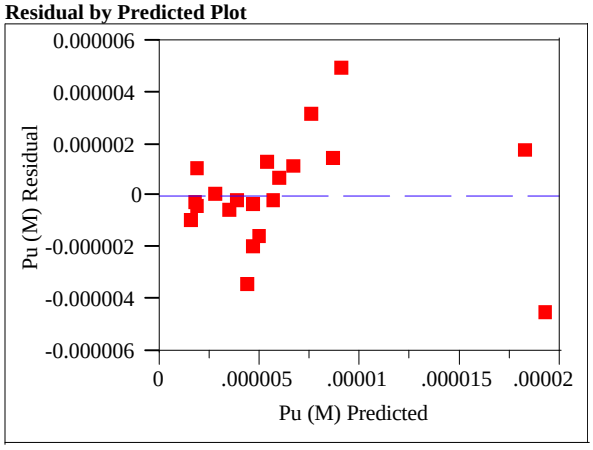
Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	4.4451e-10	6.35e-11	9.0154
Error	12	8.4524e-11	7.044e-12	Prob > F
C. Total	19	5.2904e-10		0.0006

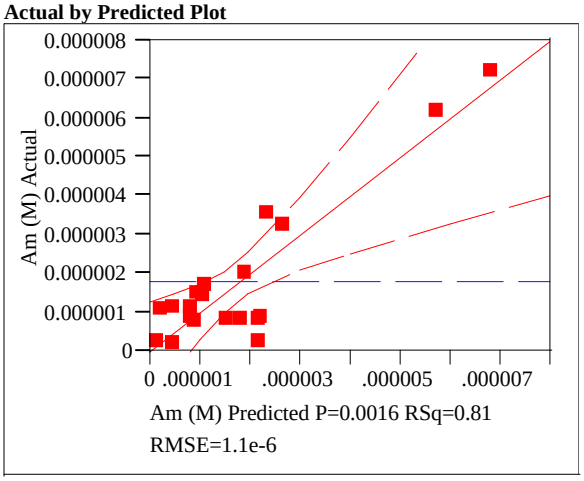
Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-3.793e-7	0.000004	-0.09	0.9276
Temp (oC)	-1.676e-8	2.167e-8	-0.77	0.4542
OH- (M)	0.0000013	2.713e-7	4.90	0.0004
Al(OH)4,- (M)	-1.743e-7	0.000003	-0.07	0.9466
SO4, 2- (M)	1.2052e-7	0.000007	0.02	0.9857
CO3,2- (M)	0.0000052	0.000002	2.52	0.0267
NO3,- (M)	4.2463e-8	6.002e-7	0.07	0.9448
NO2,- (M)	4.078e-7	4.968e-7	0.82	0.4277

Residual by Predicted Plot



Response Sample 1Am (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.807219
RSquare Adj	0.694764
Root Mean Square Error	0.000001
Mean of Response	0.000002
Observations (or Sum Wgts)	20

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	5.5745e-11	7.964e-12	7.1781
Error	12	1.3313e-11	1.109e-12	Prob > F
C. Total	19	6.9058e-11		0.0016

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	3.6472e-7	0.000002	0.22	0.8260
Temp (oC)	-1.764e-8	8.599e-9	-2.05	0.0627
OH- (M)	4.5572e-7	1.077e-7	4.23	0.0012
Al(OH)4,- (M)	2.0305e-7	0.000001	0.20	0.8442
SO4, 2- (M)	9.5676e-7	0.000003	0.37	0.7214
CO3,2- (M)	0.000001	8.144e-7	1.26	0.2319
NO3,- (M)	3.5722e-9	2.382e-7	0.01	0.9883
NO2,- (M)	1.5456e-7	1.972e-7	0.78	0.4483

Residual by Predicted Plot

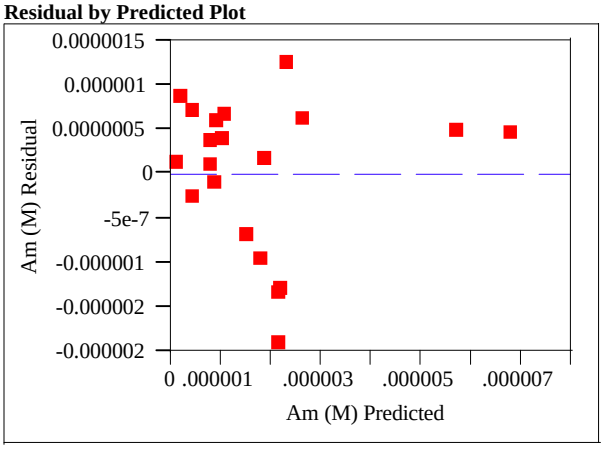
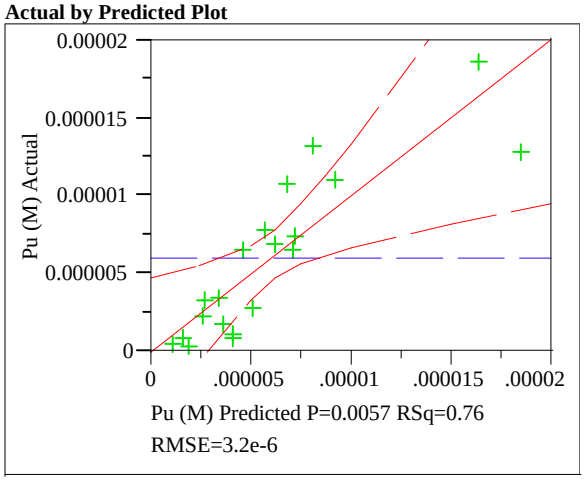


Exhibit D.3 First-Order Fits of the Am and Pu Solubility Data for Each Sample and the Average (continued)

Response Sample 2 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.75719
RSquare Adj	0.615551
Root Mean Square Error	0.000003
Mean of Response	0.000006
Observations (or Sum Wgts)	20

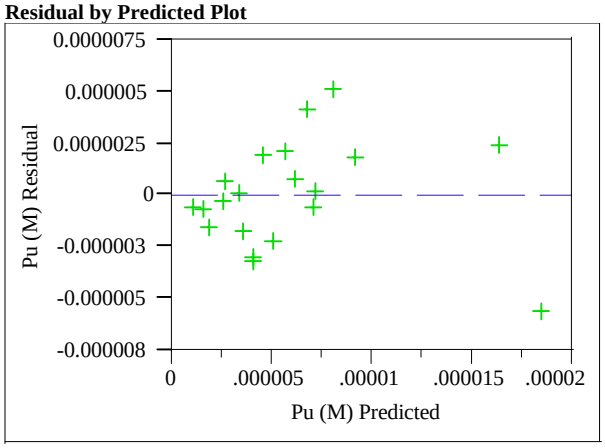
Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	3.8821e-10	5.546e-11	5.3459
Error	12	1.2449e-10	1.037e-11	Prob > F
C. Total	19	5.127e-10		0.0057

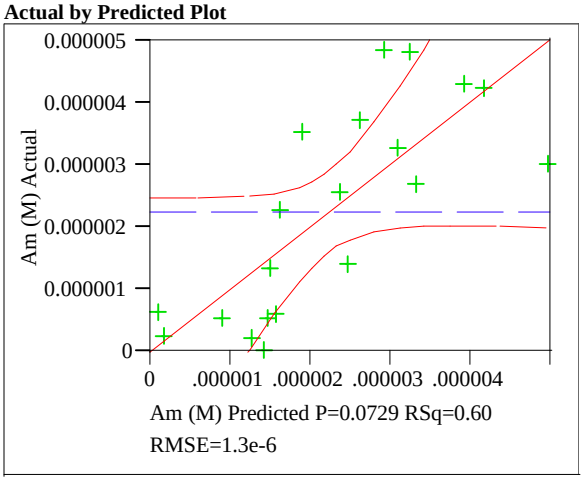
Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	0.0000014	0.000005	0.28	0.7867
Temp (oC)	-3.591e-8	2.629e-8	-1.37	0.1971
OH- (M)	0.0000012	3.293e-7	3.64	0.0034
Al(OH)4,- (M)	0.0000012	0.000003	0.40	0.6993
SO4, 2- (M)	-7.226e-7	0.000008	-0.09	0.9296
CO3,2- (M)	0.000005	0.000002	2.02	0.0664
NO3,- (M)	-1.785e-7	7.284e-7	-0.24	0.8106
NO2,- (M)	3.3814e-7	6.029e-7	0.56	0.5852

Residual by Predicted Plot



Response Sample 2 Am (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.599278
RSquare Adj	0.365523
Root Mean Square Error	0.000001
Mean of Response	0.000002
Observations (or Sum Wgts)	20

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	3.1976e-11	4.568e-12	2.5637
Error	12	2.1382e-11	1.782e-12	Prob > F
C. Total	19	5.3358e-11		0.0729

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	0.0000018	0.000002	0.88	0.3964
Temp (oC)	-2.871e-8	1.09e-8	-2.63	0.0218
OH- (M)	2.5751e-7	1.365e-7	1.89	0.0836
Al(OH)4,- (M)	0.000002	0.000001	1.59	0.1386
SO4, 2- (M)	-3.192e-7	0.000003	-0.10	0.9250
CO3,2- (M)	0.0000017	0.000001	1.67	0.1208
NO3,- (M)	-1.457e-7	3.019e-7	-0.48	0.6381
NO2,- (M)	1.6757e-7	2.499e-7	0.67	0.5151

Residual by Predicted Plot

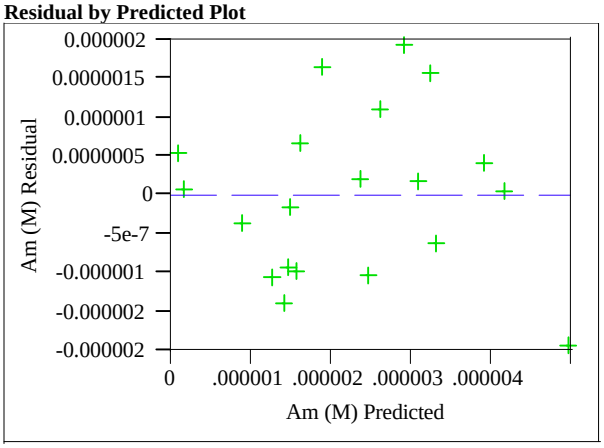
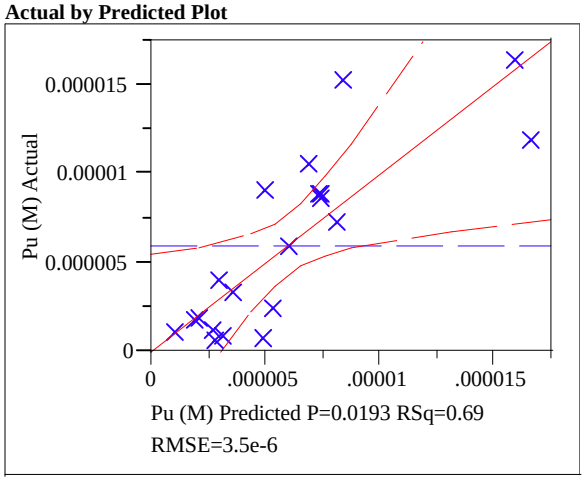


Exhibit D.3 First-Order Fits of the Am and Pu Solubility Data for Each Sample and the Average (continued)

Response Sample 3 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.693639
RSquare Adj	0.514928
Root Mean Square Error	0.000003
Mean of Response	0.000006
Observations (or Sum Wgts)	20

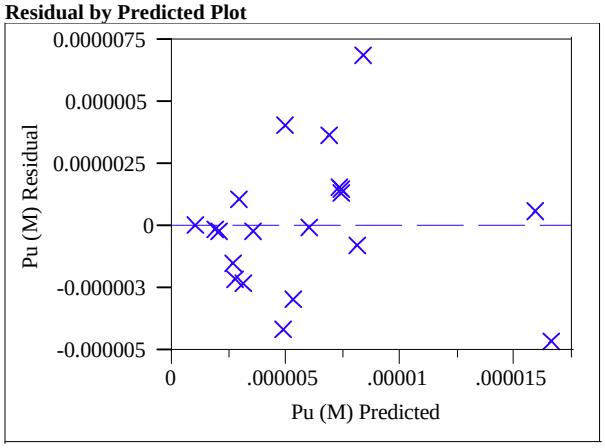
Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	3.3077e-10	4.725e-11	3.8813
Error	12	1.4609e-10	1.217e-11	Prob > F
C. Total	19	4.7686e-10		0.0193

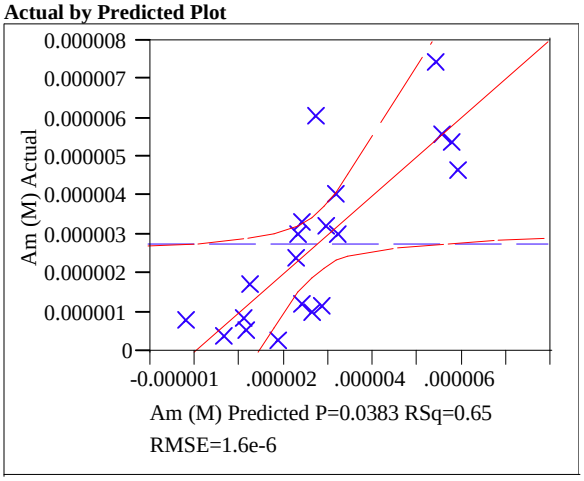
Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-0.000002	0.000005	-0.30	0.7697
Temp (oC)	-1.229e-8	2.849e-8	-0.43	0.6737
OH- (M)	0.0000012	3.567e-7	3.44	0.0049
Al(OH)4,- (M)	-0.000003	0.000003	-0.77	0.4584
SO4, 2- (M)	0.0000035	0.000009	0.40	0.6943
CO3,2- (M)	0.0000059	0.000003	2.21	0.0477
NO3,- (M)	7.6866e-7	7.891e-7	0.97	0.3492
NO2,- (M)	1.4842e-7	6.531e-7	0.23	0.8241

Residual by Predicted Plot



Response Sample 3 Am (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.649203
RSquare Adj	0.444571
Root Mean Square Error	0.000002
Mean of Response	0.000003
Observations (or Sum Wgts)	20

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	5.6901e-11	8.129e-12	3.1725
Error	12	3.0747e-11	2.562e-12	Prob > F
C. Total	19	8.7648e-11		0.0383

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-0.000001	0.000002	-0.51	0.6222
Temp (oC)	-6.401e-9	1.307e-8	-0.49	0.6331
OH- (M)	4.8184e-7	1.637e-7	2.94	0.0123
Al(OH)4,- (M)	-0.000002	0.000002	-1.12	0.2833
SO4, 2- (M)	0.0000022	0.000004	0.55	0.5935
CO3,2- (M)	0.0000026	0.000001	2.14	0.0538
NO3,- (M)	9.5232e-7	3.62e-7	2.63	0.0220
NO2,- (M)	-1.212e-8	2.996e-7	-0.04	0.9684

Residual by Predicted Plot

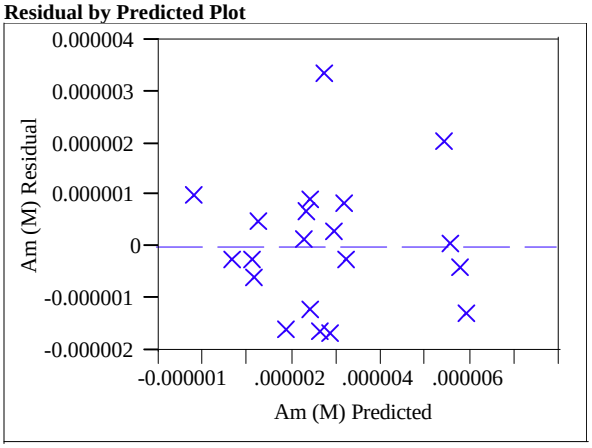
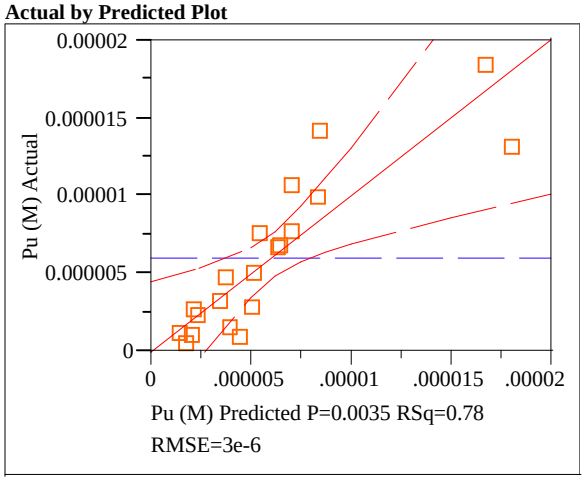


Exhibit D.3 First-Order Fits of the Am and Pu Solubility Data for Each Sample and the Average (continued)

Response Average Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.778743
RSquare Adj	0.649676
Root Mean Square Error	0.000003
Mean of Response	0.000006
Observations (or Sum Wgts)	20

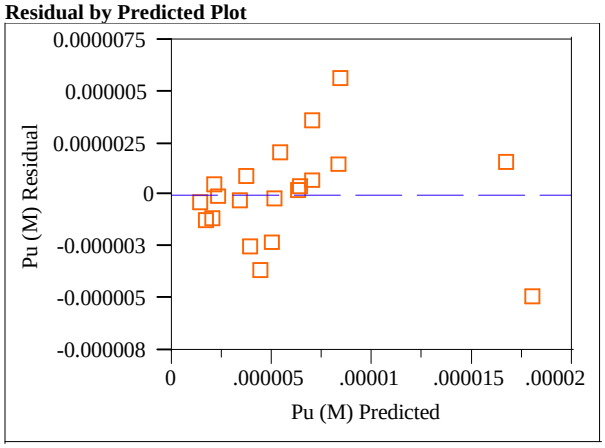
Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	3.7826e-10	5.404e-11	6.0336
Error	12	1.0747e-10	8.956e-12	Prob > F
C. Total	19	4.8573e-10		0.0035

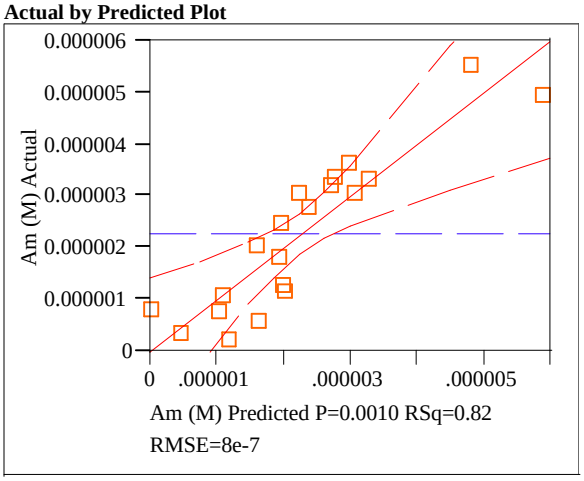
Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-2.052e-7	0.000005	-0.04	0.9652
Temp (oC)	-2.165e-8	2.443e-8	-0.89	0.3929
OH- (M)	0.0000013	3.06e-7	4.09	0.0015
Al(OH)4,- (M)	-5.058e-7	0.000003	-0.18	0.8632
SO4, 2- (M)	9.6444e-7	0.000007	0.13	0.8991
CO3,2- (M)	0.0000054	0.000002	2.33	0.0382
NO3,- (M)	2.1089e-7	6.768e-7	0.31	0.7607
NO2,- (M)	2.9812e-7	5.602e-7	0.53	0.6043

Residual by Predicted Plot



Response Average Am (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.824889
RSquare Adj	0.722741
Root Mean Square Error	7.964e-7
Mean of Response	0.000002
Observations (or Sum Wgts)	20

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	7	3.5856e-11	5.122e-12	8.0754
Error	12	7.6116e-12	6.343e-13	Prob > F
C. Total	19	4.3467e-11		0.0010

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	3.088e-7	0.000001	0.25	0.8056
Temp (oC)	-1.758e-8	6.502e-9	-2.70	0.0192
OH- (M)	3.9836e-7	8.143e-8	4.89	0.0004
Al(OH)4,- (M)	1.6994e-7	7.646e-7	0.22	0.8278
SO4, 2- (M)	9.406e-7	0.000002	0.47	0.6436
CO3,2- (M)	0.0000018	6.158e-7	2.92	0.0128
NO3,- (M)	2.7007e-7	1.801e-7	1.50	0.1596
NO2,- (M)	1.0334e-7	1.491e-7	0.69	0.5014

Residual by Predicted Plot

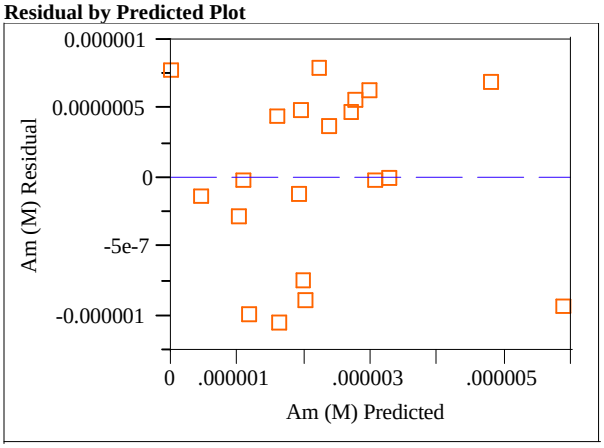
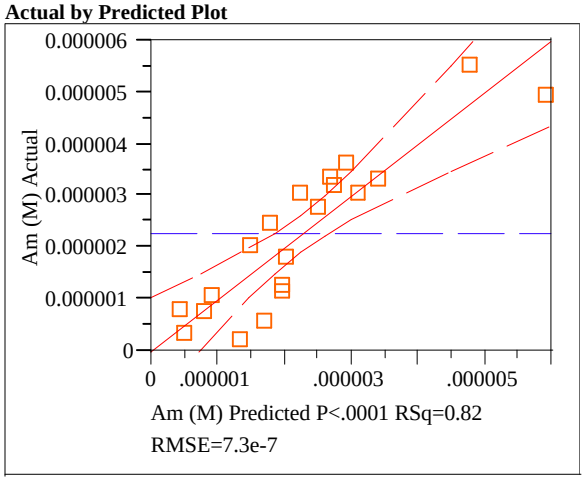


Exhibit D.4 Final First-Order Fit of the Am Solubility Data Using the Sample Averages

Response Average Am (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.815567
RSquare Adj	0.766385
Root Mean Square Error	7.311e-7
Mean of Response	0.000002
Observations (or Sum Wgts)	20

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	4	3.5451e-11	8.863e-12	16.5826
Error	15	8.0168e-12	5.345e-13	Prob > F
C. Total	19	4.3467e-11		<.0001

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	0.0000011	6.06e-7	1.77	0.0971
Temp (oC)	-1.91e-8	5.638e-9	-3.39	0.0041
OH- (M)	3.5397e-7	4.959e-8	7.14	<.0001
CO3,2- (M)	0.0000016	4.948e-7	3.16	0.0065
NO3,- (M)	2.2606e-7	1.165e-7	1.94	0.0713

Effect Tests

Source	Nparm	DF	Sum of Squares	F Ratio	Prob > F
Temp (oC)	1	1	6.1359e-12	11.4806	0.0041
OH- (M)	1	1	2.7233e-11	50.9555	<.0001
CO3,2- (M)	1	1	5.3373e-12	9.9864	0.0065
NO3,- (M)	1	1	2.013e-12	3.7664	0.0713

Residual by Predicted Plot

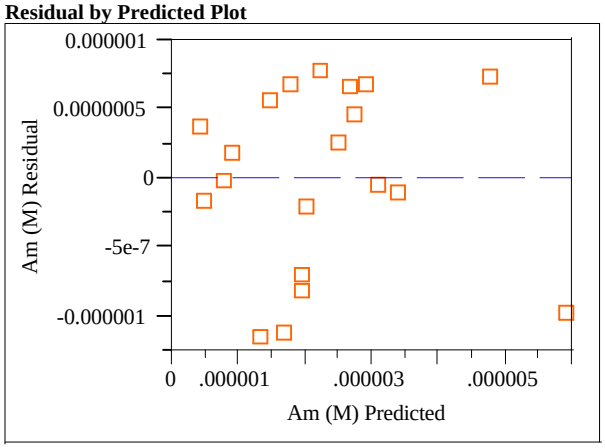
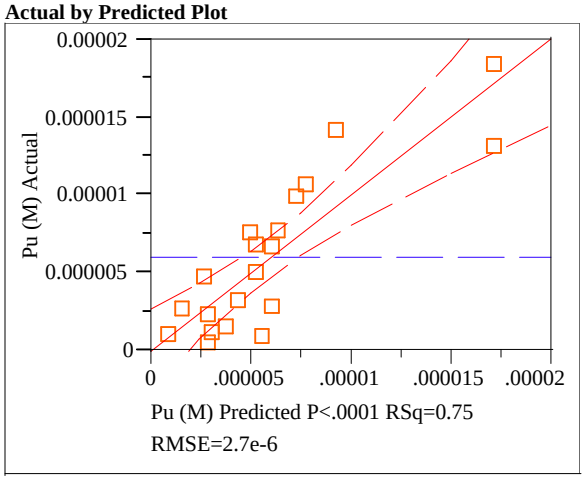


Exhibit D.5 Final First-Order Fit of the Pu Solubility Data Using the Sample Averages

Response Pu (M)
Whole Model
Actual by Predicted Plot



Summary of Fit

RSquare	0.74651
RSquare Adj	0.716688
Root Mean Square Error	0.000003
Mean of Response	0.000006
Observations (or Sum Wgts)	20

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	2	3.626e-10	1.813e-10	25.0319
Error	17	1.2313e-10	7.243e-12	Prob > F
C. Total	19	4.8573e-10		<.0001

Lack Of Fit

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	15	1.0224e-10	6.816e-12	0.6527
Pure Error	2	2.0887e-11	1.044e-11	Prob > F
Total Error	17	1.2313e-10		0.7520
				Max RSq
				0.9570

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-7.479e-8	0.000001	-0.06	0.9550
OH- (M)	0.0000012	1.629e-7	7.06	<.0001
CO3,2- (M)	0.0000049	0.000002	2.70	0.0152

Effect Tests

Source	Nparm	DF	Sum of Squares	F Ratio	Prob > F
OH- (M)	1	1	3.6109e-10	49.8555	<.0001
CO3,2- (M)	1	1	5.2766e-11	7.2853	0.0152

Residual by Predicted Plot

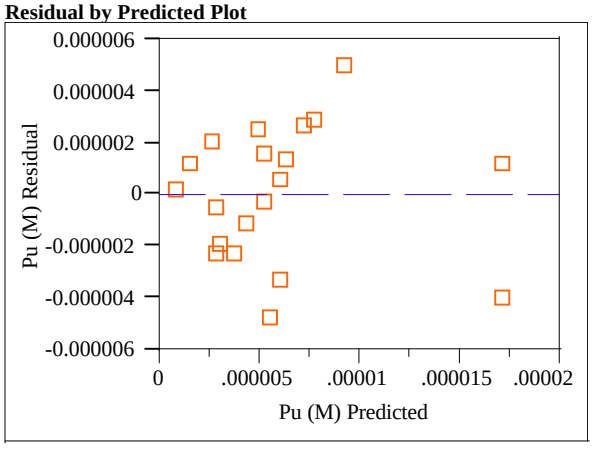
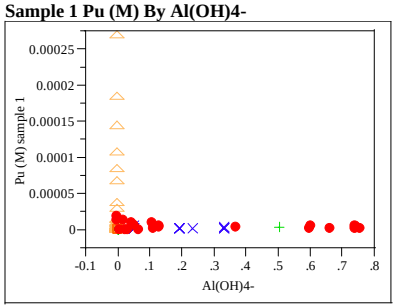
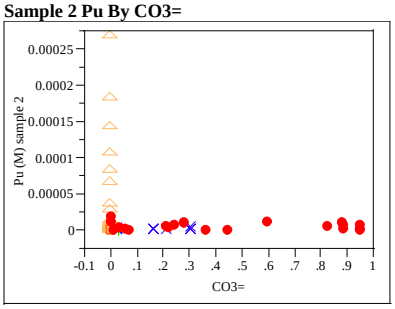


Exhibit D.6 Historical and Current Pu Solubility Data Versus the Factors of Interest

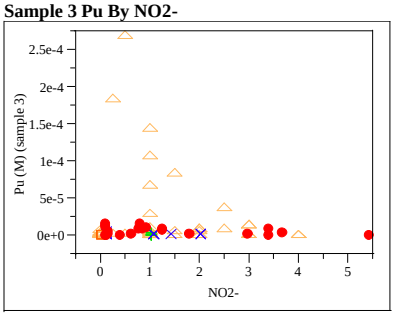
Sample 1 Pu (M) By Al(OH)4-



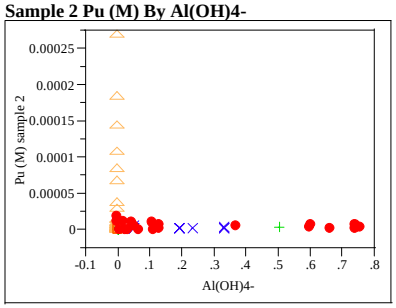
Sample 2 Pu By CO3=



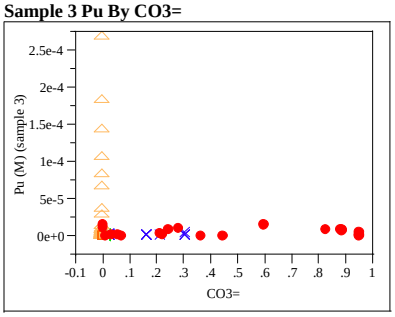
Sample 3 Pu By NO2-



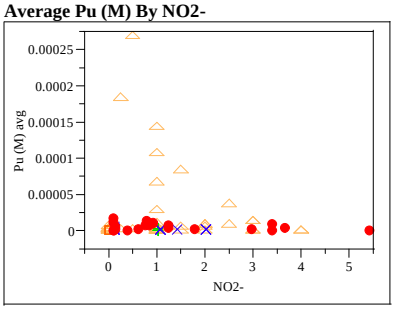
Sample 2 Pu (M) By Al(OH)4-



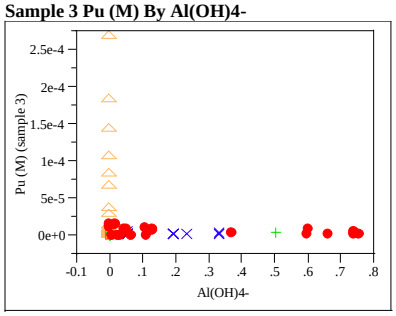
Sample 3 Pu By CO3=



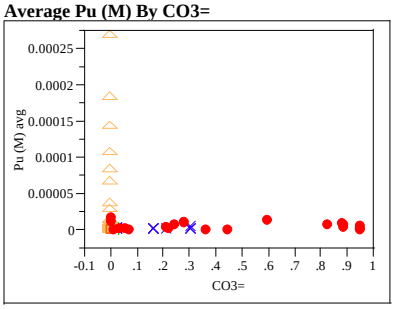
Average Pu (M) By NO2-



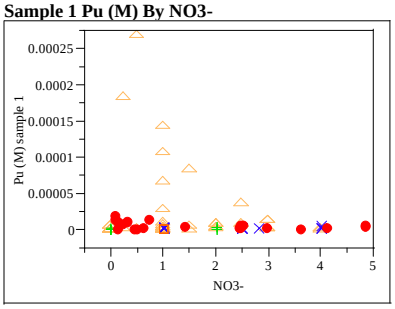
Sample 3 Pu (M) By Al(OH)4-



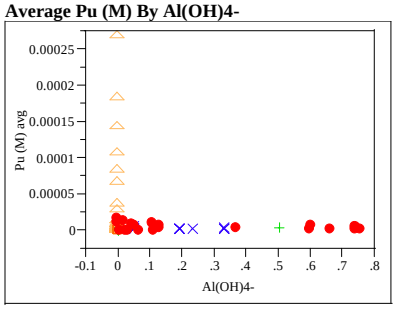
Average Pu (M) By CO3=



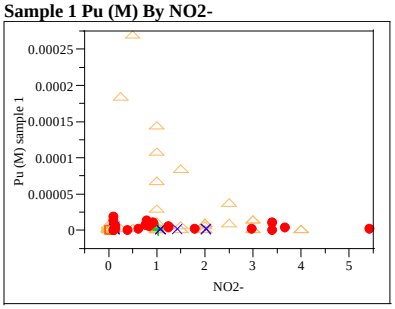
Sample 1 Pu (M) By NO3-



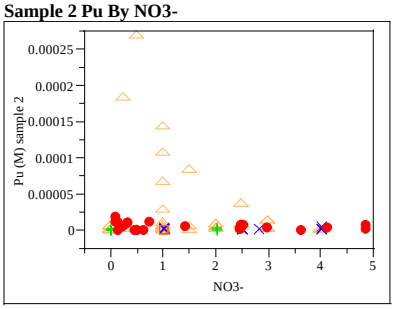
Average Pu (M) By Al(OH)4-



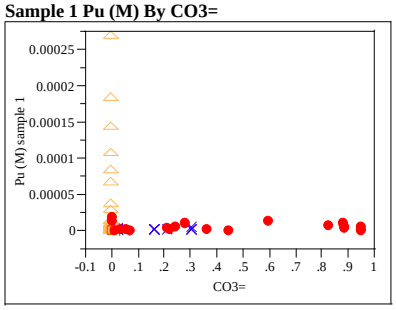
Sample 1 Pu (M) By NO2-



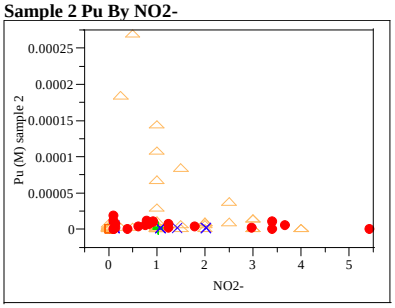
Sample 2 Pu By NO3-



Sample 1 Pu (M) By CO3=



Sample 2 Pu By NO2-



Sample 3 Pu By NO3-

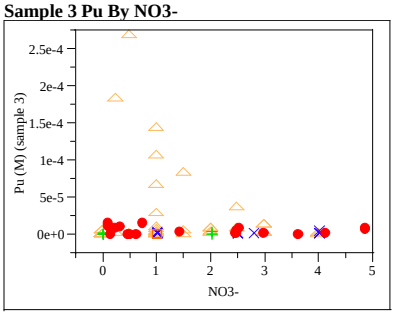
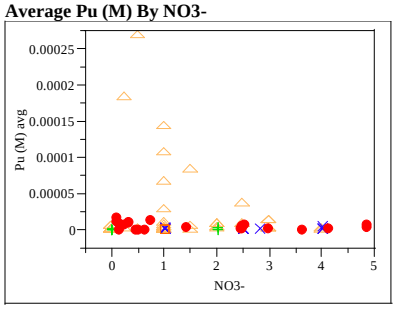
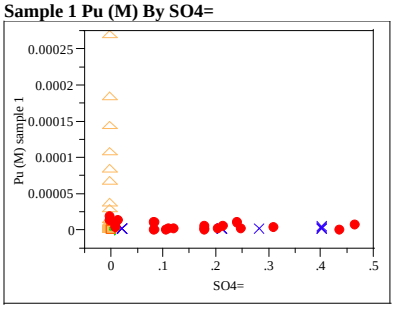


Exhibit D.6 Historical and Current Pu Solubility Data Versus the Factors of Interest continued

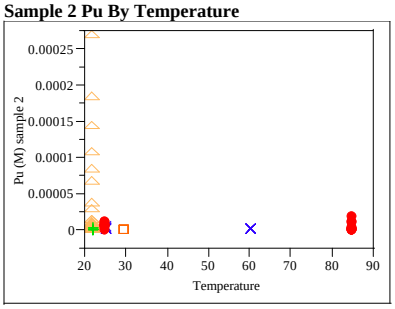
Average Pu (M) By NO3-



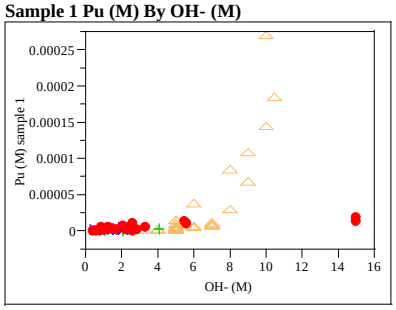
Sample 1 Pu (M) By SO4=



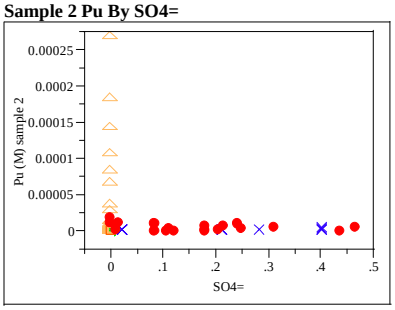
Sample 2 Pu By Temperature



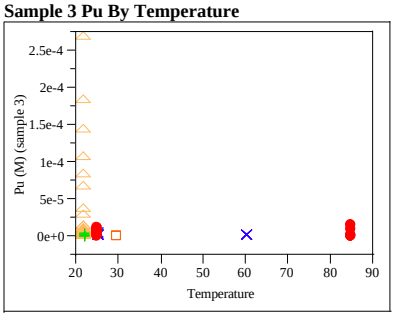
Sample 1 Pu (M) By OH- (M)



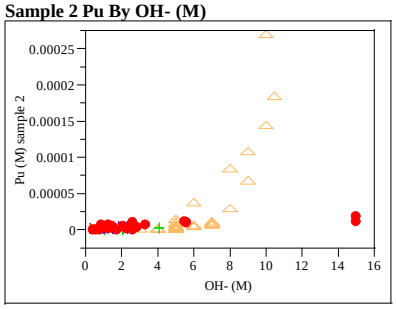
Sample 2 Pu By SO4=



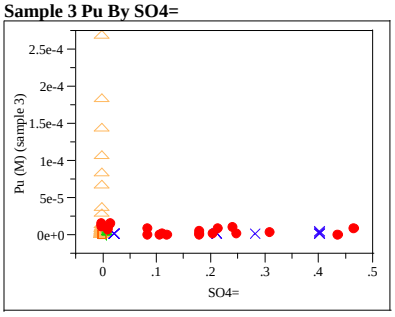
Sample 3 Pu By Temperature



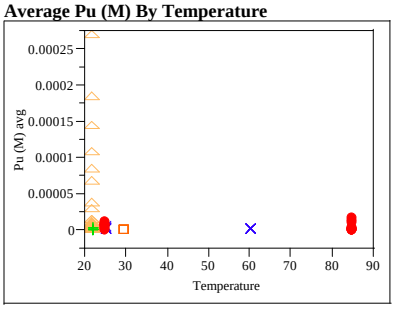
Sample 2 Pu By OH- (M)



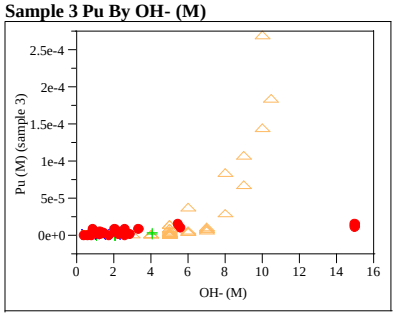
Sample 3 Pu By SO4=



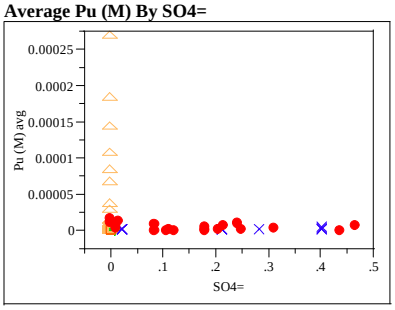
Average Pu (M) By Temperature



Sample 3 Pu By OH- (M)



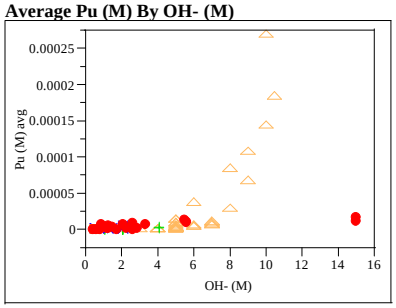
Average Pu (M) By SO4=



Legend for Plots

		Data Set	Number of Data Points
△	1	1	42
+	2	2	10
×	3	3	14
□	4	4	5
●	5	IRD Study	20

Average Pu (M) By OH- (M)



Sample 1 Pu (M) By Temperature

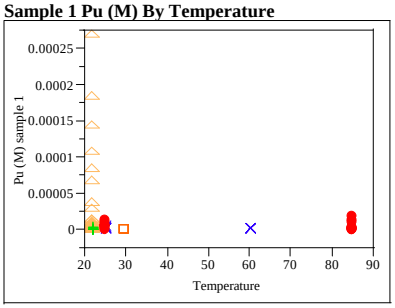
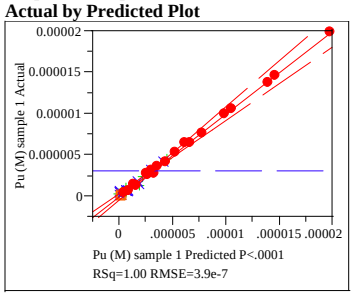


Exhibit D.7 Modified Response Surface Model Fit to Historical and Current Pu Data

Response Sample 1 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.997522
RSquare Adj	0.991504
Root Mean Square Error	3.937e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	34	8.7337e-10	2.569e-11	165.7560
Error	14	2.1696e-12	1.55e-13	Prob > F
C. Total	48	8.7554e-10		<.0001

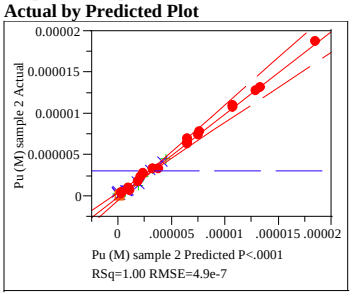
Lack Of Fit

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	10	2.1385e-12	2.138e-13	27.4897
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	14	2.1696e-12		0.0030
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	0.0000011	9.89e-7	1.15	0.2693
Al(OH)4-	0.0000078	0.000004	1.99	0.0665
CO3=	-0.000014	0.000002	-6.11	<.0001
NO2-	-8.822e-7	6.505e-7	-1.36	0.1965
NO3-	-6.875e-7	5.34e-7	-1.29	0.2188
OH- (M)	2.9373e-7	9.607e-7	0.31	0.7643
SO4=	0.0000011	0.000008	0.14	0.8924
Temperature	-3.429e-8	4.538e-8	-0.76	0.4624
CO3=*Al(OH)4-	-0.000003	0.000004	-0.79	0.4419
NO2-*Al(OH)4-	3.7219e-7	0.000002	0.24	0.8109
NO3-*Al(OH)4-	-0.000004	0.000001	-3.51	0.0035
OH- (M)*Al(OH)4-	-0.000002	7.821e-7	-2.26	0.0401
SO4=*Al(OH)4-	0.0000055	0.000008	0.65	0.5232
Temperature*Al(OH)4-	-1.036e-7	1.56e-8	-6.64	<.0001
NO2-*CO3=	0.0000021	0.000002	1.40	0.1833
NO3-*CO3=	5.8435e-7	7.248e-7	0.81	0.4336
OH- (M)*CO3=	0.0000036	3.71e-7	9.74	<.0001
SO4=*CO3=	0.0000124	0.000006	2.23	0.0424
Temperature*CO3=	2.8242e-8	2.19e-8	1.29	0.2181
NO3-*NO2-	1.6237e-7	1.269e-7	1.28	0.2214
OH- (M)*NO2-	4.3513e-8	7.235e-7	0.06	0.9529
SO4=*NO2-	0.000001	0.000002	0.54	0.5982
Temperature*NO2-	3.3772e-9	1.184e-8	0.29	0.7796
OH- (M)*NO3-	2.6833e-7	9.912e-8	2.71	0.0170
SO4=*NO3-	-0.000001	8.406e-7	-1.59	0.1344
Temperature*NO3-	-9.38e-10	4.792e-9	-0.20	0.8477
SO4=*OH- (M)	7.1202e-7	8.321e-7	0.86	0.4066
Temperature*OH- (M)	7.9972e-9	3.002e-9	2.66	0.0185
Temperature*SO4=	8.7072e-8	8.101e-8	1.07	0.3006
Al(OH)4-*Al(OH)4-	0.0000168	0.000005	3.29	0.0053
CO3=*CO3=	0.0000077	0.000002	4.27	0.0008
NO2-*NO2-	-7.286e-9	1.002e-7	-0.07	0.9431
OH- (M)*OH- (M)	2.9692e-8	6.183e-8	0.48	0.6385
SO4=*SO4=	-0.00001	0.000013	-0.79	0.4447
NO3-*NO3-	2.9828e-7	1.434e-7	2.08	0.0564

Response Sample 2 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.995948
RSquare Adj	0.986109
Root Mean Square Error	4.94e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	34	8.3972e-10	2.47e-11	101.2204
Error	14	3.416e-12	2.44e-13	Prob > F
C. Total	48	8.4314e-10		<.0001

Lack Of Fit

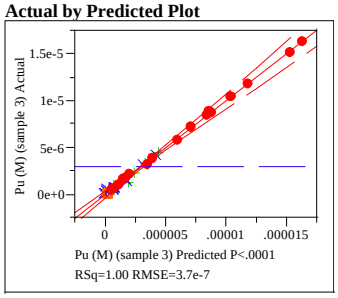
Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	10	3.3849e-12	3.385e-13	43.5121
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	14	3.416e-12		0.0012
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	6.0724e-7	0.000001	0.49	0.6322
Al(OH)4-	0.0000188	0.000005	3.83	0.0018
CO3=	-0.000009	0.000003	-3.06	0.0085
NO2-	-7.482e-7	8.163e-7	-0.92	0.3749
NO3-	-7.343e-7	6.7e-7	-1.10	0.2916
OH- (M)	-6.568e-7	0.000001	-0.54	0.5944
SO4=	-0.000016	0.00001	-1.68	0.1156
Temperature	8.4627e-9	5.695e-8	0.15	0.8840
CO3=*Al(OH)4-	-0.00001	0.000005	-1.96	0.0708
NO2-*Al(OH)4-	-0.000002	0.000002	-1.00	0.3330
NO3-*Al(OH)4-	-0.000007	0.000001	-5.14	0.0002
OH- (M)*Al(OH)4-	-0.000004	9.813e-7	-3.82	0.0019
SO4=*Al(OH)4-	0.0000424	0.000011	4.01	0.0013
Temperature*Al(OH)4-	-1.142e-7	1.957e-8	-5.83	<.0001
NO2-*CO3=	0.0000014	0.000002	0.74	0.4690
NO3-*CO3=	3.1932e-7	9.095e-7	0.35	0.7307
OH- (M)*CO3=	0.0000035	4.655e-7	7.56	<.0001
SO4=*CO3=	0.0000192	0.000007	2.75	0.0157
Temperature*CO3=	7.3196e-9	2.748e-8	0.27	0.7939
NO3-*NO2-	-1.408e-7	1.592e-7	-0.88	0.3915
OH- (M)*NO2-	8.0314e-7	9.079e-7	0.88	0.3913
SO4=*NO2-	0.0000057	0.000002	2.36	0.0330
Temperature*NO2-	-5.615e-9	1.485e-8	-0.38	0.7111
OH- (M)*NO3-	2.3209e-7	1.244e-7	1.87	0.0831
SO4=*NO3-	-0.000001	0.000001	-1.11	0.2855
Temperature*NO3-	-1.951e-8	6.013e-9	-3.24	0.0059
SO4=*OH- (M)	5.4853e-7	0.000001	0.53	0.6076
Temperature*OH- (M)	5.8729e-9	3.767e-9	1.56	0.1413
Temperature*SO4=	1.3876e-7	1.016e-7	1.37	0.1937
Al(OH)4-*Al(OH)4-	0.0000209	0.000006	3.26	0.0057
CO3=*CO3=	0.0000032	0.000002	1.41	0.1805
NO2-*NO2-	-1.85e-7	1.258e-7	-1.47	0.1635
OH- (M)*OH- (M)	8.204e-8	7.758e-8	1.06	0.3082
SO4=*SO4=	-0.000003	0.000016	-0.18	0.8576
NO3-*NO3-	7.2475e-7	1.8e-7	4.03	0.0012

Exhibit D.7 Modified Response Surface Model Fit to Historical and Current Pu Data (Continued)

Response Sample 3 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.997675
RSquare Adj	0.992028
Root Mean Square Error	3.66e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	34	8.0445e-10	2.366e-11	176.6729
Error	14	1.8749e-12	1.339e-13	Prob > F
C. Total	48	8.0633e-10		<.0001

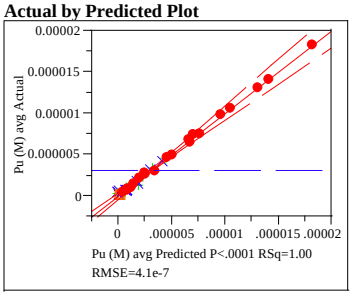
Lack Of Fit

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	10	1.8438e-12	1.844e-13	23.7017
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	14	1.8749e-12		0.0040
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	3.9965e-7	9.193e-7	0.43	0.6704
Al(OH)4-	0.0000017	0.000004	0.47	0.6423
CO3=	-0.00002	0.000002	-9.02	<.0001
NO2-	-2.078e-7	6.047e-7	-0.34	0.7363
NO3-	-2.97e-7	4.964e-7	-0.60	0.5591
OH- (M)	-2.389e-9	8.931e-7	-0.00	0.9979
SO4=	0.0000052	0.000007	0.72	0.4849
Temperature	-1.478e-8	4.219e-8	-0.35	0.7313
CO3=*Al(OH)4-	-0.000005	0.000004	-1.41	0.1795
NO2-*Al(OH)4-	5.8412e-7	0.000001	0.41	0.6868
NO3-*Al(OH)4-	-0.000002	0.000001	-1.96	0.0699
OH- (M)*Al(OH)4-	0.0000015	7.27e-7	2.00	0.0658
SO4=*Al(OH)4-	0.0000055	0.000008	0.70	0.4925
Temperature*Al(OH)4-	-1.52e-7	1.45e-8	-10.48	<.0001
NO2-*CO3=	5.1294e-7	0.000001	0.36	0.7229
NO3-*CO3=	0.0000025	6.738e-7	3.77	0.0021
OH- (M)*CO3=	0.0000046	3.449e-7	13.21	<.0001
SO4=*CO3=	0.0000177	0.000005	3.42	0.0041
Temperature*CO3=	3.2152e-8	2.036e-8	1.58	0.1367
NO3-*NO2-	1.0746e-7	1.179e-7	0.91	0.3777
OH- (M)*NO2-	3.5933e-7	6.726e-7	0.53	0.6016
SO4=*NO2-	-6.42e-7	0.000002	-0.36	0.7228
Temperature*NO2-	-1.016e-8	1.1e-8	-0.92	0.3713
OH- (M)*NO3-	2.1613e-7	9.215e-8	2.35	0.0343
SO4=*NO3-	-0.000002	7.815e-7	-2.19	0.0457
Temperature*NO3-	8.9304e-9	4.455e-9	2.00	0.0647
SO4=*OH- (M)	5.1816e-8	7.736e-7	0.07	0.9475
Temperature*OH- (M)	5.9143e-9	2.791e-9	2.12	0.0524
Temperature*SO4=	1.3449e-8	7.53e-8	0.18	0.8608
Al(OH)4-*Al(OH)4-	0.0000105	0.000005	2.20	0.0448
CO3=*CO3=	0.0000101	0.000002	6.01	<.0001
NO2-*NO2-	7.089e-8	9.319e-8	0.76	0.4594
OH- (M)*OH- (M)	3.9105e-8	5.747e-8	0.68	0.5074
SO4=*SO4=	-0.000005	0.000012	-0.45	0.6596
NO3-*NO3-	4.2735e-8	1.333e-7	0.32	0.7533

Response Average Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.997152
RSquare Adj	0.990237
Root Mean Square Error	4.087e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	34	8.1881e-10	2.408e-11	144.1851
Error	14	2.3384e-12	1.67e-13	Prob > F
C. Total	48	8.2115e-10		<.0001

Lack Of Fit

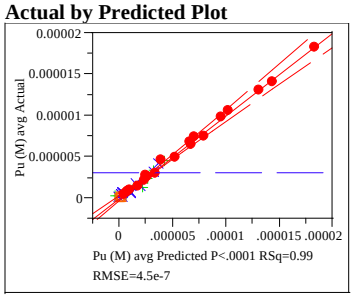
Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	10	2.3072e-12	2.307e-13	29.6592
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	14	2.3384e-12		0.0026
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	7.148e-7	0.000001	0.70	0.4977
Al(OH)4-	0.0000094	0.000004	2.32	0.0357
CO3=	-0.000014	0.000002	-5.88	<.0001
NO2-	-6.127e-7	6.753e-7	-0.91	0.3796
NO3-	-5.73e-7	5.544e-7	-1.03	0.3189
OH- (M)	-1.218e-7	9.974e-7	-0.12	0.9045
SO4=	-0.000003	0.000008	-0.42	0.6826
Temperature	-1.354e-8	4.712e-8	-0.29	0.7781
CO3=*Al(OH)4-	-0.000006	0.000004	-1.46	0.1654
NO2-*Al(OH)4-	-3.213e-7	0.000002	-0.20	0.8422
NO3-*Al(OH)4-	-0.000004	0.000001	-3.78	0.0020
OH- (M)*Al(OH)4-	-0.000001	8.119e-7	-1.67	0.1173
SO4=*Al(OH)4-	0.0000178	0.000009	2.04	0.0609
Temperature*Al(OH)4-	-1.233e-7	1.619e-8	-7.61	<.0001
NO2-*CO3=	0.0000014	0.000002	0.86	0.4057
NO3-*CO3=	0.0000011	7.525e-7	1.52	0.1498
OH- (M)*CO3=	0.0000039	3.852e-7	10.12	<.0001
SO4=*CO3=	0.0000164	0.000006	2.85	0.0129
Temperature*CO3=	2.2571e-8	2.274e-8	0.99	0.3377
NO3-*NO2-	4.3019e-8	1.317e-7	0.33	0.7488
OH- (M)*NO2-	4.0199e-7	7.511e-7	0.54	0.6009
SO4=*NO2-	0.000002	0.000002	1.02	0.3261
Temperature*NO2-	-4.134e-9	1.229e-8	-0.34	0.7416
OH- (M)*NO3-	2.3885e-7	1.029e-7	2.32	0.0359
SO4=*NO3-	-0.000001	8.727e-7	-1.61	0.1293
Temperature*NO3-	-3.838e-9	4.975e-9	-0.77	0.4533
SO4=*OH- (M)	4.3745e-7	8.639e-7	0.51	0.6205
Temperature*OH- (M)	6.5948e-9	3.117e-9	2.12	0.0527
Temperature*SO4=	7.976e-8	8.41e-8	0.95	0.3590
Al(OH)4-*Al(OH)4-	0.000016	0.000005	3.03	0.0090
CO3=*CO3=	0.000007	0.000002	3.73	0.0022
NO2-*NO2-	-4.046e-8	1.041e-7	-0.39	0.7033
OH- (M)*OH- (M)	5.0279e-8	6.419e-8	0.78	0.4465
SO4=*SO4=	-0.000006	0.000013	-0.46	0.6523
NO3-*NO3-	3.5526e-7	1.489e-7	2.39	0.0317

Exhibit D.7 Stepwise Model Fit to the Historical Data with, in turn, the Average and Individual Sample Pu Data from this Study

Response Average Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.993479
RSquare Adj	0.987962
Root Mean Square Error	4.538e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	22	8.1579e-10	3.708e-11	180.0582
Error	26	5.3545e-12	2.059e-13	Prob > F
C. Total	48	8.2115e-10		<.0001

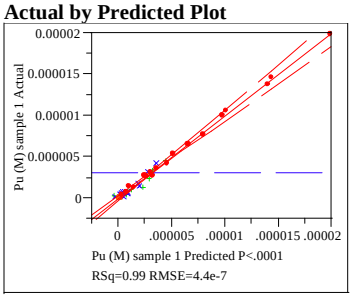
Lack Of Fit

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	22	5.3234e-12	2.42e-13	31.1050
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	26	5.3545e-12		0.0021
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-1.015e-7	2.976e-7	-0.34	0.7358
Al(OH)4-	0.000002	0.000002	0.94	0.3540
CO3=	-0.000015	0.000002	-6.68	<.0001
NO2-	-2.371e-7	1.44e-7	-1.65	0.1117
NO3-	7.5769e-7	1.229e-7	6.17	<.0001
OH- (M)	7.3388e-7	5.063e-8	14.49	<.0001
SO4=	-0.000002	0.000003	-0.65	0.5225
Temperature	-3.535e-8	8.106e-9	-4.36	0.0002
CO3=*Al(OH)4-	-0.000004	0.000001	-2.80	0.0096
NO2-*Al(OH)4-	0.0000011	4.122e-7	2.78	0.0100
NO3-*Al(OH)4-	-0.000002	4.818e-7	-4.08	0.0004
SO4=*Al(OH)4-	0.0000129	0.000005	2.54	0.0173
Temperature*Al(OH)4-	-1.164e-7	1.493e-8	-7.79	<.0001
NO2-*CO3=	0.0000015	3.115e-7	4.82	<.0001
NO3-*CO3=	0.0000013	2.153e-7	6.12	<.0001
OH- (M)*CO3=	0.0000037	2.411e-7	15.42	<.0001
SO4=*CO3=	0.0000123	0.000004	3.51	0.0016
Temperature*CO3=	2.2612e-8	1.028e-8	2.20	0.0369
SO4=*NO3-	-0.000001	4.791e-7	-2.85	0.0084
Temperature*OH- (M)	8.1391e-9	9.45e-10	8.61	<.0001
Temperature*SO4=	9.2169e-8	3.052e-8	3.02	0.0056
Al(OH)4-*Al(OH)4-	0.0000108	0.000003	3.65	0.0012
CO3=*CO3=	0.0000089	0.000001	6.00	<.0001

Response Sample 1 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.994357
RSquare Adj	0.989581
Root Mean Square Error	4.359e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	22	8.706e-10	3.957e-11	208.2314
Error	26	4.9411e-12	1.9e-13	Prob > F
C. Total	48	8.7554e-10		<.0001

Lack Of Fit

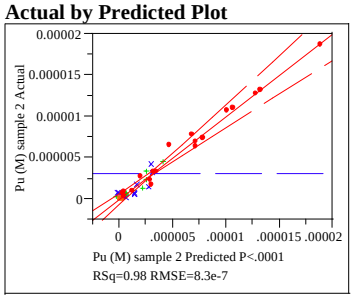
Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	22	4.91e-12	2.232e-13	28.6894
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	26	4.9411e-12		0.0025
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-4.931e-7	2.859e-7	-1.73	0.0964
Al(OH)4-	6.0509e-7	0.000002	0.30	0.7650
CO3=	-0.000015	0.000002	-6.93	<.0001
NO2-	-1.561e-7	1.383e-7	-1.13	0.2693
NO3-	7.7117e-7	1.18e-7	6.53	<.0001
OH- (M)	8.3717e-7	4.864e-8	17.21	<.0001
SO4=	7.8905e-7	0.000003	0.29	0.7772
Temperature	-2.717e-8	7.787e-9	-3.49	0.0017
CO3=*Al(OH)4-	-0.000002	0.000001	-1.66	0.1088
NO2-*Al(OH)4-	0.000001	3.96e-7	2.58	0.0158
NO3-*Al(OH)4-	-0.000002	4.628e-7	-4.16	0.0003
SO4=*Al(OH)4-	0.0000046	0.000005	0.94	0.3547
Temperature*Al(OH)4-	-1.022e-7	1.435e-8	-7.12	<.0001
NO2-*CO3=	0.0000017	2.993e-7	5.66	<.0001
NO3-*CO3=	9.1776e-7	2.068e-7	4.44	0.0001
OH- (M)*CO3=	0.0000033	2.316e-7	14.26	<.0001
SO4=*CO3=	0.0000118	0.000003	3.51	0.0016
Temperature*CO3=	2.6772e-8	9.875e-9	2.71	0.0117
SO4=*NO3-	-0.000001	4.602e-7	-2.76	0.0105
Temperature*OH- (M)	7.9575e-9	9.08e-10	8.76	<.0001
Temperature*SO4=	4.9842e-8	2.932e-8	1.70	0.1011
Al(OH)4-*Al(OH)4-	0.0000129	0.000003	4.55	0.0001
CO3=*CO3=	0.0000091	0.000001	6.34	<.0001

Exhibit D.7 Stepwise Model Fit to the Historical Data with, in turn, the Average and Individual Sample Pu Data from this Study (continued)

Response Sample 2 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.978685
RSquare Adj	0.96065
Root Mean Square Error	8.314e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	22	8.2517e-10	3.751e-11	54.2645
Error	26	1.7971e-11	6.912e-13	Prob > F
C. Total	48	8.4314e-10		<.0001

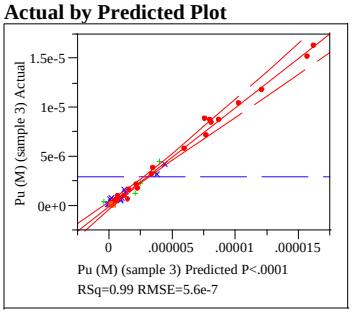
Lack Of Fit

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	22	1.794e-11	8.155e-13	104.8262
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	26	1.7971e-11		0.0002
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	5.3117e-7	5.452e-7	0.97	0.3389
Al(OH)4-	1.9755e-7	0.000004	0.05	0.9592
CO3=	-0.000009	0.000004	-2.18	0.0381
NO2-	-4.019e-7	2.638e-7	-1.52	0.1397
NO3-	7.8594e-7	2.251e-7	3.49	0.0017
OH- (M)	6.5095e-7	9.276e-8	7.02	<.0001
SO4=	-0.000004	0.000005	-0.83	0.4131
Temperature	-5.748e-8	1.485e-8	-3.87	0.0007
CO3=*Al(OH)4-	-6.279e-7	0.000003	-0.23	0.8192
NO2-*Al(OH)4-	0.000002	7.552e-7	2.63	0.0143
NO3-*Al(OH)4-	-0.000001	8.827e-7	-1.42	0.1672
SO4=*Al(OH)4-	0.0000192	0.000009	2.07	0.0488
Temperature*Al(OH)4-	-1.08e-7	2.736e-8	-3.95	0.0005
NO2-*CO3=	0.0000015	5.708e-7	2.61	0.0150
NO3-*CO3=	5.515e-7	3.944e-7	1.40	0.1738
OH- (M)*CO3=	0.0000032	4.417e-7	7.32	<.0001
SO4=*CO3=	0.000005	0.000006	0.78	0.4435
Temperature*CO3=	-3.393e-9	1.883e-8	-0.18	0.8584
SO4=*NO3-	-0.000001	8.777e-7	-1.62	0.1166
Temperature*OH- (M)	1.0529e-8	1.732e-9	6.08	<.0001
Temperature*SO4=	1.6584e-7	5.591e-8	2.97	0.0064
Al(OH)4-*Al(OH)4-	0.0000066	0.000005	1.22	0.2339
CO3=*CO3=	0.0000073	0.000003	2.69	0.0124

Response Sample 3 Pu (M)
Actual by Predicted Plot



Summary of Fit

RSquare	0.989925
RSquare Adj	0.981399
Root Mean Square Error	5.59e-7
Mean of Response	0.000003
Observations (or Sum Wgts)	49

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	22	7.9821e-10	3.628e-11	116.1166
Error	26	8.124e-12	3.125e-13	Prob > F
C. Total	48	8.0633e-10		<.0001

Lack Of Fit

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	22	8.0929e-12	3.679e-13	47.2878
Pure Error	4	3.1117e-14	7.779e-15	Prob > F
Total Error	26	8.124e-12		0.0009
				Max RSq
				1.0000

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	-3.425e-7	3.665e-7	-0.93	0.3587
Al(OH)4-	0.0000051	0.000003	1.99	0.0577
CO3=	-0.000021	0.000003	-7.62	<.0001
NO2-	-1.533e-7	1.774e-7	-0.86	0.3954
NO3-	7.1595e-7	1.513e-7	4.73	<.0001
OH- (M)	7.1354e-7	6.237e-8	11.44	<.0001
SO4=	-0.000002	0.000004	-0.56	0.5772
Temperature	-2.14e-8	9.985e-9	-2.14	0.0417
CO3=*Al(OH)4-	-0.000009	0.000002	-5.17	<.0001
NO2-*Al(OH)4-	4.2841e-7	5.078e-7	0.84	0.4065
NO3-*Al(OH)4-	-0.000003	5.935e-7	-4.57	0.0001
SO4=*Al(OH)4-	0.0000149	0.000006	2.38	0.0249
Temperature*Al(OH)4-	-1.389e-7	1.84e-8	-7.55	<.0001
NO2-*CO3=	0.0000013	3.838e-7	3.45	0.0019
NO3-*CO3=	0.0000025	2.652e-7	9.37	<.0001
OH- (M)*CO3=	0.0000046	2.97e-7	15.56	<.0001
SO4=*CO3=	0.0000201	0.000004	4.65	<.0001
Temperature*CO3=	4.4457e-8	1.266e-8	3.51	0.0016
SO4=*NO3-	-0.000001	5.901e-7	-2.38	0.0252
Temperature*OH- (M)	5.9309e-9	1.165e-9	5.09	<.0001
Temperature*SO4=	6.0825e-8	3.759e-8	1.62	0.1177
Al(OH)4-*Al(OH)4-	0.0000128	0.000004	3.53	0.0016
CO3=*CO3=	0.0000104	0.000002	5.68	<.0001