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UC-2000

Fiscal Year 1995 Laboratory Scale Studies of Cs Elution in Tank 8D-1 and Sludge Dissolution in Tank 8D-2

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April 1996

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
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QA Level

This work was performed to satisfy WVSP FY 1996 milestone number 0603-A and was conducted in accordance with Impact Level II quality assurance requirements as defined by the WVSP Quality Assurance Plan WTC-007 and the PNNL *Quality Assurance Manual* PNNL-MA-70.

Summary

During phase I of the West Valley Demonstration Project (WVDP) waste remediation, an estimated 95% of the zeolite currently in tank 8D-1 will be transferred to tank 8D-2, leaving behind residual Cs-loaded zeolite which will require treatment to remove the Cs. After phase I vitrification, tank 8D-2 will contain residual waste, which will consist of waste from the Plutonium and Uranium Extraction (PUREX) and Thorium Extraction (THOREX) processes and spent Cs-loaded zeolite. The residual waste will require treatment to dissolve the waste and transfer the radionuclides to the vitrification plant.

Oxalic acid has been proposed as a reagent for the elution of Cs from zeolite in tank 8D-1 and for the dissolution of sludge and mobilization of radionuclides in tank 8D-2. Nonradioactive laboratory-scale tests were performed to determine optimum Cs elution and sludge dissolution conditions in terms of acid-to-zeolite/sludge ratio, contact time, and temperature of acid addition, and to evaluate the effects of multiple contacts, long-term contacts, presence of corrosion products, lack of agitation, temperature of tank contents, and oxalic acid concentration. An evaluation of the potential for increased corrosion of tank 8D-1 during Cs elution was also performed, using laboratory-scale nonradioactive tests with mild-steel specimens.

The optimum conditions for Cs elution were selected based on process simplicity, as no one set of parameters tested resulted in a distinguishably higher Cs elution efficiency. The optimum conditions selected were 40 L of 8 wt% oxalic acid per kg of zeolite added to the tank at ~25°C. A multiple-contact process that involves three two-hour contacts with oxalic acid, with removal of the acid after each contact, appears to be optimal for Cs elution; 99% of the Cs present is eluted during the process. Contact times longer than two hours did not appear to increase elution of Cs.

The optimum conditions for sludge dissolution were also selected based on process simplicity; no one set of parameters was distinguishably better, although slightly higher dissolution of many of the ions present occurred at higher acid-to-sludge ratios (40 and 80 L acid/kg sludge) and higher acid addition temperature (80°C). The optimum conditions selected were 40 L of 8 wt% oxalic acid per kg sludge added to the tank at ~25°C, with an optimum contact time of two hours. Under these conditions, 70-75 wt% of the solids present in the sludge can be dissolved, although the dissolution of the radionuclides may be significantly lower. Under these conditions, 90-100% of the Fe, the major sludge component, may be dissolved.

Tests in which Fe_2O_3 was added to simulate corrosion products showed that up to 90% of loose Fe in the form of rust may be dissolved by treatment with 8 wt% oxalic acid. This amount may be unacceptable in terms of the vitrification flowsheet. Cesium elution tests that used 4 and 1 wt% oxalic acid showed that only 15-20% of the Fe may be dissolved; however, only 75% of the Cs was eluted in these tests.

Laboratory-scale tests designed to evaluate the potential for increased corrosion of tank 8D-1 during the oxalic acid Cs elution process examined mild-steel specimens at elevated temperature (50°C) in two acid concentrations (4 and 8 wt%) for durations of up to 3 weeks. These tests showed that even at relatively modest temperatures (50°C) and short exposures to oxalic acid (1-3 weeks), the corrosion rates observed in the test specimens were quite high, greater than

150 mils per year in some cases. Corrosion rates were found to be higher on specimens contacted with 4 wt% oxalic acid than on those contacted with 8 wt% oxalic acid. The corrosion observed in these tests appeared as localized pitting and crevice corrosion, especially at 4 wt% oxalic acid. Vapor-space corrosion was negligible at both 4 and 8 wt% oxalic acid. Considering the uncertainty in corrosion rates because of the short exposure times, vapor-space corrosion rates were comparable to corresponding results from similar testing.

Future testing should include identification of surface films to help understand the reason for lower corrosion rates at higher acid concentrations. Follow-on testing should include agitation of some of the vessels to estimate the impact of mixer-pump operation on tank corrosion during Cs elution processing. As potential processing conditions become better defined, the temperature investigated for these tests (50°C) may not be appropriate. Corrosion testing should be performed at or above the maximum expected Cs elution processing temperature.

Computer modeling of oxalate-treated sludge at thermodynamic equilibrium indicated that essentially all of the species in the sludge, including the radionuclides U and Th, are readily soluble in this system. The oxalic acid-sludge system was not solubility-limited for any of the species investigated, except Ce, Si and Zr, which supports the experimental findings that essentially all of the Fe present in the tank as iron compounds or iron oxides is capable of being dissolved. Cerium was used as a surrogate for the radionuclides U and Th, and was chosen by the virtue that it appeared to form similar oxalate compounds to U and Th. The modeling work showed that Ce was a poor choice of surrogate; as the modeling results showed it to be insoluble, while U and Th would be completely soluble.

Acronyms

DOE	Department of Energy
EPA	Environmental Protection Agency
ESP	Environmental Simulation Program
FY	Fiscal Year
HLW	High Level Waste
IC	Ion Chromatography
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
PNNL	Pacific Northwest National Laboratory
PUREX	Plutonium and Uranium Extraction
QA	Quality Assurance
THOREX	Thorium Extraction Process
WVDP	West Valley Demonstration Project
WVNS	West Valley Nuclear Services
WVSP	West Valley Support Project
XRF	X-Ray Fluorescence

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1.0 Introduction

This report details work performed as part of the West Valley Support Program (WVSP) Subtask 0604, conducted by the Pacific Northwest National Laboratory (PNNL), and is in support of residual waste removal during high-level waste (HLW) tank stabilization activities performed by the West Valley Demonstration Project (WVDP). The HLW, approximately 2 million liters, originated from a commercial nuclear fuel-reprocessing plant formerly located at West Valley, New York.

The largest volume fraction of waste was generated during reprocessing of spent U fuel using the PUREX process. Waste from the PUREX process was neutralized with NaOH for storage in a carbon-steel tank designated 8D-2. Neutralization resulted in a precipitated hydroxide sludge which settled to the bottom of the tank and is covered by a supernatant salt solution. Acidic THOREX waste was generated by the processing of Th fuel from the Indian Point I reactor using the THOREX process. This waste, approximately 55,000 liters, was stored in a stainless-steel tank designated 8D-3, and has recently been transferred to 8D-2. Cesium-loaded zeolite was generated by a supernatant decontamination process involving ion exchange. The exchange columns and the spent zeolite from this processing are stored in a carbon-steel tank designated 8D-1 (Cadoff 1991).

During phase I of the WVDP waste remediation, an estimated 95 wt% of the zeolite will be removed from 8D-1 and transferred to 8D-2, leaving behind residual Cs-loaded zeolite, which will require treatment to remove the highly radioactive Cs(a). Tank 8D-2 will contain residual waste after phase I vitrification, consisting of waste from the PUREX and THOREX processes and spent Cs-loaded zeolite. The residual waste remaining in 8D-2 will also require treatment to dissolve the waste and transfer the radionuclides to the vitrification plant.

Oxalic acid has been studied as a decontamination reagent for nuclear reactors and equipment (Meservey 1970). Oxalic acid may also be used to elute Cs from zeolite and to dissolve sludge. Previous laboratory studies(b) show that oxalic acid can be used to elute Cs from zeolite in tank 8D-1 at a level of 88-90% over a period of 11 days (264 hours) when 0.8 M (7 wt%) oxalic acid is added to zeolite at ~20 L acid/kg zeolite. These studies have also found that a maximum of 66 wt% of washed sludge can be dissolved using 0.8 M oxalic acid. In addition, Bradley and Hill (1977) reported that over 95 vol% of the sludge from the Savannah River Plant's Tank 16H could be dissolved using 8 wt% oxalic acid at 85°C with agitation in a two-step dissolution process (50 hours per step), and with an initial oxalic acid-solution-to-sludge volume of 20:1.

(a) Schiffhauer, MA. 1994. "Review of Zeolite Transfer Operation and Estimated Duration," Internal West Valley Demonstration Project Memo dated March 24, 1994. West Valley Demonstration Project, West Valley, NY.

(b) Bray, LA. 1986. "Washed Sludge and Cesium/Zeolite Dissolution Studies," Letter report WVST 86/104. West Valley Nuclear Services, West Valley, NY.

This report summarizes the findings of laboratory experiments that were performed with the following primary objectives:

- to determine the optimum process conditions which cause maximum Cs elution (target value 99 wt%) from zeolite in tank 8D-1 using oxalic acid, with a target value of 99 wt%;
- to determine the optimum process conditions to cause maximum Cs elution while minimizing Fe dissolution;
- to determine the optimum process conditions to cause maximum sludge dissolution by oxalic acid in tank 8D-2;
- to investigate the effect of tank corrosion products on the oxalic acid treatment process in tanks 8D-1 and 8D-2; and
- to investigate the potential for increased corrosion of tank 8D-1 because of contact with oxalic acid.

2.0 Conclusions and Recommendations

2.1 Cs-Elution Studies

Twenty-nine Cs-elution tests were run in six sets of experiments. The experiments included (1) determination of optimum process conditions for the elution of Cs from zeolite; (2) repeatability tests, in which one set of test conditions was run 4 times; (3) long-term tests with contact times up to 11 days; (4) multiple-contact tests using three two-hour contacts; (5) tests to determine the effects of condition modifications, such as the presence of corrosion products and carbon-steel coupons, no agitation, and temperature of tank contents; and (6) tests to determine the effect of varying oxalic acid concentration (1 wt% - 8 wt%) on Cs elution and Fe dissolution.

The optimum process conditions for Cs elution were selected based on process simplicity, as no one set of parameters tested resulted in a distinguishably higher Cs-elution efficiency. The optimum conditions selected were 40 L of 8 wt% oxalic acid per kg of zeolite added to the tank at ~25°C for one contact lasting a minimum of two hours. Under these conditions, ~80 wt% of the Cs was eluted from the zeolite.

The majority of the Cs elution appeared to occur during the first two hours of contact, and longer contact times did not improve Cs elution. Contact times as long as 11 days did not result in significantly higher levels of Cs elution.

A multiple-contact process involving three two-hour contacts with oxalic acid, with removal of the acid after each contact, dramatically improved Cs elution over the single-contact process, eluting 99 wt% of the Cs present. In full-scale operations, the multiple-contact option would increase the duration of the overall process and the volume of water to be evaporated.

In the tests involving effects of condition modifications, addition of corrosion products, presence of carbon-steel coupons, and lack of agitation all produced approximately the same level of Cs elution as the control after 50 hours of contact with oxalic acid. Using a 25°C bath instead of a 50°C bath appeared to decrease Cs elution slightly. Addition of Fe_2O_3 did not appear to have a significant effect on the elution of Cs, indicating that the presence of corrosion products in tank 8D-1 will probably not have a large effect on plant-scale Cs-elution operations at the ratio of 40 L of 8 wt% oxalic acid per kg of zeolite. However, almost all of the Fe_2O_3 added to the system dissolved, indicating that the rust present in the tank may be extremely soluble in 8 wt% oxalic acid, which would result in increased amounts of Fe going to vitrification. This could increase the number of glass logs produced by vitrification. The presence of carbon-steel coupons increased the amount of Fe in solution beyond what would be expected from the Fe content of the zeolite, suggesting that oxalic acid may be somewhat corrosive to carbon steel under the test conditions.

Tests using lower concentrations of oxalic acid (4 and 1 wt%) indicated that decreasing the oxalic-acid concentration decreases Fe solubilization to 15-20 wt% of the Fe in the system. However, Cs-elution efficiency also drops to ~75 wt% in three contacts.

In summary, the "optimum" conditions chosen for Cs elution were 40 L 8 wt% oxalic acid/kg zeolite added to the tank at ~25°C. The data indicate that a multiple-contact process involving three two-hour contacts with oxalic acid, with removal of the supernatant after each contact, is optimal for Cs elution, eluting 99 wt% of the Cs present during the course of the tests. Under the conditions examined, a two-hour contact time appeared to be effective, but contact times up to 11 days do not appear detrimental to the elution of Cs from zeolite (although contact times this long may certainly be detrimental to the tank). Since the elution of Cs from zeolite is a physical process rather than a chemical one, it stands to reason that contact with any agent that can dissolve zeolite will release some portion of the Cs. If tank corrosion is a concern with the oxalic acid treatment process, perhaps another reagent can be found which will dissolve the zeolite but will not contribute to tank structure corrosion.

2.2 Sludge-Dissolution Studies

Sixteen sludge-dissolution tests were run in five sets of experiments. The experiments included (1) determination of optimum process conditions for the dissolution of sludge; (2) repeatability tests, in which one set of test conditions was run 4 times; (3) long-term tests with contact times of up to 11 days; (4) multiple-contact tests using three two-hour contacts; and (5) tests to determine the effects of condition modifications such as the presence of corrosion products and carbon-steel coupons, no agitation, and temperature of tank contents.

The optimum process conditions for sludge dissolution were chosen based on process simplicity, as no one set of parameters resulted in distinguishably higher levels of dissolution of all sludge elements. The conditions chosen were 40 L 8 wt% oxalic acid per kg sludge added at 25°C in a single contact lasting a minimum of four hours. Contact time did not appear to be a factor in sludge dissolution; significant changes in the percentage of each ion dissolved were not observed after 4 hours contact time, and in most cases, >70 wt% of each ion present had dissolved after 2 hours contact time.

Long-term testing indicated that the fraction of most sludge elements dissolved remains constant upon long term (up to 11 days) exposure to oxalic acid (Mg and Sr, which show a decrease in solubility with time, are the only exceptions).

Multiple-contact tests involving three two-hour contacts with oxalic acid were not found to be advantageous in sludge-dissolution experiments. The fraction of sludge elements remained constant, within experimental error, over all three contacts.

In tests involving the effects of condition modifications, addition of corrosion products, presence of carbon-steel coupons, lack of agitation, and a 25°C bath temperature (as opposed to the 50°C bath used for most of the tests) did not have a significant effect on the dissolution of most of the sludge elements present in the simulant. Addition of Fe_2O_3 had very little effect on laboratory-scale sludge dissolution, suggesting that the rust present in tank 8D-2 will probably not have a large effect on plant-scale sludge-dissolution operations at the ratio of 40 L 8 wt% oxalic acid per kg sludge. However, because most of the Fe_2O_3 ended up in solution, its presence could increase the number of glass logs produced by vitrification. Presence of carbon-steel coupons increases the amount of Fe in solution, suggesting that oxalic acid may be somewhat corrosive to carbon steel under the test conditions.

In summary, the "optimum" conditions for sludge dissolution were chosen to be 40 L 8 wt% oxalic acid per kg sludge, added at ~25°C, with most dissolution occurring within 4 hours. The data indicate that under these conditions, 70-75 wt% of the solids present in the sludge can be dissolved. Under these conditions, 90-100 wt% of the Fe, the major sludge component, may be dissolved.

2.3 Mild-Steel Corrosion Studies

The following conclusions and recommendations are based on results of work presented in this report to evaluate the corrosiveness of oxalic acid solutions to mild steel.

Even at relatively modest temperatures (50°C) and short exposures to oxalic acid (1 to 3 weeks), the corrosion rate of A516 Grade 55 mild-steel test specimens is quite high (> 150 mils per year). It is uncertain how similar the effect would be with Tank 8D-1, since the present condition of the tank has not been well characterized (moderate to severe degrees of corrosion on the inside and outside surfaces of the tank is suspected). Oxalic acid dissolution of the already corroded tank wall may be significantly different from the rates observed on the initially smooth, clean test specimens. The presence of heavy oxide scale on the tank wall and other surfaces will probably affect corrosion of the steel below. Also, because of the corrosion that has already occurred in the tank, the metal surface is no longer smooth. Pitting may proceed faster on a surface that, unlike the test coupons, is already corroded.

The corrosion appeared as localized pitting and crevice corrosion, especially with the 4 wt% acid test specimens. However, the pitting occurred evenly over the entire surface of the submerged specimens, so the results calculated from weight-loss measurements are considered to be reasonably accurate estimates of the actual penetration rates.

Corrosion rates measured during testing were higher on specimens contacted with the 4 wt% acid solution than with the 8 wt% acid solution (> 150 mils per year). The magnitude of this difference was not expected, and the reason for this seemingly reversed order is not yet clear; it may be due to different surface films formed by reaction products at the different conditions, and/or the solubility of oxalic acid at these conditions.

A thick yellow layer of iron oxalate accumulated on the surfaces of the test specimens in the 4 wt% acid. A much thinner layer of iron oxalate was found on the specimens in the 8 wt% acid. The effects of this layer on corrosion of the steel was not evaluated as part of this effort; however, it was observed that the conditions which produced the thicker layer (4 wt% acid) also resulted in the higher corrosion rates. These tests were conducted without agitation. An agitated system (resembling the anticipated mixing of the tanks during Cs elution) may result in significantly different corrosion rates.

Vapor-space corrosion was negligible at these conditions for specimens in both the 4 wt% and 8 wt% acid vessels (1 to 2 mils per year). Considering the uncertainty in calculated corrosion rates due to the short exposure times of the tests, vapor-space corrosion rates were comparable to corresponding results from other testing (e.g., sludge-wash corrosion tests, nitrogen-inerting corrosion tests, etc.) previously conducted at PNNL for the WVDP.

Since the testing reported here resulted in some unexpected observations, and because of potential impacts to Cs-elution operations, it is recommended that some of this testing be repeated, and additional conditions be tested (e.g., 4, 6, and 8 wt% acid). Higher acid concentrations resulting in lower corrosion rates on mild steel may be very desirable for Cs elution. The reason for this observed result needs to be further investigated and better understood.

Additional follow-on testing should include agitation in some of the vessels to assess the impact of mixer-pump operation on tank corrosion during Cs-elution processing. Agitation is expected to increase corrosion rate for both 4 wt% and 8 wt% acid conditions; however, because the iron-oxalate precipitate was much more loosely adhered in the 4 wt% acid tests, relatively more should be removed by stirring. Therefore, the corrosion rate increase due to agitation is expected to have a greater effect with the 4 wt% acid tests.

As potential processing conditions become better defined, the temperature that was used for these tests (50°C) may not be appropriate. Corrosion testing should be performed slightly above the maximum expected Cs-elution processing temperature.

2.4 Modeling Studies

Results of computer modeling of oxalate-treated sludge at thermodynamic equilibrium show that most of the components of constituents associated with the sludge are completely soluble in quantities of oxalic acid greater than 20 L 8 wt% oxalic acid/kg sludge. Exceptions are Ce, which was used as a surrogate for U and Th in laboratory-scale testing, and Zr and Si, which are found in the zeolite. The solubility of these three constituents does not appear to be influenced by the quantity of oxalic acid added; all three elements are essentially insoluble.

In contrast, both U and Th were dissolved readily, indicating that Ce was a poor choice of surrogate for U and Th in this system.

In summary, the modeling work indicates several things of importance with respect to the use of oxalic acid for the dissolution of sludge:

(1) The oxalic acid-sludge system is not solubility-limited for any species investigated, except Ce and Zr. This finding is important as it indicates that essentially all of the Fe present in the tank as iron compounds (e.g., iron oxides) is capable of being dissolved. In addition, if a corresponding redox couple is available, it is possible that Fe from the walls of the tank could be dissolved into the system.

(2) Additional data for zeolite IE-96 should be obtained to improve the accuracy of the predictions of this model further.

3.0 Cesium Elution: Use of Oxalic Acid to Elute Cs from Zeolite in Tank 8D-1

3.1 Procedures

3.1.1 Zeolite Loading

The IE-96 zeolite was loaded to a level of 0.8862 g Cs/kg zeolite (a concentration of 886 ug/g) by contacting it with $CsNO_3$ overnight (see Appendix A for calculation of the required level of Cs loading). The contact conditions are shown in Table 3.1. After contact, the solution was stirred, allowed to settle, and the $CsNO_3$ solution was decanted. A sample of the zeolite was placed in a drying oven and analyzed by X-ray fluorescence (XRF) and inductively coupled plasma mass spectroscopy (ICP-MS) for Cs content. The results are shown in Table 3.1. As the table shows, both ICP-MS and XRF indicated higher levels of Cs present than calculated; however, the XRF number is well within experimental error, and the ICP-MS number is only slightly higher than the ±10% experimental error expected in the measurements. Because there is no impact level II procedure for ICP-MS on solids in place, the XRF value was used in calculations as the initial Cs loading in the zeolite.

When exposed to air, zeolite absorbs some of the moisture in the air. The amount of moisture absorbed is a function of the relative humidity of the air, and thus can vary depending on ambient conditions. For this reason, it is usual to refer to the weight of zeolite in terms of dry weight rather than absolute weight. The two weights are related by a quantity known as the F-factor, as shown in Equation 3.1. The weight of an amount of zeolite used can then be scaled to the dry weight by multiplying the weight used by the F-factor. For these experiments, the F-factor was determined by weighing ~1g of zeolite (wet weight), placing it in a drying oven at ~105°C for 24 hours, and weighing again (dry weight). Using this method, an F-factor of 0.9482 was determined for the zeolite immediately after loading.

$$\text{F factor} = \text{dry weight} / \text{wet weight} \qquad 3.1$$

The Cs-elution experiments took place over a period of several months, and it was noted after the third set of tests had been run that the appearance of the loaded zeolite had changed from damp to dry, presumably due to evaporation of residual $CsNO_3$ solution. A new F-factor of 0.7241 was determined, significantly different from the original F-factor. The fact that the new

Table 3.1. Contact Conditions and Analytical Results for Cs Loading of Zeolite

Contact Conditions		Analytical Results	
Dry weight zeolite, g	250	Calculated Cs loading, ug/g	885
Weight $CsNO_3$, g	0.3253	Average Cs loading by XRF, ug/g	957
Volume water, mL	500	Average Cs loading by ICP-MS, ug/g	977

F-factor is smaller than the original F-factor indicates that the moisture content of the zeolite had actually increased rather than decreased. It is likely that instead of evaporating, the residual $CsNO_3$ was actually absorbed, along with moisture from the air.

Through the course of the first three experiments, the wet weight of zeolite used did not change, and therefore as time went on, smaller and smaller dry weights were being used. This means that in the first three experiments, there was less Cs present than calculated, and weight percent of Cs eluted was therefore low for these experiments. Since the zeolite appearance changed relatively quickly (the change was noted after the first experiment, but not acted on immediately), it was decided that the new F-factor of 0.724 would be more representative than the original one, and so this value was used in the calculations for the first three experiments. Since it is impossible to know the exact rate of moisture absorption, it is not possible to correct the results any further than this. Note that this error only affects absolute amounts of Cs eluted; any trends seen in the Cs-elution tests would still be valid. In subsequent testing, the F-factor was determined before running each experiment.

3.1.2 Cesium-Elution Experiments: General Procedure

The goal of the Cs-elution experiments was to determine the amount of Cs that could be eluted from the zeolite using an 8 wt% oxalic acid solution. The experiments were performed in 500 mL Pyrex® reaction vessels, with ports for thermocouples, stir shafts, and sampling, as shown in Figure 3.1. Cesium-loaded zeolite was placed in the bottom of the reaction vessel and covered with NaOH at pH ~11 (to simulate the basic environment in tank 8D-1). The 8 wt% oxalic acid was then added. The temperature of the added acid was either 25°C (room temperature), or 80°C. (The reason for testing heated acid is that at 25°C, 8 wt% is near the solubility limit for oxalic acid; oxalic acid is much more soluble at 80°C, and so adding heated acid to the tanks was thought to be an option.) The vessel was sealed using vacuum grease and placed in a heating mantle with a temperature controller programmed to hold the temperature at 50°C (the expected temperature of the material in tank 8D-1(a)). A stirring shaft was placed in each vessel and the contents of the vessel were agitated throughout the test period to simulate the effects of mixing pumps in tank 8D-1. Tests were run for 50 hours unless otherwise noted, and sampling was performed at 2, 4, 20, 28, and 50 hours of contact time, unless otherwise noted. When sampling, agitation of the material in the vessel was stopped and the material was allowed to settle for five minutes. After five minutes, a 5 mL sample was drawn from the supernatant and analyzed by ICP-MS for Cs content. A discussion of sources of experimental error arising from these procedures can be found in Appendix B. A listing of all of the Cs-elution tests performed can be found in Appendix C.

3.1.3 Determination of Optimum Process Conditions

This first experiment involved six tests, and was designed to determine the optimum process conditions for Cs elution from zeolite. The variables in this experiment were the ratio of oxalic acid to zeolite (20, 40, and 80 L acid/kg zeolite), the temperature of the acid (25°C or 80°C), and the contact time (2, 4, 20, 28, or 50 hours). The various experimental conditions are tabulated in Table 3.2.

(a) Fahey, SC, fax to KD Wiemers dated October 20, 1994.

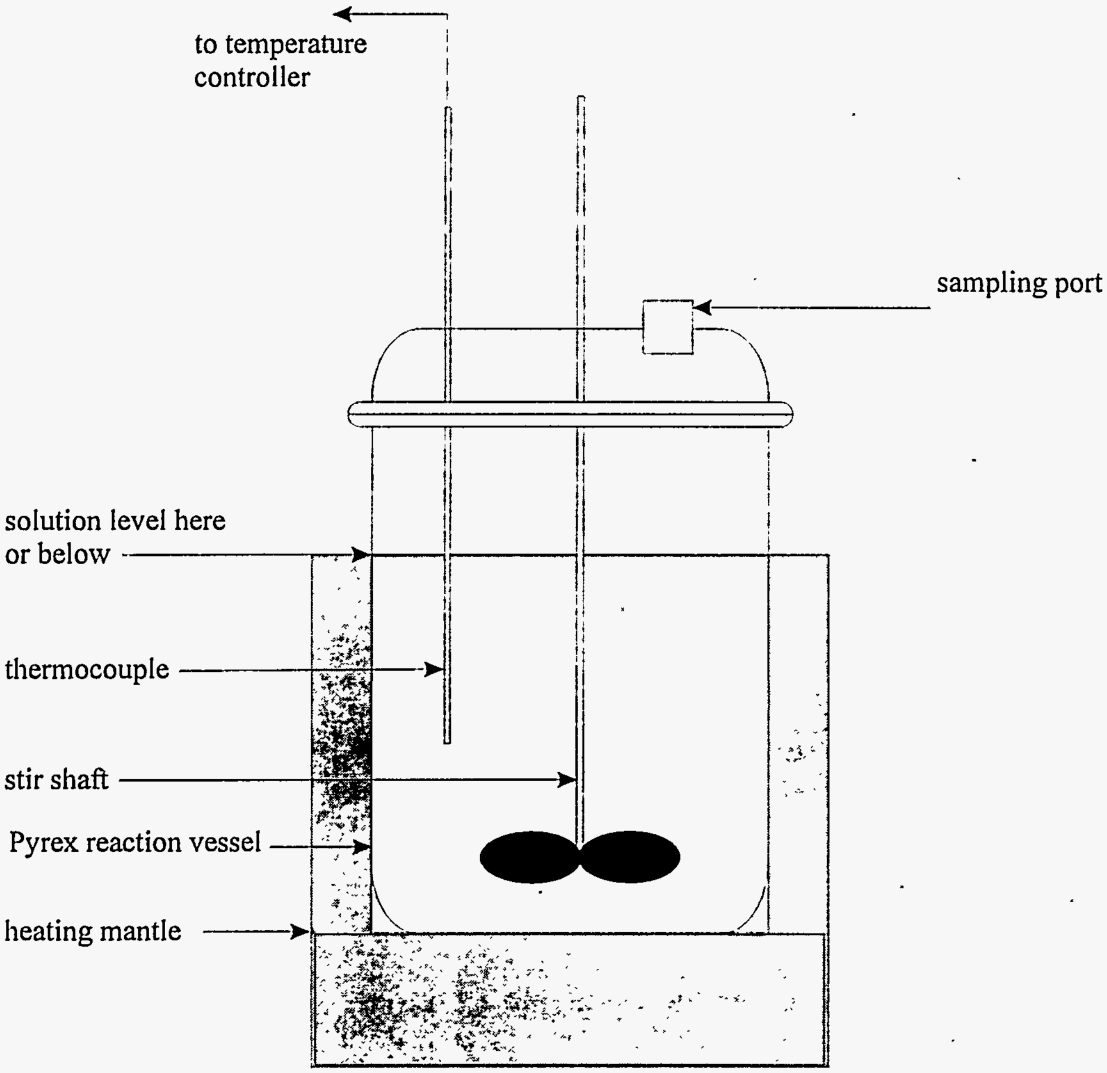

Figure 3.1. Five-Hundred mL Reaction Vessel Assembly Used in Cs-Elution and Sludge-Dissolution Tests

Table 3.2. Experimental Conditions for Determination of Optimum Process Conditions for Cs Elution from Zeolite

	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Oxalic acid/zeolite ratio (L/kg)	20	40	80	20	40	80
Temperature oxalic acid (°C)	25	25	25	80	80	80
Amount Cs loaded zeolite (g)	5.93	5.93	5.93	5.93	5.93	5.93
Amount NaOH (pH ~ 11) (g)	2.77	2.77	2.77	2.77	2.77	2.77
Amount 8 wt% oxalic acid solution (g)	114.6	229.3	458.6	114.6	229.3	458.6
Bath temperature (°C)	50	50	50	50	50	50
Agitation?	yes	yes	yes	yes	yes	yes

3.1.4 Repeatability Experiment

The repeatability experiment involved four tests; it was designed to determine the repeatability of the Cs-elution results obtained in the first experiment by running four identical tests, using the optimum process conditions determined under the first experiment. The optimum process conditions were determined to be 40 L acid/kg zeolite added at 25°C, with a two-hour contact time. It was determined, however, that one set of data points at two hours contact was not enough to make an accurate determination of repeatability, and so these tests were run for 50 hours, with sampling at 2, 4, 20, 28, and 50 hours contact time. The experimental conditions used in the repeatability tests are shown in Table 3.3.

3.1.5 Long-Term Experiment

The third Cs-elution experiment involved two tests, and was designed to determine the effect of long contact times (up to 11 days) on Cs elution from zeolite. The first test, Test 11, used the optimum process conditions of 40 L acid/kg zeolite added at 25°C and was held at 50°C for 11 days; the second test, Test 12, was an attempt to reproduce data obtained in previous PNNL

Table 3.3. Experimental Conditions for Cs-Elution Repeatability Tests

	Test 7	Test 8	Test 9	Test 10
Oxalic acid/zeolite ratio (L/kg)	40	40	40	40
Temperature oxalic acid (°C)	25	25	25	25
Amount Cs loaded zeolite (g)	5.93	5.93	5.93	5.93
Amount NaOH (pH ~ 11) (g)	2.77	2.77	2.77	2.77
Amount 8 wt% oxalic acid solution (g)	229.3	229.3	229.3	229.3
Bath temperature (°C)	50	50	50	50
Agitation?	yes	yes	yes	yes

Table 3.4. Experimental Conditions for Long-Term Cs-Elution Tests

	Test 11	Test 12
Oxalic acid/zeolite ratio (L/kg)	40	20
Temperature oxalic acid (°C)	25	25
Amount Cs loaded zeolite (g)	5.93	5.93
Amount NaOH (pH ~ 11) (g)	2.77	2.77
Amount 8 wt% oxalic acid solution (g)	229.3	229.3
Bath temperature (°C)	50	70
Agitation?	yes	yes

experimental work.[(a)] It used 20 L acid/kg zeolite added at 25°C and was held at 70°C for 11 days. The experimental conditions for the long-term tests are summarized in Table 3.4. Samples (5 mL) were taken at contact times of 2 hrs, 4 hrs, 1 day, 4 days, 7 days, and 11 days.

3.1.6. Multiple-Contact Experiment

The fourth Cs-elution experiment involved four tests, and was designed to determine the effect of multiple contacts with oxalic acid on the elution of Cs from zeolite. Using the optimum process conditions determined in the first experiment, Cs-loaded zeolite was contacted with fresh oxalic acid three times, for two hours each. Two-hour contact times were chosen because the first experiment indicated that the majority of the Cs was eluted after two hours. After each contact, a 5 mL sample of supernatant was drawn from each reaction vessel for analysis, and the rest of the supernatant was removed from the vessel. Two types of tests were performed,

Table 3.5. Experimental Conditions for Multiple-Contact Cs-Elution Tests

	Test 13	Test 14	Test 15	Test 16
Oxalic acid/zeolite ratio (L/kg)	40	40	40	40
Temperature oxalic acid (°C)	25	25	25	25
Amount Cs loaded zeolite (g)	7.77	7.76	7.77	7.77
Amount NaOH (pH ~ 11) (g)	2.77	2.77	2.78	2.77
Bath Temperature (°C)	50	50	50	50
Amount 8 wt% oxalic acid solution--1st contact	229.3	229.3	229.3	229.3
Amount first water rinse	*	7.76	*	7.78
Amount 8 wt% oxalic acid solution--2nd contact	229.3	229.3	229.3	229.3
Amount second water rinse	*	7.77	*	7.78
Amount 8 wt% oxalic acid solution--3rd contact	229.3	229.3	229.3	229.3
Agitation?	yes	yes	yes	yes

* water rinse not used in these tests

(a) Bray, LA, 1986. *Washed Sludge and Cesium/Zeolite Dissolution Studies*. WVST 86/104, Pacific Northwest National Laboratory, Richland, Washington.

one involving a water rinse (1:1 deionized water : zeolite) of the zeolite after each contact, and one with no water rinse. The experimental conditions used for the multiple-contact tests are summarized in Table 3.5.

3.1.7. Effects of Condition Modifications

The fifth Cs-elution experiment involved five tests and was designed to determine the effects on Cs elution from zeolite of several modifications of the optimum process conditions. The following modifications were made: presence of corrosion products, presence of carbon-steel coupons, 25°C bath temperature, and lack of agitation. The experimental conditions for this series of tests are shown in Table 3.6.

The effect of corrosion products in tank 8D-1 on Cs elution was examined in Test 17. Corrosion products were simulated by adding Fe_2O_3 to the vessel at a level equivalent to 2400 kg of rust at the bottom of the tank.[a]

The effect of the presence of carbon-steel coupons on the elution of Cs from zeolite was examined in Test 18. Two carbon-steel coupons were pre-weighed, suspended inside the reaction vessel, and completely submerged in the oxalic acid. The coupons were prepared from archived tank 8D-1 carbon steel which had been part of earlier WVNS tank farm support corrosion testing. The coupons were coated with a layer of corrosion product ("rust"). The depth of the oxide layer varied among the coupons and was not characterized before testing. The use of archived 8D-1 steel in the "as received" condition was thought to be of a significant benefit to the tests by minimizing uncertainties surrounding the use of "clean" specimens. A pair

Table 3.6. Experimental Conditions for Cs-Elution Tests Involving Condition Modifications

	Test 17 Fe_2O_3	Test 18 Coupons	Test 19 No stir	Test 20 25°C Bath	Test 21 Control
Oxalic acid/zeolite ratio (L/kg)	40	40	40	40	40
Temperature oxalic acid (°C)	25	25	25	25	25
Amount Cs loaded zeolite (g)	7.76	7.76	7.76	7.76	7.76
Amount NaOH (pH ~ 11) (g)	2.76	2.76	2.76	2.76	2.76
Amount 8 wt% oxalic acid solution (g)	229.3	229.3	229.3	229.3	229.3
Bath temperature (°C)	50	50	50	25	50
Amount Fe_2O_3	4.13	*	*	*	*
Steel Coupons?	no	yes	no	no	no
Agitation?	yes	yes	no	yes	yes

* Fe_2O_3 not added

(a) Schiffhauer, MA, fax to KD Wiemers dated 27 September, 1994.

Table 3.7. Initial Carbon-Steel Coupon Data for Cs-Elution Test 18

	Cleaned Coupon	Rusted Coupon
Coupon ID	X056C	X056R
Length (cm)	2.9477	3.0544
Width (cm)	1.0439	1.0422
Thickness (cm)	0.2548	0.2573
Weight (g)	5.3276	5.5399

of coupons for this test was prepared from larger U-bend coupons from the prior tests. One coupon of the pair was measured, weighed, and otherwise left "as received" with a rusted layer on the surface. The other coupon was cleaned in an inhibited acid cleaning solution to remove the rust layer. Once cleaned, the "clean" coupons were measured and weighed. The initial dimensions and weights of the coupons are shown in Table 3.7. When the test was complete, the coupons were removed from the vessel, rinsed in deionized water, and dried at room temperature. The coupons were then re-weighed to determine weight loss, from which corrosion rates were estimated.

The effect of no agitation on Cs elution was examined in Test 19. The vessel for this test was set up without a stir shaft. The effect of a 25°C bath temperature over 50 hours, rather than the 50°C bath temperature, was examined in Test 20. The 25°C bath temperature reflects conditions under which the tank contents are at ambient temperature, while the 50°C bath temperature reflects the effects of both radiolytic decay heat and heat from the pumps used to agitate the tank contents. Test 21 was a control run at optimum process conditions. In addition to ICP-MS analysis for Cs concentration, the samples in this set of tests were also analyzed by ICP-MS for Fe and Al, the oxides of which are major components of the zeolite.

3.1.8 Effect of Varying Oxalic Acid Concentration on Cs Elution and on the Amount of Fe Dissolved

The sixth Cs-elution experiment was designed and run after the results from the first five experiments (i.e., sets of tests) were analyzed. The results of Tests 17 (Fe_2O_3) and 18 (steel coupons) raised some concern about whether the amounts of iron dissolved by the Cs-elution process would be low enough to be acceptable for vitrification activities. The objective of this experiment was to refine the optimum process conditions for Cs elution based on engineering information received from WVNS.

Three major engineering constraints were accommodated (see Appendix A for pertinent calculations):

(1) A three-foot (0.91 meter) minimum level in Tank 8D-1 is required for agitation. Assuming 5 wt% of the zeolite remains in the tank, this translates to ~100 L acid/kg zeolite.

(2) A maximum pumping rate of 100 gallons per minute (0.455 L/minute) has been identified. At this rate, it will take ~15 hours to pump in enough oxalic acid to reach the minimum three-

foot (0.91 meter) depth. It will take this long to remove the same amount of oxalic acid from the tank. Therefore, a minimum contact time of 30 hours is required.

(3) Minimization of the number of glass logs produced is an issue of vital importance. The 8 wt% oxalic acid used in the previous five experiments was shown to solubilize >90 wt% of the Fe present as corrosion products, which, based on WVNS recent assessment(a), may result in an unacceptable number of logs for this mission. Lower concentrations of oxalic acid (4 wt% and 1 wt%) were therefore used in this experiment to reduce Fe solubilization.

Test 22 served as a control for the 4 wt% oxalic acid tests; Test 26 served as a control for the 1 wt% oxalic acid tests. The effect of corrosion products was examined in Tests 23 and 27 by adding Fe_2O_3; Test 23 examined its effect in 4 wt% oxalic acid, and Test 27 in 1 wt% oxalic acid. The amount of Fe_2O_3 was adjusted based on engineering information received from WVNS(a); the calculation can be found in Appendix A. Tests 24 and 28 examined the effects of the presence of carbon-steel coupons at 4 and 1 wt% oxalic acid, respectively. Tests 25 and 29 examined the effects of carbon-steel coupons, in the absence of zeolite, on the amount of Fe in solution, at 4 and 1 wt% oxalic acid, respectively. The initial weights of the coupons used in Tests 24, 25, 28, and 29 are shown in Table 3.8. In these tests, the coupons were of such a size that the same relative amount of tank surface was exposed to the oxalic acid solution as would be exposed if Tank 8D-1 were filled to a three foot depth with the oxalic acid solution. The calculations of the required coupon size can be found in Appendix A.

Table 3.8. Initial Carbon-Steel Coupon Data for Cs-Elution Tests 24-29

Test	Coupon ID	Initial wt (g)
Test 24	599	13.2214
Test 25	449	13.5338
Test 28	450	12.9148
Test 29	593	13.4012

Previous tests (Tests 13-16) showed that three two-hour contacts eluted 99.9 wt% of the Cs present. This experiment was run using three 47 hour contacts during which the reaction vessels were maintained at 50°C and stirred continuously. Sampling occurred at 2, 4, 20, 28, and 47 hours from the beginning of each contact. After each contact, supernatant was removed from each vessel and replaced with fresh oxalic acid solution. Table 3.9 summarizes the conditions used for these tests.

(a) Phone conference with CS King and MA Schiffhauer, April 20, 1995.

Table 3.9. Experimental Conditions for Tests with Varying Oxalic Acid Concentrations

	Test 22 Control	Test 23 Fe_2O_3	Test 24 Coupons	Test 25 Coupons	Test 26 Control	Test 27 Fe_2O_3	Test 28 Coupons	Test 29 Coupons
Oxalic acid/zeolite ratio (L/kg)	100	100	100	100	100	100	100	100
Oxalic acid concentration	4 wt%	4 wt%	4 wt%	4 wt%	1 wt%	1 wt%	1 wt%	1 wt%
Temperature oxalic acid (°C)	25	25	25	25	25	25	25	25
Amount Cs-loaded zeolite (g)	5.40	5.40	5.40	0	5.40	5.40	5.40	0
Amount NaOH (pH ~ 11) (g)	2.46	2.47	2.47	0	2.47	2.47	2.47	0
Amount oxalic acid (g)	508.7	508.7	508.7	508.7	501.5	501.5	501.5	501.5
Bath temperature (°C)	50	50	50	50	50	50	50	50
Amount Fe_2O_3	*	5.90	*	*	*	5.90	*	*
Steel coupons?	n	n	y	y	n	n	y	y
Number of contacts	3	3	3	3	3	3	3	3
Time per contact, hours	47	47	47	47	47	47	47	47

* Fe_2O_3 not added

3.2 Results and Discussion

3.2.1 Determination of Optimum Process Conditions

The first experiment involved six tests and was designed to determine the optimum process conditions for elution of Cs from zeolite. The variables examined were the amount of acid (20, 40, or 80 litres of 8 wt% oxalic acid per kg of Cs-loaded zeolite); the initial temperature of the added oxalic acid (25°C or 80°C); and the total contact time (2, 4, 20, 28, or 50 hours).

The results of this set of tests are summarized in Table 3.10 and Figure 3.2. Temperature and pH data collected during the experiment are found in Appendix D; sample calculations showing the method by which raw data were converted to wt% Cs eluted are found in Appendix E. The data were corrected for sample removal; this correction is shown in the calculations in Appendix E. As Figure 3.2 shows, no one set of conditions consistently produced the highest elution level at each contact time. Cesium elution ranged from a minimum of 52 wt% (at 40 L acid/kg zeolite, 25°C acid addition, 2 hour contact time) to 90 wt% (at 20 L acid/kg zeolite, 80°C acid addition, 50 hour contact time). Tests 2, 4, and 6 showed consistently high elution at longer contact times. In the interest of keeping plant operations simple (i.e., heating the oxalic acid to 80°C would add difficulty to the process and may not significantly improve the Cs elution) and minimizing waste, 40 L acid/kg zeolite added at 25°C was chosen as the optimum process condition. The majority of the Cs elution appears to occur during the first two hours of contact, with ~60 wt% of the Cs eluted in this time, and so a two-hour contact time was chosen as optimum for the laboratory-scale tests. A contact time this short may not be feasible in full-scale operations.

3.2.2 Repeatability Experiment

The second experiment addressed the issue of repeatability. Four identical tests were run, using the optimum process conditions of 40 L 8 wt% oxalic acid/kg zeolite added at 25°C.

Table 3.10. Percent Cs Eluted from Zeolite in Experiment to Determine Optimum Process Conditions*(a)

Hours	Test 1 20 L/kg, 25°	Test 2 40 L/kg, 25°	Test 3 80 L/kg, 25°	Test 4 20 L/kg, 80°	Test 5 40 L/kg, 80°	Test 6 80 L/kg, 80°
2	60	52	62	63	57	58
4	58	53	59	65	62	62
20	69	70	65	72	65	63
28	66	66	64	73	73	60
50	70	80	66	90	64	72

* All values in table have a ±10% uncertainty due to analytical error

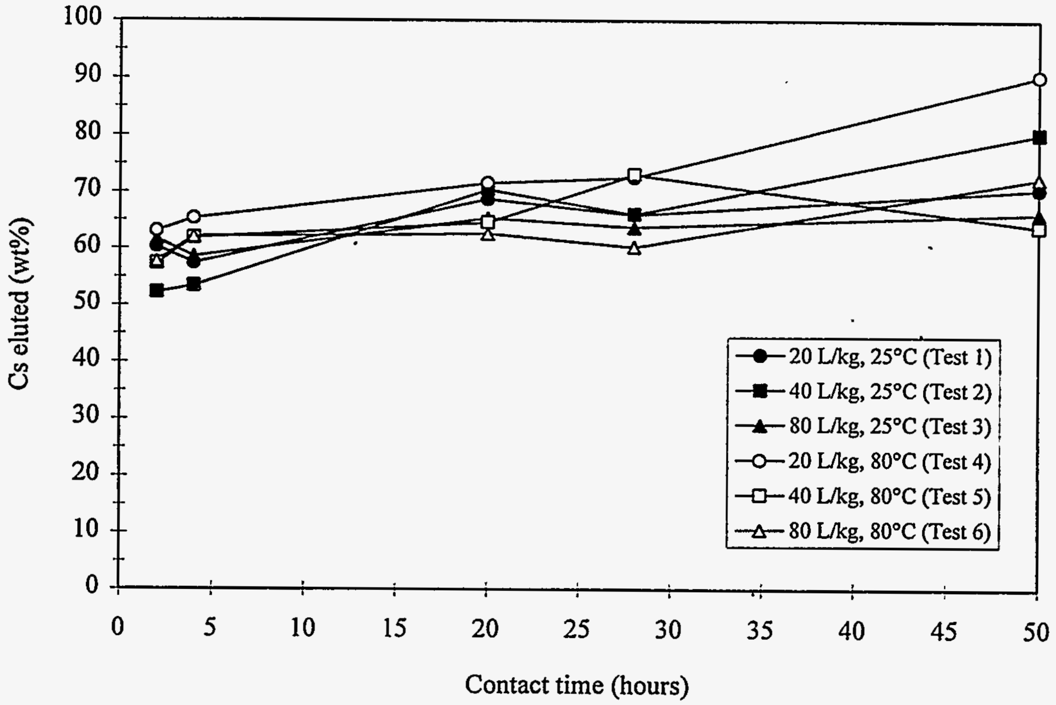

Figure 3.2. Percent Cs Eluted from Zeolite in Experiment to Determine Optimum Process Conditions (100% = Total Amount of Cs Loaded onto Zeolite)

The results of this set of tests are summarized in Table 3.11 and Figure 3.3. Temperature and pH data collected during the experiment are shown in Appendix D; sample calculations in which raw data are converted to wt% Cs eluted are found in Appendix E. The data were corrected for sample removal, and the calculations in Appendix E reflect this correction. Cesium elution

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(a) Because of the absorption of moisture by the zeolite during the course of the first three experiments, the percentages reported in Table 3.10 may be low. This affects the results of Tests 1-12. See section 3.1.1 for details.

Table 3.11. Percent Cs Eluted from Zeolite in Repeatability Tests Run Using 40 L Acid/kg Zeolite at 50°C[(a)] *

Hours	Test 7	Test 8	Test 9	Test 10
2	64	55	60	62
4	64	56	60	61
20	75	57	60	64
28	70	65	62	65
50	68	54	64	71

* All values in the table have a ±10% uncertainty because of analytical error

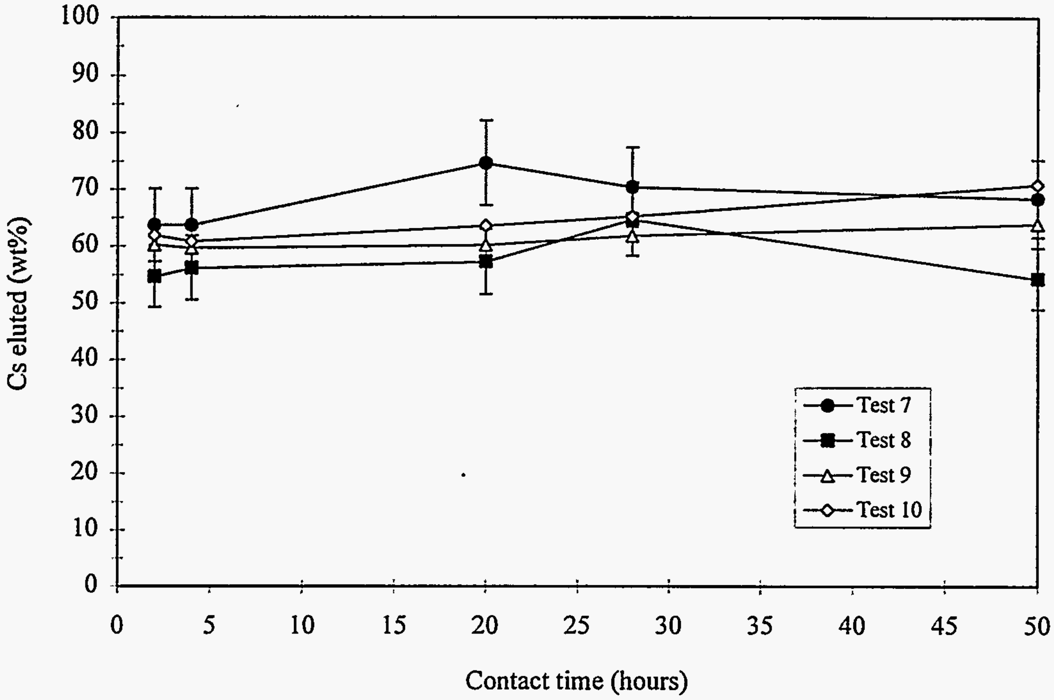

Figure 3.3. Percent Cs Eluted from Zeolite in Repeatability Tests (100% = Total Amount of Cs Loaded onto Zeolite)

(a) Because of the absorption of moisture by the zeolite during the course of the first three experiments, the percentages reported in Table 3.11 may be low. This affects the results of Tests 1-12. See section 3.1.1 for details.

ranged from a minimum of 54 wt% at 50 hours contact time, to a maximum of 75 wt% at 20 hours contact time. These results fall into the same range as those from the first experiment, in which optimum process conditions were determined. Error bars for two of the data sets are shown in Figure 3.3, reflecting the ±10% analytical error in the ICP-MS measurement of Cs concentration in the samples. This is considered to be the minimum error associated with this data (see Appendix B). Although there is some scatter in the data, for the most part, the data points seem to fall within the bounds of experimental error.

3.2.3 Long-Term Experiment

The third experiment examined the effects of long-term exposure to oxalic acid on Cs elution using two test conditions. The first test used 40 L 8 wt% oxalic acid/kg zeolite added at 25°C, a vessel temperature of 50°C, and an 11-day contact time. The second test attempted to reproduce data obtained in previous studies[(a)], and used 20 L acid/kg zeolite added at 25°C, a vessel temperature of 70 °C, and an 11-day contact time. The data indicated that Cs elutions as high as 88-90 wt% might be obtained under these conditions.

The results of this experiment are shown in Table 3.12 and Figure 3.4. Temperature and pH data collected during the experiment are found in Appendix D; sample calculations showing the method by which the raw data were converted to wt% Cs eluted are shown in Appendix E. The data were corrected for sample removal; this correction is reflected in the calculations shown in Appendix E. As Figure 3.4 shows, both tests attained high levels of Cs elution, with the 70°C bath temperature showing higher levels at all contact times. In the 50°C test, the maximum amount of Cs eluted was 96 wt% after 96 hours (4 days) contact time; in the 70°C test, the maximum amount of Cs eluted was 130 wt% after 264 hours (11 days) contact time. Elutions greater than 100 wt% are most likely due to an error in the amount of moisture in the zeolite (see section 3.1.1).

Table 3.12. Percent Cs Eluted from Zeolite in Long-Term Tests[(b)] *

Hours	Test 11 50°C Bath	Test 12 70°C Bath
2	75	92
4	82	100
24	87	130
96	96	110
168	80	130
264	88	130

* All values in table have a ±10% uncertainty because of analytical error

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(a) Bray, LA 1986. *Washed Sludge and Cesium/Zeolite Dissolution Studies.* WVST 86/104. Pacific Northwest Laboratory, Richland, Washington 99352.

(b) Because of the absorption of moisture by the zeolite during the course of the first three experiments, the percentages reported in Table 3.12 may be low. This affects the results of Tests 1-12. See section 3.1.1 for details.

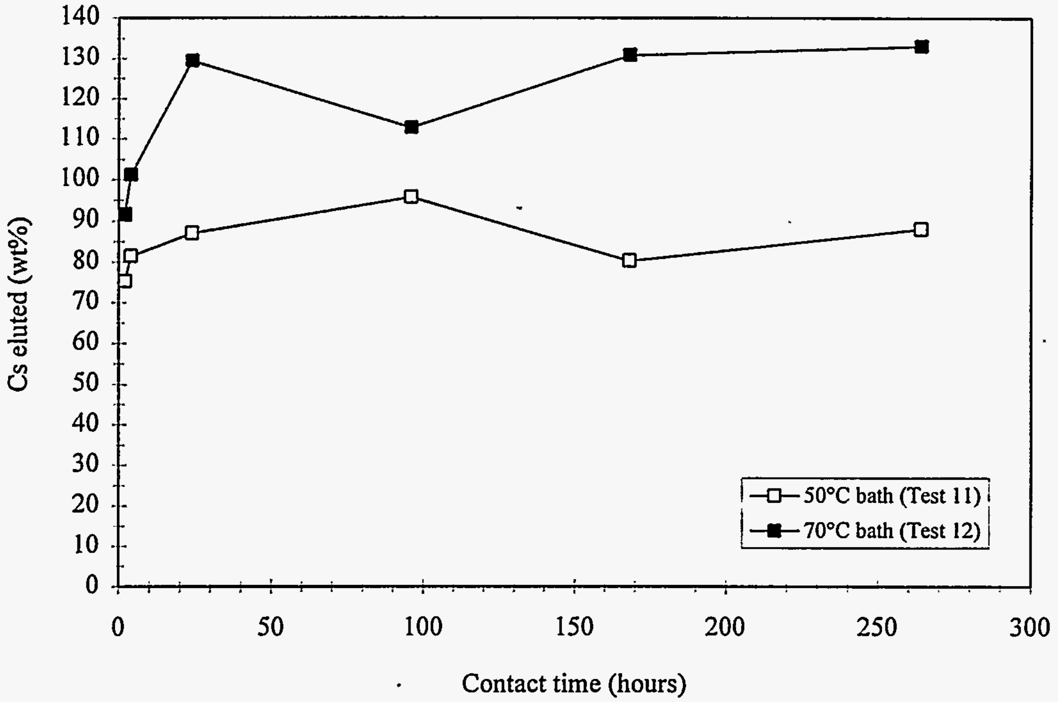

Figure 3.4. Percent Cs Eluted from Zeolite in Long-Term Tests (100% = Total Amount of Cs Loaded onto Zeolite)

The data indicate that in both cases either the maximum or a value quite close to it was observed after 96 hours contact time. Longer contact times do not improve or impede Cs elution. Near-maximum dissolution is achieved after 24 hours contact for the 70°C bath. In the sense that high percentages of Cs can be eluted from zeolite, these tests are in agreement with those done previously[(a)]; however, these tests show that 11 days of contact is not necessary to achieve the maximum results. Although long-term contacts are probably feasible in a full-scale operation, it is improbable that a system could be engineered to maintain the tank contents at 70°C.

3.2.4 Multiple-Contact Tests

The fourth experiment examined the effects of multiple contacts with oxalic acid on Cs elution. Cesium-loaded zeolite was contacted with fresh oxalic acid (at 40 L 8 wt% oxalic acid/kg zeolite, 25°C acid addition, 2 hour contact time) three times. Two-hour contacts were used because >50 wt% of the Cs was eluted after two hours contact time in the first experiment. Two types of tests were performed, one involving a water rinse after each contact, and one with no water rinse.

The results of these tests are shown in Table 3.13 and Figure 3.5. Temperature and pH data collected during the experiment are found in Appendix D; sample calculations showing the method by which the raw data were converted to wt% Cs eluted are shown in Appendix E. The data were corrected for sample removal, which is reflected in the calculations shown in Appendix E. As shown in Figure 3.5, close to 100 wt% Cs elution was observed using two and

Table 3.13. Percent Cs Eluted from Zeolite in Multiple-Contact Tests*

Contacts	Test 13 No Rinse	Test 14 Rinse	Test 15 No Rinse	Test 16 Rinse
1	88	90	85	90
2	100	100	110	110
3	110	100	110	110

* All values in the table have a ±10% uncertainty because of analytical error

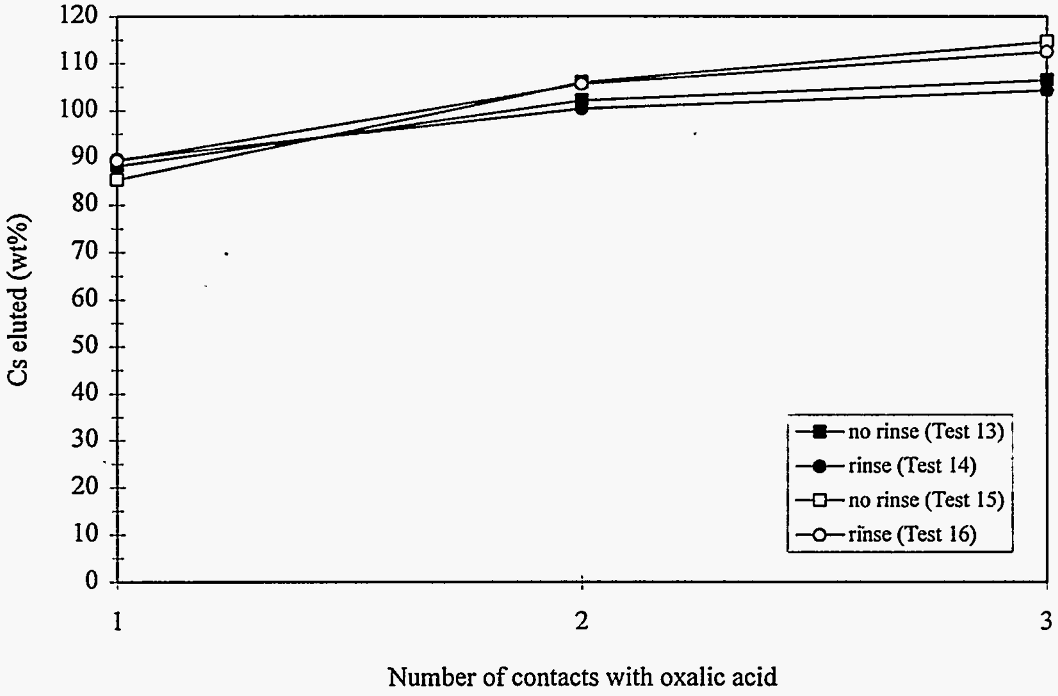

Figure 3.5. Percent Cs Eluted from Zeolite in Multiple-Contact Tests (100% = Total Amount of Cs Loaded onto Zeolite)

three consecutive two-hour contacts with oxalic acid. A water rinse (1:1 deionized water : initial zeolite) did not appear to improve or hamper Cs elution significantly. Cesium elution increased from an average of 88 wt% after one contact to 100 wt% after two contacts, up to an average of 110 wt% after three contacts.

3.2.5 Effects of Condition Modifications

The fifth experiment examined the effects on Cs elution of several modifications of the optimum process conditions (40 L 8 wt% oxalic acid/kg zeolite added at 25°C). The additional conditions were: addition of Fe_2O_3 to simulate corrosion products (rust) in the tank; addition of carbon steel coupons (one pre-corroded, one clean) to simulate the sides of the tank and to obtain preliminary carbon-steel corrosion data; no agitation; and a 25°C bath temperature instead of 50°C.

In addition to measuring Cs concentrations in all of the supernatant samples, the concentrations of Fe and Al were measured. Aside from SiO_2, which constitutes 60.1 wt% of the zeolite, the other major components of the zeolite are Al_2O_3 (15.5 wt%), Na_2O (7.0 wt%), and Fe_2O_3 (3.5 wt%)[(a)]. Concentrations of Na were not determined because the amount of Na present from NaOH would dominate the results, making any trends in the data difficult to identify. Silicon concentrations were not measured because of high instrument background interference, possibly resulting from internal contamination of the instrument.

The results of this experiment are shown in Table 3.14 and Figure 3.6 through Figure 3.8. Temperature and pH data collected during the experiment are found in Appendix D; sample calculations showing the method by which the raw data were converted to wt% Cs eluted are shown in Appendix E. The data were corrected for sample removal, and this is reflected in the calculations shown in Appendix E. Tables showing the data from the Fe and Al measurements (which are plotted in Figures 3.7 and 3.8, respectively) are contained in Appendix F. As shown in Figure 3.6, after 50 hours contact time, all conditions except the 25°C bath resulted in approximately the same amount of Cs eluted, an average of 91 wt%, with a standard deviation of 2.5 wt%. As would be expected, the 25°C bath condition resulted in lower levels of Cs elution at longer contact times; however, the difference is within experimental error. The no-stir condition showed relatively low levels of Cs eluted at short contact times, but by 20 hours contact time, the Cs-elution level had risen from 29 wt% to 78 wt%, and by 50 hours, it was at 88 wt%. The reason for the delayed Cs elution in this test is not known. Neither the addition of Fe_2O_3 nor the presence of carbon-steel coupons appeared to have any effect on Cs elution at contact times greater than 2 hours.

Addition of Fe_2O_3

The addition of Fe_2O_3 to simulate corrosion products in the tank did not appear to have a significant effect on the elution of Cs from zeolite (Figure 3.6, Table 3.14). This is not surprising, as there is so much oxalate in the system that a competition between the mechanisms of Cs elution and Fe dissolution is not necessary (see calculations in Appendix A). It is believed that as the amount of oxalate in the system decreases, Cs-elution efficiency would also decrease; further testing may be needed to determine how low the oxalate concentration can go without compromising Cs elution. Addition of Fe_2O_3 does not appear to have an effect on the dissolution of Al in zeolite (Figure 3.8 and Appendix F). However, it does have an effect on the amount of Fe present in solution. In Figure 3.7, 100% Fe dissolved means 100% of the Fe in the system (i.e., Fe in the zeolite and Fe in the added Fe_2O_3). Hence, although Test 17 and Test 21 (the control) both appear to have dissolved the same percentage of Fe, the total amount of Fe in solution is much higher for Test 17. The data indicate that 92 wt% of the Fe in the system—including loose rust at the bottom of the tank—is in solution after 50 hours contact time. This additional Fe may increase the number of glass logs produced by vitrification. It is interesting to note that it appears to take some time for the Fe concentration in solution to

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(a) Olson, KM, ML Elliott, JW Shade, and HD Smith. 1990. *Fabrication, Characterization, and Evaluation of West Valley Sludge Glass-1 (WVSG-1), a Fully Radioactive Glass Made with High Level Waste From the West Valley Demonstration Project.* Letter report, Pacific Northwest National Laboratory, Richland, WA 99352.

Table 3.14. Percent Cs Eluted from Zeolite in Tests Involving Condition Modifications*

Hours	Test 17 Fe_2O_3	Test 18 Coupons	Test 19 No stir	Test 20 25°C Bath	Test 21 Control
2	68	61	15	51	81
4	82	83	29	74	89
20	100	98	78	73	90
28	91	92	84	82	94
50	94	89	88	80	91

* All data have a ±10% uncertainty due to analytical error

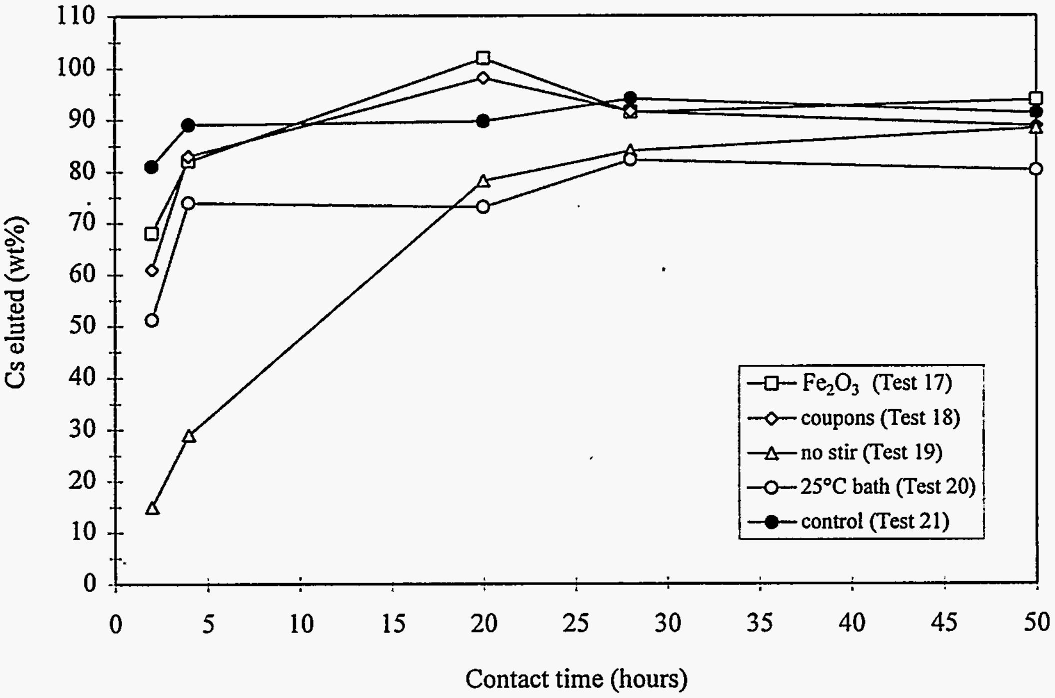

Figure 3.6. Percent Cs Eluted from Zeolite in Tests Involving Condition Modifications (100% = Total Amount of Cs Loaded onto Zeolite)

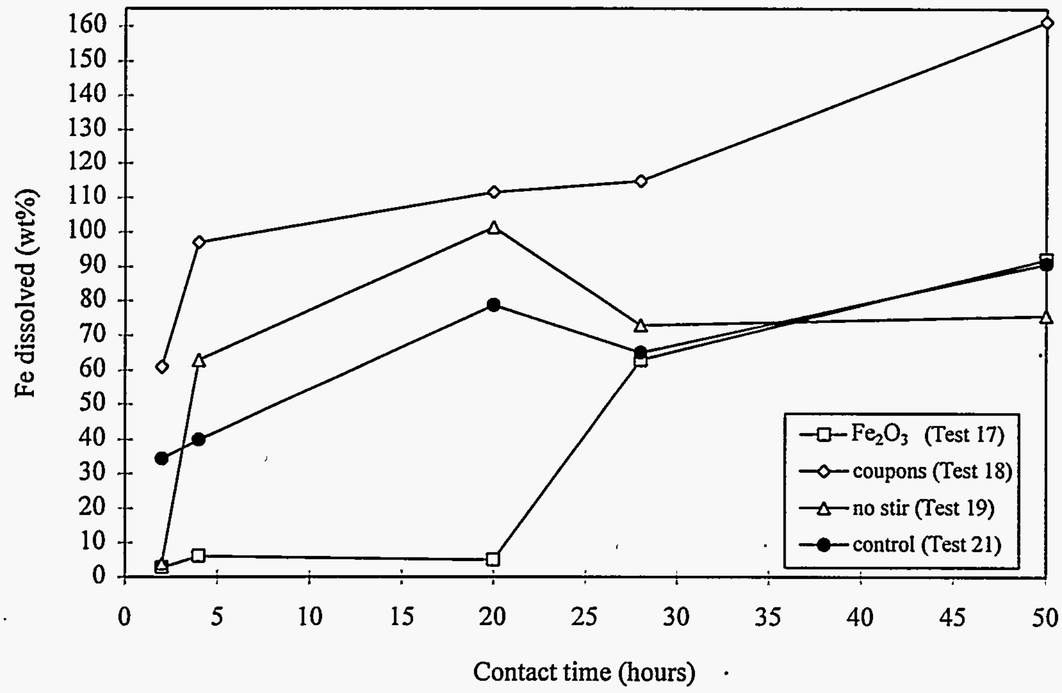

Figure 3.7. Weight Percent Fe Dissolved in Tests Involving Condition Modifications

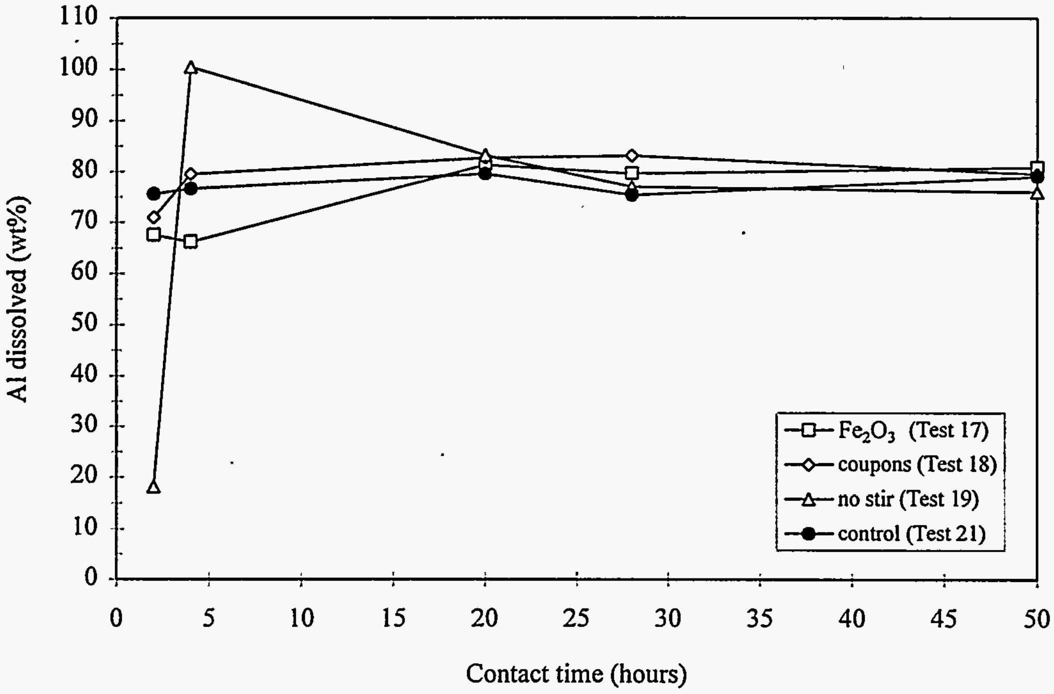

Figure 3.8. Weight Percent Al Dissolved in Tests Involving Condition Modifications

rise. At 20 hours contact time, only about 5 wt% of the Fe in the system is in solution (this is the amount one would expect if only the Fe in the zeolite were in solution.) By 28 hours, this amount has risen to 63 wt%.

Carbon-Steel Coupons

The presence of carbon steel coupons in the oxalic acid solution had no significant effect on either the elution of Cs from zeolite or on the dissolution of Al_2O_3 in zeolite, as shown in Figures 3.6 and 3.8. Amounts of Fe greater than 100 wt% in solution at longer contact times are shown in Figure 3.7. In this test, 100% Fe is based on the amount of Fe in the zeolite; therefore, amounts greater than 100% would have to be attributed to Fe dissolved from the carbon-steel coupons. The control test indicates that at 50 hours contact time, about 91 wt% of the Fe from the zeolite is in solution; this suggests that amounts of Fe above about 91 wt% in the coupon test are probably coming from the dissolution of the coupons themselves.

The results of the coupon tests are summarized in Table 3.15. The reported corrosion rates are based on weight loss after cleaning in the inhibited acid cleaning solution, and assume that the weight loss, and therefore the corrosion rate, is uniform over the whole surface of each coupon. For these tests, this is a reasonable assumption because no localized corrosion (i.e. pitting) was observed on any of the specimens. The calculated corrosion rates resulting from these tests are higher than anticipated. The rates may be unrealistically high because of the short duration of the tests. Corrosion rates commonly decrease with time as corrosion products accumulate on the specimen and passivate the surface, thereby inhibiting further corrosion. Longer-term tests would be needed to determine the true long-term corrosion rate under these conditions more accurately. However, since the oxalic acid does remove the oxide coating of the carbon steel, the surface may not passivate under these conditions. If so, the corrosion rates determined from these short tests may be indicative of longer-term corrosion rates.

The results indicate that the weight losses, and therefore the corrosion rates, for the rusted coupons are higher than for the cleaned coupons. If true, the oxide coating is apparently dissolved at a higher rate than the base metal. Because of the limited number of coupons used in this testing, the significance of the differences in the corrosion rates cannot be determined. These results tend to suggest that the surface would not be passivated under these conditions, and that

Table 3.15. Corrosion Rates for Carbon-Steel Coupons in Cs-Elution Test 18

	Cleaned Coupon	Rusted Coupon
Coupon ID	X056C	X056R
Weight change, g	0.0938	0.1648
Surface area, cm^2	7.524	7.817
Corrosion rate, mpy*	109	185
Corrosion rate, um/yr	2770	4700

*mpy = mils (thousandths of an inch) per year the longer-term corrosion rates may well be high. Without further testing, one would expect that the corrosion rate for carbon steel under these conditions is between 100 and 200 mils per year (2500 and 5000 um/yr).

No Stirring

A lack of agitation in the reaction vessel contents only appeared to affect Cs elution at short contact times. At 2 and 4 hours, 15 wt% and 29 wt% of the Cs was eluted, respectively, but after 20 hours, the amount of Cs eluted increased to 78 wt%, and by 50 hours had reached the same level as the control, as shown in Figure 3.6. These results suggest that when contact times are long (i.e., greater than 28 hours), a lack of agitation should not be detrimental to Cs elution. It is important to note, however, that the small scale of these tests made it impossible to examine conditions in which the zeolite is in a pile and thus has less surface area exposed to the oxalic acid. Under such conditions, Cs elution may be limited by diffusion of oxalic acid into the zeolite pile, especially if agglomerates exist in the pile. The lack of agitation had a similar effect on the dissolution of Fe and Al in the zeolite.

25°C Bath

The 25°C bath temperature resulted in lower levels of Cs elution at most contact times, as shown in Figure 3.6. This suggests that the temperature of the system could have an impact on the efficiency of Cs-elution operations. If tank temperatures fall below the 50°C used in these tests, it may be necessary to extend the duration of contact times and/or implement multiple-contact treatments.

3.2.6. Effect of Varying Oxalic Acid Concentration on Cs Elution and on Amount of Fe Dissolved

The sixth experiment involved eight tests and was designed to refine the optimum process conditions for Cs elution based on the results of the first five experiments and on engineering information received from WVNS. Variables in the tests were adjusted to accommodate three major engineering constraints. The concentration of oxalic acid used was reduced from 8 wt% to 4 wt% and 1 wt%, with the objective of reducing the amount of Fe dissolved during contact with oxalic acid. The ratio of oxalic acid-to-zeolite was changed from 40 L acid/kg zeolite to 100 L acid/kg zeolite to maintain a minimum depth of three feet (0.91 meters) in the tank, which will allow for agitation of the tank contents. A minimum contact time of 30 hours was identified; these tests used 47 hours/contact. The original multiple-contact experiment (Tests 13-16) indicated that 99.9 wt% of the Cs could be eluted in three contacts, and so three contacts were again used in this set of tests.

The results of this set of tests are summarized in Figure 3.9 through Figure 3.15. The Cs-elution data and the Fe dissolution data are contained in Appendix G. Temperature and pH data collected during the experiment are contained in Appendix D; sample calculations showing the method by which raw data were converted to wt% Cs eluted and wt% Fe dissolved are shown in Appendix E. The data were corrected for sample removal, and this correction is shown in the calculations in Appendix E.

Cesium Elution at Various Oxalic Acid Concentrations

Tests 22 and 26 were controls, run with 4 wt% and 1 wt% oxalic acid, respectively. The results of these tests plotted with the results of Test 21, which was a control for tests using 8 wt% oxalic acid are shown in Figure 3.9. Only data for the first contact (47 hours) is shown in this plot; Test 21 was only run for 50 hours. It is important to note that Test 21 was also run using 40 L oxalic acid/kg zeolite, rather than the 100 L/kg used in Tests 22 and 26. The figure shows that even at a lower acid:zeolite ratio, 8 wt% oxalic acid is more effective for Cs elution than 4 or 1 wt% oxalic acid. At 8 wt% (Test 21), 91 wt% of the Cs was eluted, while at 4 wt% (Test 22), 72 wt% was eluted, and at 1 wt% (Test 26), 63 wt% of the Cs was eluted at the completion of the contact.

The results of Tests 22 (4 wt%) and 26 (8 wt%) over the entire experiment are shown in Figure 3.10. Also plotted are results from Test 13, which was a multiple-contact test run using 8 wt% oxalic acid at 40 L acid/kg zeolite and three two-hour contacts, as opposed to the three 47-hour contacts used in Tests 22 and 26. The figure shows that the 8 wt% oxalic acid elutes the most Cs in three contacts, even though the contacts used in Test 13 were only two hours long. Throughout most of the experiment, 4 wt% oxalic acid eluted more Cs than 1 wt% oxalic acid. The last two data points for Test 26 (1 wt%) indicate that the amount of Cs eluted suddenly increased dramatically at 122 hours total contact time. This is likely because of experimental error. If these two points are discounted, and the data for the final contact are examined, then it appears that 1 wt% and 4 wt% oxalic acid both elute ~75 wt% of the Cs present.

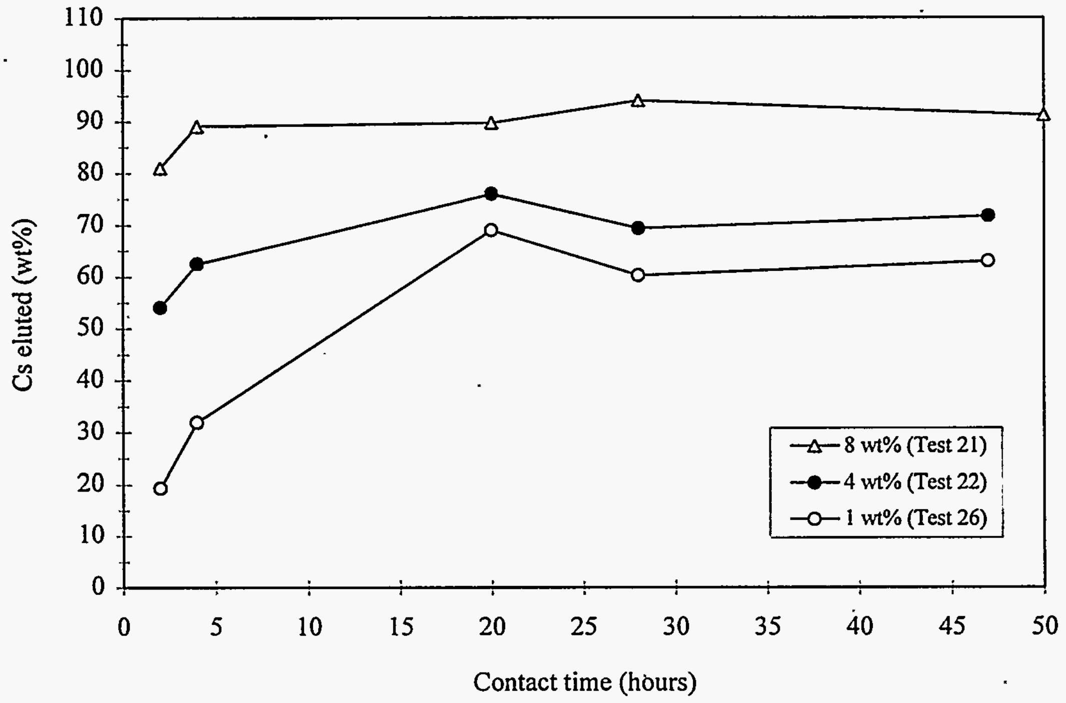

Figure 3.9. Percent Cs Eluted from Zeolite by 8, 4, and 1 wt% Oxalic Acid During One Contact

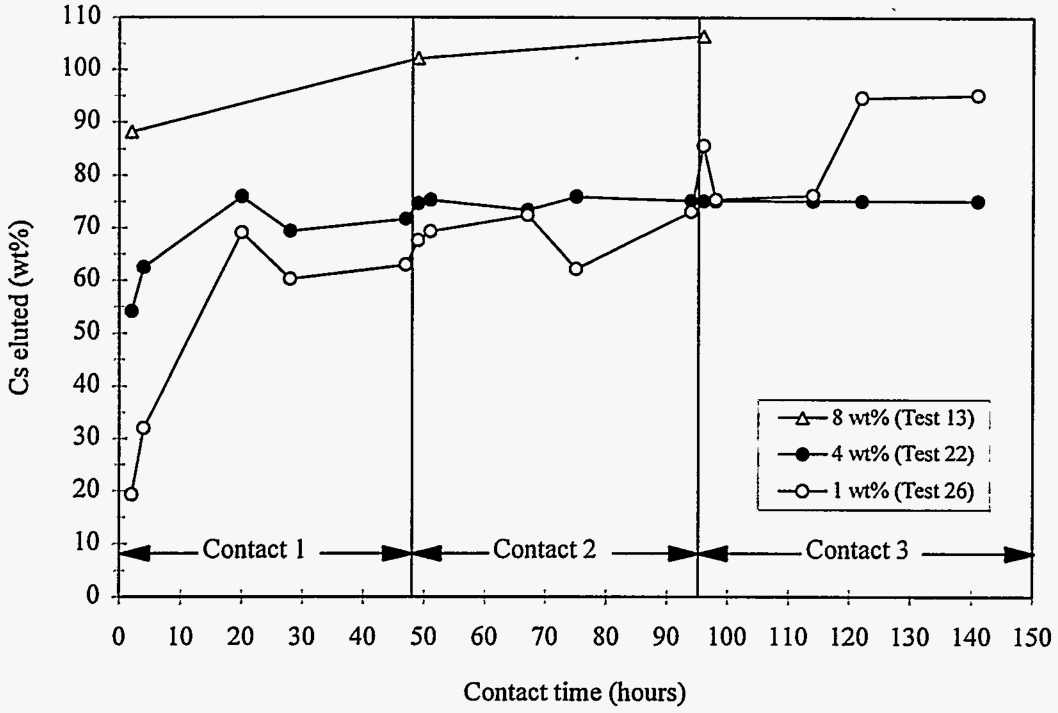

Figure 3.10. Percent Cs Eluted from Zeolite During Multiple-Contact Tests Using 8, 4, and 1 wt% Oxalic Acid and Three Contacts

Cesium Elution in the Presence of Fe

Tests 23, 24, 27, and 28 examined the effect of Fe on Cs elution. In Tests 23 and 27, Fe_2O_3 was added in amounts equivalent to the amount of loose iron, or "rust," expected to be on the bottom of the tank. The results of these tests are shown in Figure 3.11, along with the data from the controls. The results of Test 17, an 8 wt% oxalic acid test run in the presence of Fe_2O_3, are not shown because few of the process conditions were the same as in Tests 23 and 27, including the amount of Fe_2O_3 added to the system. As Figure 3.11 shows, 4 wt% oxalic acid was more effective at eluting Cs than was 1 wt% oxalic acid. After three 47-hour contacts, 4 wt% oxalic acid had eluted ~85 wt% of the Cs. If one discounts the last two data points in the 1 wt% oxalic acid test as experimental error, then it appears that 1 wt% oxalic acid eluted ~74 wt% of the Cs. The data show that although the Cs elution appears to start slower when Fe_2O_3 is present, the amount of Cs eluted by the middle of the third contact (at 114 hours contact time) is within experimental error of the control. Four wt% oxalic acid is slightly more effective than 1 wt%, but this, too, is within experimental error.

In Tests 24 and 28, pre-corroded carbon-steel coupons were suspended in the oxalic acid solution to simulate internal tank structures contacted by oxalic acid. The results of these tests plotted with the data from the controls are shown in Figure 3.12. In this series of tests, 4 wt% oxalic acid appears to be slightly more effective at Cs elution than 1 wt% oxalic acid at the beginning of the test, but by the middle of the third contact, the amounts of Cs eluted are within experimental error of each other and of the controls. The presence of corroded carbon steel then appears to have no effect on the elution of Cs from zeolite.

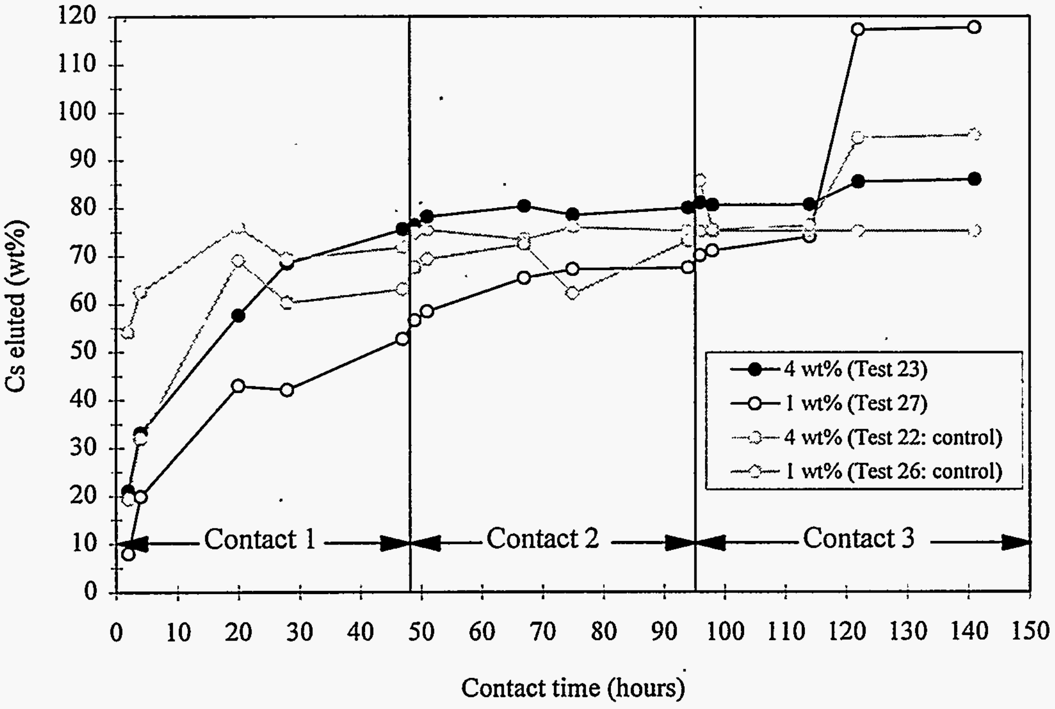

Figure 3.11. Percent Cs Eluted from Zeolite During Multiple-Contact Tests Using 4 and 1 wt% Oxalic Acid in the Presence of Additional Fe_2O_3

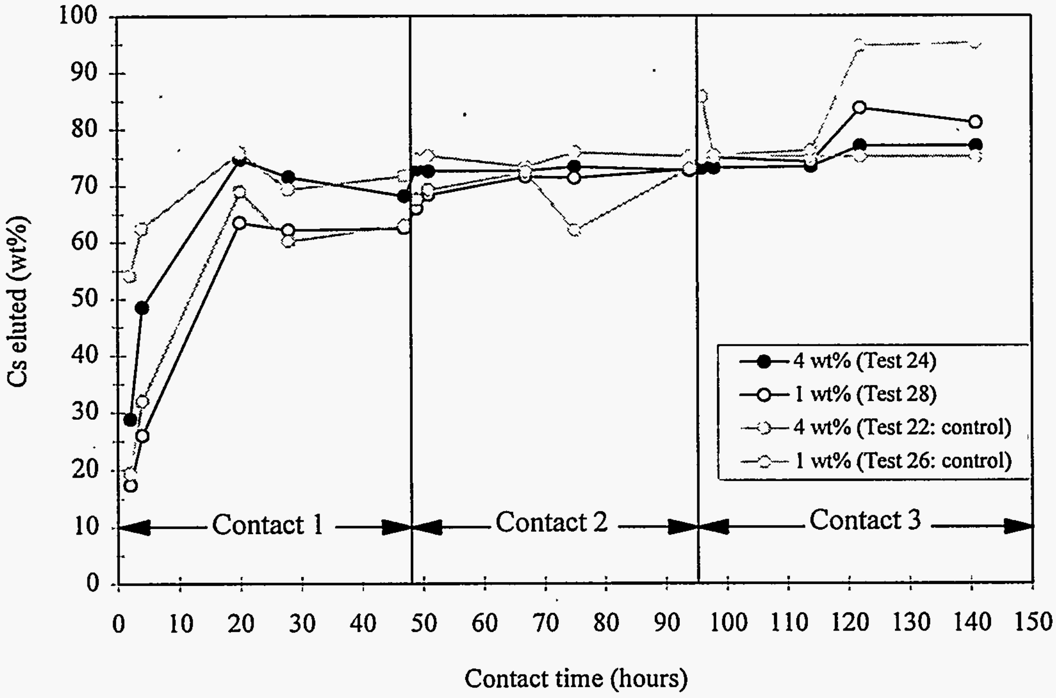

Figure 3.12. Percent Cs Eluted from Zeolite in Multiple-Contact Tests Using 4 and 1 wt% Oxalic Acid in the Presence of Carbon-Steel Coupons

Summary of Cs-Elution Results

Within experimental error, 4 wt% oxalic acid is no more effective than 1 wt% oxalic acid in the elution of Cs from zeolite. The presence of Fe in the form of rust and in the form of corroded carbon steel appears to have little effect on the elution of Cs from zeolite in laboratory-scale tests. In all of the tests, approximately 75 wt% of the Cs was eluted from the zeolite. This is compared to 100 wt% Cs eluted in the 8 wt% oxalic acid multiple-contact tests (Tests 13 through 16).

Amount of Fe Dissolved

Tests 22 and 26 were controls, run using only zeolite and 4 and 1 wt% oxalic acid, respectively. Four wt% oxalic acid dissolved ~90 wt% of the Fe present in the zeolite, while 1 wt% oxalic acid dissolved ~83 wt% of the Fe in the zeolite. In Tests 23 (4 wt% oxalic acid) and 27 (1 wt% oxalic acid), Fe_2O_3 was added to the system in amounts equivalent to the amount of loose iron, or "rust," expected to be on the bottom of the tank. The data from these tests are plotted in Figure 3.13. In these tests, the amount of Fe contributed by the zeolite is about 3 wt% of the total amount of Fe in the system; the remaining Fe is contributed by the added Fe_2O_3. The figure shows that the amount of Fe dissolved in these tests was quite a bit lower than the amount dissolved in Test 17, using 8 wt% oxalic acid, in which >90 wt% of the Fe in the system was dissolved. In both 4 and 1 wt% oxalic acid, ~15 wt% of the Fe in the system was dissolved by the middle of the third contact. Within experimental error, there is no difference in the amount of Fe dissolved by 4 wt% oxalic acid vs. 1 wt% oxalic acid.

In Tests 24, 25, 28, and 29, pre-corroded carbon-steel coupons were suspended in the oxalic acid to simulate internal tank structures contacted by oxalic acid. The results of Tests 24 and 28, in which zeolite was present, are shown in Figure 3.14. The results of Tests 22 and 26, the controls, are also plotted for comparison. The coupons used in this test were of slightly different dimensions than those used in the Test 18 (using 8 wt% oxalic acid), and the oxalic acid ratios were different, so the results of Test 18 are not included on the plot. The plot shows that at 4 wt% oxalic acid (Test 24), three times as much Fe is dissolved when carbon-steel coupons with surface areas proportional to the surface area of internal tank structures are present compared to the control (Test 22). At 1 wt% oxalic acid (Test 28), about two and one-half times as much Fe is dissolved when carbon-steel coupons are present compared to the control (Test 26). Tests 25 and 29 also contained carbon-steel coupons of proportional surface area to the internal tank structures. In these tests, however, there was no zeolite present. These tests were performed to determine whether the presence of the zeolite has any effect on the solubilization of Fe. Because these tests contained no Fe other than what was in the coupons, the actual amounts of Fe in solution were plotted. The data for Tests 24, 25, 28, and 29 in terms of ug Fe in solution are shown in Figure 3.15. At short contact time (i.e., during the first contact), the results are what one would expect: higher amounts of Fe are found in solution for tests in which zeolite and carbon-steel coupons were present (Tests 24 and 28). However, at longer times (i.e., during the third contact), in the case of 4 wt% oxalic acid (Tests 24 and 25), more Fe was found in solution when no zeolite was present, indicating that the presence of zeolite may slightly inhibit dissolution of carbon-steel coupons. If the dissolution of the coupons were the same in both tests, then one would expect there to be slightly more Fe in solution in Test 24, because the presence of Fe from the zeolite.

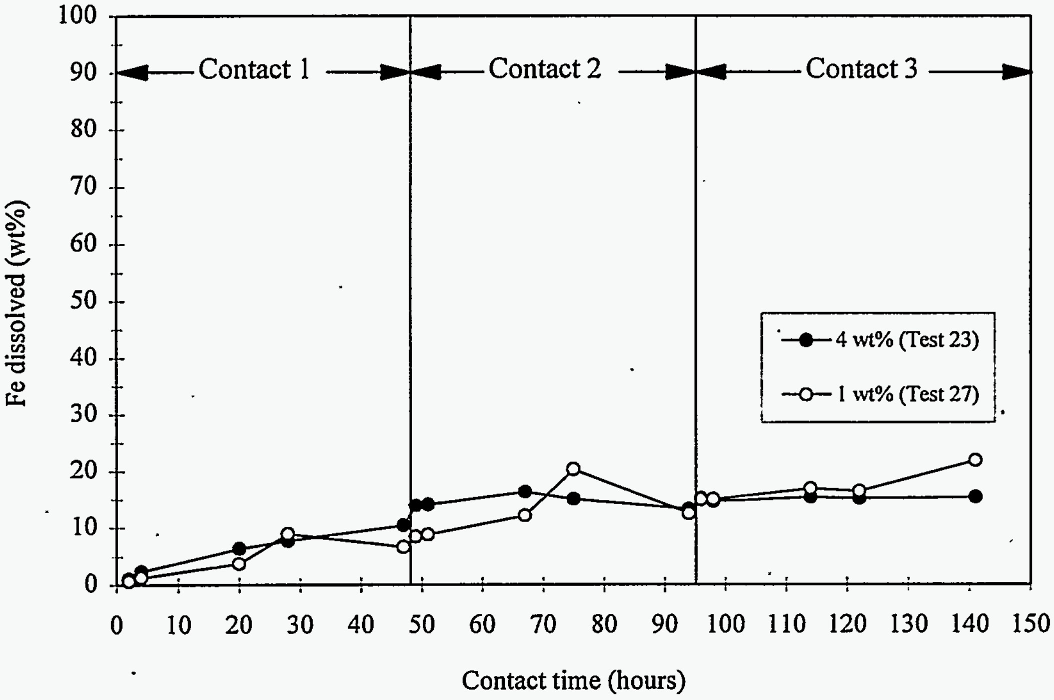

Figure 3.13 Percent Fe Dissolved in Cs-Elution Tests Using 4 and 1 wt% Oxalic Acid in the Presence of Additional Fe_2O_3

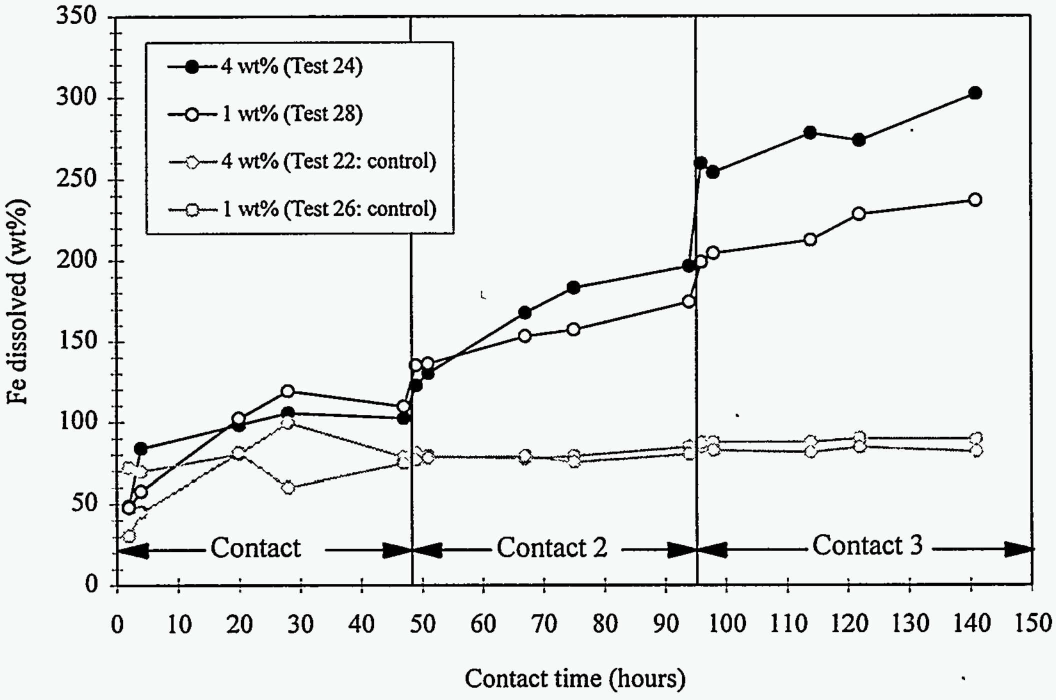

Figure 3.14. Percent Fe Dissolved in Cs-Elution Tests Using 4 and 1 wt% Oxalic Acid in the Presence of Carbon-Steel Coupons (100% = 100% of the Zeolite in the System Dissolved)

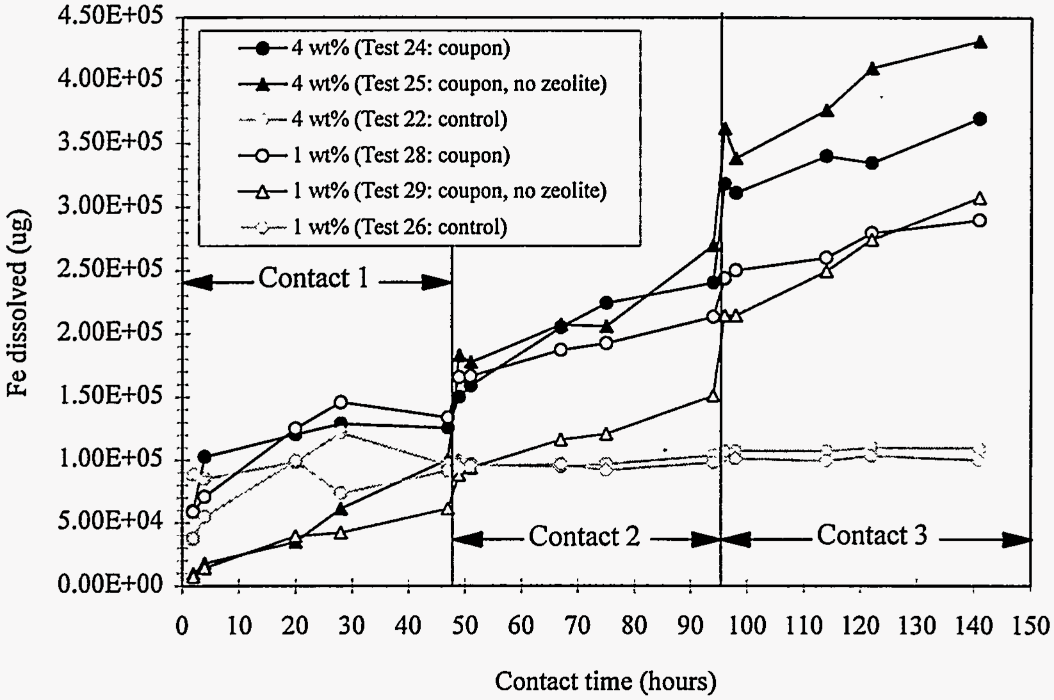

Figure 3.15. Amount of Fe Dissolved in Cs-Elution Tests Using 4 and 1 wt% Oxalic Acid in the Presence of Carbon-Steel Coupons

Corrosion of Carbon Steel Coupons

The results of the coupon tests are summarized in Table 3.16. When the coupons were first removed from the reaction vessels, they were coated with a substantial layer of soft yellow solids. Additional yellow solid material had precipitated in the vessels. Before cleaning the coupons, the yellow precipitate was analyzed by X-ray diffraction (XRD). The XRD results showed that the material was composed almost entirely of crystalline "Humboldine" ($C_2FeO_4 \cdot 2H_2O$). Because this precipitate was not noted in the earlier tests at 8 wt% oxalic acid, it is presumed that precipitation is due to lower solubility for this material in the less-concentrated acid solution.

Following XRD analysis, the coupons were cleaned, weighed, and visually examined. As shown in Table 3.16, based on weight loss, the calculated uniform corrosion rates vary noticeably; however, this is not surprising considering the short duration of the test. In spite of the variability there is a significant difference between the 1 wt% and 4 wt% tests; higher acid concentrations produced higher corrosion rates. In general, these corrosion rates are similar to those seen in the earlier 8 wt% acid tests. Based on that and the absence of any precipitate during the 8 wt% tests, it appears that the precipitated coating of Humboldine does not protect the surface of the steel from corrosion.

The coupons were examined microscopically for localized corrosion, such as pitting. None was observed. The corrosion, although quite severe, occurs uniformly over the entire exposed surfaces of the coupons.

Based on the corrosion results shown in Table 3.16, the presence of zeolite would appear to reduce corrosion of the steel. However, because there is so little zeolite material in the solution, it is not clear how this would act to inhibit steel corrosion; the apparent effect on corrosion rate may be only coincidental. Additional testing with multiple coupons would be necessary to assess this possibility better.

Table 3.16. Corrosion of Carbon-Steel Coupons During Cs-Elution Tests 24 - 29

Test	Coupon ID	Test Conditions	Weight Loss (g)	Corrosion Rate (mpy)
Test 24	599	4 wt% oxalic acid	0.9803	171
Test 25	449	4 wt% oxalic acid, no zeolite	1.1905	207
Test 28	450	1 wt% oxalic acid	0.5682	101
Test 29	593	1 wt% oxalic acid, no zeolite	0.7576	131

4.0 Sludge Dissolution: Use of Oxalic Acid to Dissolve Sludge in Tank 8D-2

4.1 Procedures

4.1.1 Sludge Simulant Preparation

The simulant used in these experiments was prepared during FY 1994 for the WVSP 0301 subtask[a] [b]. The target composition for the simulant is shown in Table 4.1. The simulated HLW was made to represent sludge and supernatant from the PUREX process, solution from the THOREX process, and the zeolite from the supernatant decontamination process. Simulant preparation simulates the process history and the chemical composition of the waste. In summary, NO_3^- solutions of the primary constituents are first prepared and co-precipitated as hydroxides. These hydroxides are then washed, if necessary, to reduce the amounts of Na^+ and NO_3^- present. Finally, other chemicals (oxides, hydroxides, and carbonates) and zeolite are added to complete the HLW simulant.

In the HLW simulant preparation, SiO_2 and ZrO_2 are used as surrogates for radioactive U and Th. For the sludge-dissolution studies, it was necessary to choose a surrogate which has similar solubility in oxalic acid and forms similar oxalate compounds to U and Th. Using these criteria, Ce was chosen as a suitable surrogate (Weast 1981) and was added to the simulant in the form of $Ce(OH)_4$ in an amount equivalent to the amounts of U and Th (on a mole basis) expected to be in the actual sludge.

4.1.2 Sludge-Dissolution Experiments: General Procedure

The objective of the sludge-dissolution experiments was to determine how much of the solid portion of the sludge simulant could be dissolved using an 8 wt% oxalic acid solution. Ideally, dissolution would mobilize the radionuclides Th and U, which are simulated in these experiments by Ce. The experiments were run in 500 mL Pyrex® reaction vessels, with ports for thermocouples, stir shafts, and sampling, as shown in Figure 3.1. An appropriate amount of sludge simulant was added to the reaction vessel, followed by an appropriate amount of 8 wt% oxalic acid. The vessel was then sealed using vacuum grease, and placed in a heating mantle with a temperature controller programmed to hold the temperature at 50°C (unless otherwise noted). Fifty degrees Celsius is the expected temperature of the material in tank 8D-2[c] A stirring shaft was placed in each vessel and the contents of the vessel were agitated throughout the test period to simulate the effects of pumps in tank 8D-2. Tests were run for 50 hours unless otherwise noted, and sampling was performed at 2, 4, 20, 28, and 50 hours of contact time, unless otherwise noted. When sampling, agitation of the material in the vessel was stopped and the material was allowed to settle for five minutes. After five minutes, approximately 5 mL were drawn from the supernatant and analyzed by ICP-MS for Al, Ca, Ce, Cr, Fe, K, Mg, Mn, Na, Ni, Sr, Ti, and Zr. Although Si also appears in the simulant, it was not analyzed for because of high

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(a) Patello, GK 1994. *Test Instruction for Preparation of West Valley HLW Simulant.* WTC-007-041-TI-1. Pacific Northwest Laboratory, Richland, Washington.

(b) Patello, GK 1994. *Preparation of High Level Waste Simulant for West Valley Melter Feed Studies.* WTC-007-041, Rev. 0. Pacific Northwest Laboratory, Richland, Washington.

(c) Fax from SC Fahey to KD Wiemers dated October 20, 1994.

Table 4.1. Initial HLW Simulant Target and Measured Compositions

Ion	Target* (ug/g)**	Measured (ug/g)	Source of Measured Value	Error
Al	6967	6537	ICP-ES†	±10%
Ca	1713	1693	ICP-ES	±10%
Ce	10437	392	ICP-MS‡	±10%
Cr	406	546	ICP-ES	±10%
Fe	39855	37900	ICP-ES	±10%
K	4021	4370	ICP-MS	±10%
Mg	227	280	ICP-MS	±10%
Mn	2908	2790	ICP-ES	±10%
Na	362591	12800	ICP-MS	±10%
Ni	679	679	ICP-ES	±10%
Sr	12127	23400	ICP-ES	±10%
Ti	9	85	ICP-ES	±10%
Zr	3417	794	ICP-ES	±10%

* based on sludge simulant preparation

** ug/g sludge simulant at 21.24 wt% total solids

† ICP-ES run in 4/95, after simulant was prepared

‡ ICP-MS run in 1/95, before testing began

instrument background interference, possibly caused by internal contamination of the instrument. In some of the experiments, the supernatant was also analyzed by ion chromatography (IC) for oxalate concentration. In these cases, an extra 5 mL sample was drawn and submitted separately. At the completion of most of the experiments, the solids were separated from the supernatant by vacuum filtration, dried, and weighed, to determine the total solids dissolved during the experiment. A discussion of sources of experimental error arising from these procedures can be found in Appendix B. A listing of all of the sludge-dissolution tests performed can be found in Appendix C.

4.1.3 Determination of Optimum Process Conditions

This first sludge-dissolution experiment involved six tests and was designed to determine the optimum process conditions for sludge dissolution. The variables in this test were the ratio of oxalic acid to sludge (20, 40, and 80 L acid/kg sludge), the initial temperature of the acid (25°C or 80°C), and the contact time (2, 4, 20, 28, or 50 hours). The various experimental conditions are tabulated in Table 4.2.

4.1.4 Repeatability Experiment

The second experiment involved four tests, and was designed to determine the repeatability of the results obtained in the first experiment. The repeatability experiment used the optimum process conditions which were determined in the first experiment, 40 L 8 wt% oxalic

Table 4.2. Experimental Conditions for Determination of Optimum Process Conditions for Sludge Dissolution

	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Oxalic acid/zeolite ratio (L/kg)	20	40	80	20	40	80
Oxalic acid temperature (°C)	25	25	25	80	80	80
Amount sludge simulant (g)	5.63	5.63	5.63	5.63	5.63	5.63
Amount 8 wt% oxalic acid (g)	114.6	229.3	458.6	114.6	229.3	458.6
Bath temperature (°C)	50	50	50	50	50	50
Agitation?	yes	yes	yes	yes	yes	yes

acid/kg sludge added at 25°C. Results from the first experiment indicated that sludge was dissolved most efficiently with contact times as short as two hours. The experimental conditions are shown in Table 4.3. Data from Test 2 (determination of optimum process conditions, see section 4.1.3, Table 4.2) and Test 16 (tests involving condition modifications, see section 4.1.7, Table 4.6) are used as repeatability tests. Tests 7 and 8 were run separately as repeatability tests, but only sampled at 2 and 4 hours. This yielded four data sets to compare for repeatability of results.

4.1.5 Long-Term Experiment

This experiment was designed to determine the effect of long-term exposure to oxalic acid on sludge dissolution. One test was run under the optimum process conditions (40 L 8 wt% oxalic acid/kg sludge added at 25°C), and sampling was done at 2 and 4 hours, and 1, 4, 7, and 11 days. The experimental conditions for this test are shown in Table 4.4.

Table 4.3 Experimental Conditions for Sludge-Dissolution Repeatability Tests

	Test 2	Test 7	Test 8	Test 16
Oxalic acid/zeolite ratio (L/kg)	40	40	40	40
Oxalic acid temperature (°C)	25	25	25	25
Amount sludge simulant (g)	5.63	5.63	5.63	5.63
Amount 8 wt% oxalic acid solution (g)	229.3	229.3	229.3	229.3
Bath temperature (°C)	50	50	50	50
Agitation?	yes	yes	yes	yes

Table 4.4. Experimental Conditions for Long-Term Sludge-Dissolution Test

	Test 9
Oxalic acid/zeolite ratio (L/kg)	40
Oxalic acid temperature (°C)	25
Amount sludge simulant (g)	5.66
Amount 8 wt% oxalic acid (g)	229.3
Bath temperature (°C)	50
Agitation?	yes

4.1.6 Multiple-Contact Experiment

The fourth sludge-dissolution experiment involved two tests and was designed to determine the effect of multiple contacts with oxalic acid on the dissolution of sludge. Using the optimum process conditions determined in the first experiment, sludge simulant was contacted with fresh oxalic acid three times for two hours. A two-hour contact time was determined to be the optimum contact time in the first experiment. After each contact, a 5 mL sample of supernatant was drawn from each reaction vessel and sent for analysis. Two types of tests were performed: in the first test (Test 10), the oxalic acid in the vessel from each contact was removed after the contact and fresh oxalic acid was added; in the second test (Test 11), the oxalic acid was not removed after each contact. The experimental conditions used for these tests are summarized in Table 4.5.

Table 4.5. Experimental Conditions for Multiple-Contact Sludge-Dissolution Tests

	Test 10	Test 11*
Oxalic acid/zeolite ratio (L/kg)	40	40
Temperature oxalic acid (°C)	25	25
Amount sludge simulant (g)	5.64	5.64
Bath temperature (°C)	50	50
Amount 8 wt% oxalic acid solution--1st contact (g)	229.3	229.3
Total amount oxalic acid in vessel--1st contact (g)	229.3	229.3
Amount 8 wt% oxalic acid solution--2nd contact (g)	229.3	229.3
Total amount oxalic acid in vessel--2nd contact (g)	229.3	458.6
Amount 8 wt% oxalic acid solution--3rd contact (g)	229.3	229.3
Total amount oxalic acid in vessel--3rd contact (g)	229.3	687.9
Agitation?	yes	yes

* The reaction vessel used for Test 11 had a 2 L capacity rather than 500 mL

4.1.7 Effects of Condition Modifications

The fifth sludge-dissolution experiment involved five tests and was designed to determine the effects on sludge dissolution of several modifications of the optimum process conditions (40 L 8 wt% oxalic acid/kg zeolite, with acid added at 25°C). The following modifications were made: presence of corrosion products, presence of carbon-steel coupons, 25°C bath temperature, and lack of agitation. The experimental conditions for this series of tests are shown in Table 4.6.

The effect of the presence of corrosion products in tank 8D-2 on sludge dissolution was examined in Test 12. Corrosion products were simulated by adding Fe_2O_3, equivalent to 2400 kg of rust at the bottom of the tank[a].

The effect of the presence of carbon-steel coupons on the dissolution of sludge was examined in Test 13. Two steel coupons were prepared as described in section 3.1.7. The initial dimensions and weights of the coupons are shown in Table 4.7. When the test was complete, the coupons were removed from the vessel, rinsed in deionized water, and dried at room temperature. The coupons were then reweighed to determine weight loss and estimate corrosion rates.

The effect of no agitation on sludge dissolution was examined in Test 14. The vessel for this test was set up in the usual manner, but without a stir shaft. The effect of a 25°C bath temperature over 50 hours, rather than the 50°C bath temperature, was examined in Test 15. Test 16 was a control, run using the optimum process conditions.

Table 4.6. Experimental Conditions for Sludge-Dissolution Tests Involving Condition Modifications

	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
Oxalic acid/zeolite ratio (L/kg)	40	40	40	40	40
Oxalic acid temperature (°C)	25	25	25	25	25
Amount sludge simulant (g)	5.64	5.63	5.65	5.63	5.63
Amount 8 wt% oxalic acid solution (g)	229.3	229.3	229.3	229.3	229.3
Bath temperature (°C)	50	50	50	25	50
Amount Fe_2O_3 (g)	0.096	*	*	*	*
Steel Coupons?	no	yes	no	no	no
Agitation?	yes	yes	no	yes	yes

* Fe_2O_3 not added

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[a] Fax from MA Schiffhauer, WVNS to KD Wiemers, PNNL dated September 27, 1994.

Table 4.7. Initial Carbon Steel Coupon Data for Sludge-Dissolution Test 13

	Cleaned Coupon	Rusted Coupon
Coupon ID	X027C	X027R
Length (cm)	2.9840	3.0678
Width (cm)	1.0376	1.0490
Thickness (cm)	0.2565	0.2591
Weight (g)	5.4380	5.6522

4.1.8 Determination of Solids Remaining

An initial weight-percent solids determination was performed on the sludge simulant(a), and yielded a result of 21.24 wt% solids. This number includes solids that were dissolved in the supernatant as well as undissolved solids.

At the completion of each experiment, most of the supernatant was drained off and collected. The remaining material was poured through a vacuum-filtration apparatus and the solids were collected on a piece of filter paper of known weight. The solids were dried at room temperature for 24 hours and then weighed. Percent solids dissolved during the experiment could then be calculated using the total weight of simulant used in the experiment, and the fact that the simulant contained 21.24 wt% solids.

Percent solids remaining was not determined in the first experiment, in which optimum process conditions were defined, nor in the repeatability tests, due to unavailability of vacuum-filtration equipment.

4.2 Results and Discussion

4.2.1 Determination of Optimum Process Conditions

The first experiment consisted of six tests designed to determine the optimum process conditions for sludge simulant dissolution in 8 wt% oxalic acid. The variables studied were: the ratio of oxalic acid to sludge simulant (20, 40, and 80 L acid/kg sludge simulant); the temperature of the acid as it was added (25°C or 80°C); and the contact time (2, 4, 20, 28, or 50 hours).

The pH and temperature of the contents of each vessel were recorded at each sampling time. These data can be found in Appendix H.

(a) Elmore, M.R. 1988. *Laboratory Procedure for the Physical and Rheological Characterization of Newtonian and Non-Newtonian Fluids.* WTC-006-4, Rev. 1. Pacific Northwest Laboratory, Richland, Washington.

The concentration of soluble oxalate was measured by IC after each contact time in each of the six tests. The results are shown graphically in Figure 4.1. The data set can be found in Appendix I. Throughout the experiment, changes in the oxalate concentration were found to be less than the analytical error of ±10%. These results suggest that within experimental error, no precipitation of oxalate was detected based on IC results.

The concentration of each sludge element in solution was measured by ICP-MS after each contact time in each of the six tests. The data can be found in the form of tables and plots of weight percent of each sludge element dissolved under the various experimental conditions in Appendix I. Sample calculations showing how ICP-MS data were converted to weight percent of each sludge element dissolved are shown in Appendix J. The data were corrected for sample removal, and this correction is reflected in the calculations in Appendix J. The ranges of dissolution for each sludge element are shown in Figure 4.2, where each dark circle represents one data point, and each data point for a given sludge element represents a different set of conditions (L acid/kg, temperature of acid addition, contact time). The analytical error associated with each data point is ±10%. For most sludge elements, a number of data points lie within experimental error of the maximum dissolution, making it difficult to state with any certainty that one particular set of conditions was the most effective for a given sludge element; however, it was possible to observe trends in the data. Upon examination of the tables and plots in Appendix I, it becomes apparent that 20 L acid/kg sludge and 25°C acid addition seem to lead to lower dissolutions, while 40 or 80 L acid/kg sludge and 80°C acid addition seem, in general, to lead to higher dissolutions. Contact time did not appear to play a major role in the dissolution of most of the sludge elements; significant changes were not observed after 4 hours contact time, and in most cases, at least 70 wt% of the sludge element had dissolved after 2 hours contact time.

For most of the sludge elements analyzed (the only exceptions were Zr and Ce), maximum dissolutions were found to be over 70 wt%. For Ca and Cr, maximum dissolutions of over 110 wt% (100% + 10% analytical error) were observed for at least one of the test conditions, leading to the conclusion that the initial values or the experimental values (or both) for Ca and Cr must not be accurate to within ±10%. The two high data points for Ce dissolution occur at 40 L acid/kg sludge and 25°C acid addition, at 28 and 50 hours contact time. Based on the fact that ICP-MS was able to detect less than 4 wt% of the Ce added to the initial simulant, it is unlikely that these data points are anything other than "flyers" (i.e., data points that are scattered beyond experimental error and cannot be explained in terms of the general trends observed). In the case of Mg, there appears to be a group of data points showing higher dissolutions and another group showing lower dissolutions. Examination of the data in Appendix I (see Figure I.7 and Table I.7) shows that the lower dissolutions occur in Tests 3 and 6, which used 80 L acid/kg sludge added at 25 and 80°C, respectively.

Since Ce was chosen as a surrogate for the radionuclides U and Th, the low dissolution of Ce from the simulant is a cause for some concern. The ICP-MS measurements of the initial simulant failed to detect more than 4 wt% of the amount added to the simulant (as Table 4.1 shows); it is entirely possible that these low percentages are due to limitations in the capability of the ICP-MS to detect Ce under these conditions. It is also possible that Ce simply does not dissolve, or if it does dissolve, it may form a cerium-oxalate type of compound and thus not be found in solution. X-ray diffraction of the solids remaining indicated that a cerium-oxalate compound was present in the solids (see section 4.2.7) but a quantitative analysis was not possible, and it is therefore unknown whether all of the Ce was tied up in this compound, or only

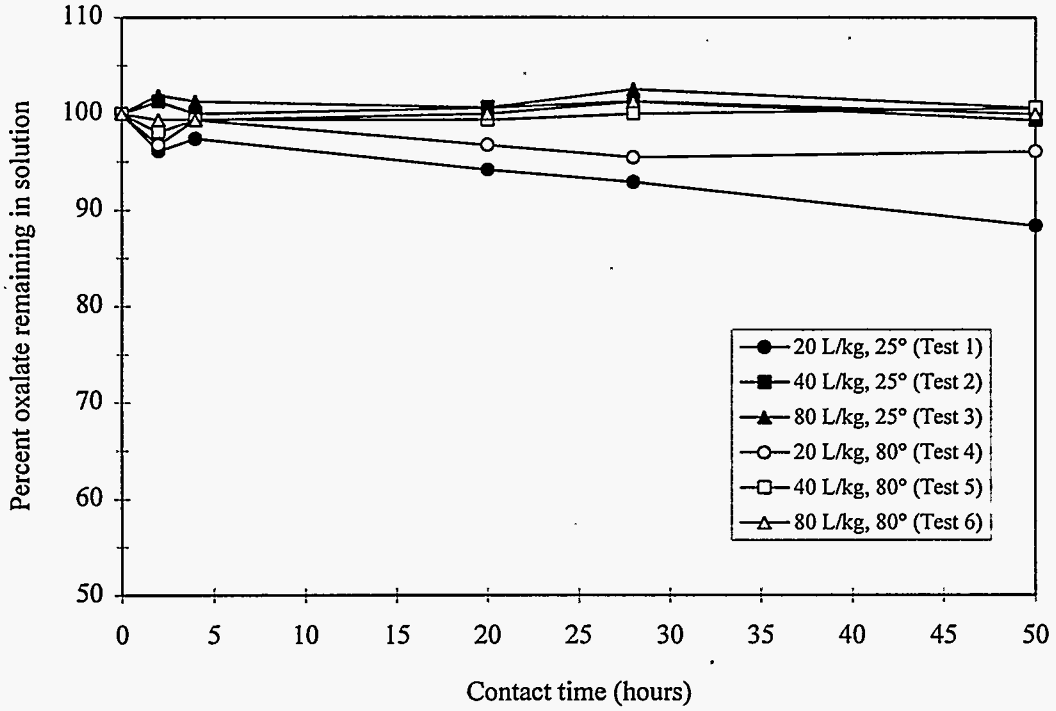

Figure 4.1. Amount of Oxalate Remaining in Solution in Experiment to Determine Optimum Process Conditions for Sludge Dissolution

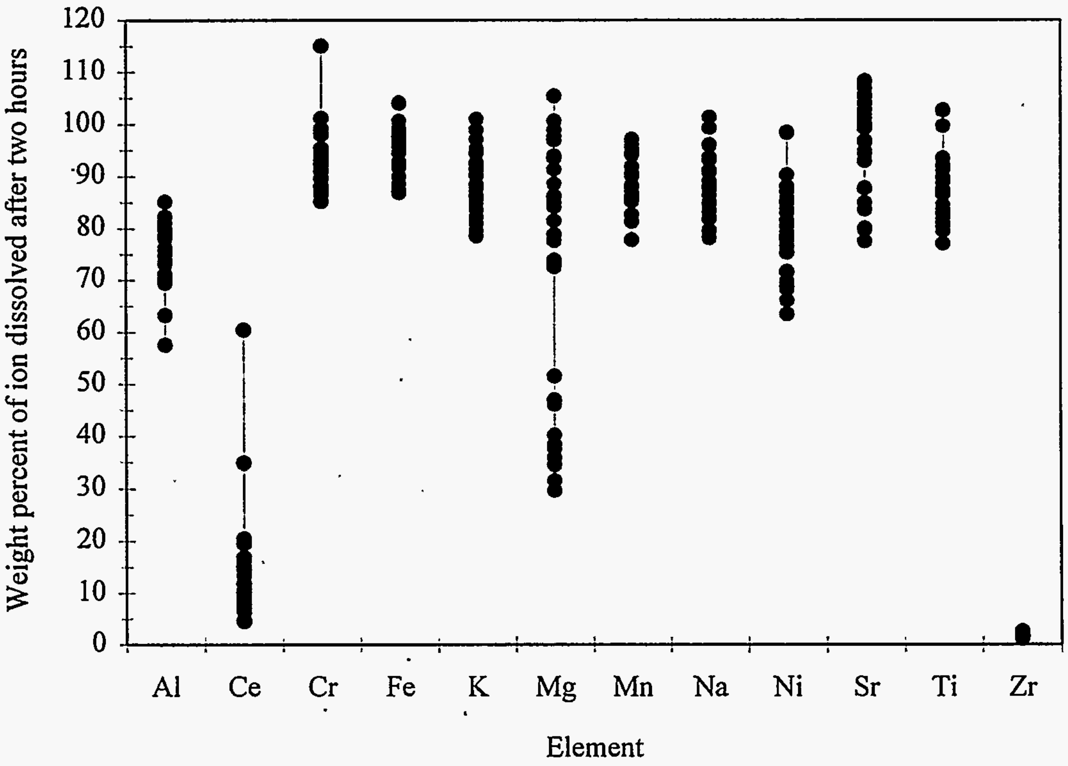

Figure 4.2. Range of Percentages of Sludge Elements Dissolved as Measured by ICP-MS

a small amount of it. Without further studies, it is difficult to say whether or not Ce was a good choice for a U and Th surrogate. These results may suggest that U and Th will not dissolve well under the conditions tested; or they may simply be an indication that Ce does not dissolve well under the conditions tested.

Optimum process conditions for sludge dissolution were chosen based on the results of these six tests and on the ease of use during plant operation. The optimum process conditions chosen were 40 L 8 wt% oxalic acid/kg sludge added at 25°C. Forty L acid/kg sludge was chosen over 80 L 8 wt% acid/kg sludge in the interest of waste minimization and because 80 L acid/kg sludge was not significantly better than 40. Twenty-five degrees Celsius acid addition was chosen because 80°C acid addition did not uniformly increase dissolution, and in cases where it did increase dissolution, the increase was on the order of a few percent. In addition, although oxalic acid is more soluble at 80°C, heating the acid would present additional operational requirements in full-scale operations.

4.2.2 Repeatability Experiment

The second experiment examined the repeatability of the sludge-dissolution tests. Each of the four tests was run using the optimum process conditions (40 L 8 wt% oxalic acid/kg sludge added at 25°C). Test 2 was run as part of the determination of the optimum process conditions (section 4.2.1); Tests 7 and 8 were run together as repeatability tests; and Test 16 was run as a control for the experiment involving the effects of condition modifications (section 4.2.5). Tests 7 and 8 were run for 4 hours, with samples taken at 2 and 4 hours; Tests 2 and 16 were run for 50 hours, but only the 2 and 4 hour data points were used for examining repeatability. The pH and temperature logs are located in Appendix H; sample calculations showing how the raw ICP-MS data were converted to percent sludge elements dissolved are shown in Appendix J. The data were corrected for sample removal, and this is reflected in the calculations in Appendix J.

The percentages of each sludge element dissolved at 2 and 4 hours of contact for each of the four tests, as well as the average and standard deviation for each data set, are shown in Table 4.8. With the exception of Ca and K, and of Ni and Al at 2 hours, the standard deviation is found to be within experimental error. The differences in dissolution of a given sludge element between 2 and 4 hours were also found to be within experimental error. The range of dissolutions for each sludge element at 2 hours contact time is shown in Figure 4.3, where the dark circle represents the average value over the four tests, and the line shows the range of percentages over which the dissolutions fell.

In general, >70 wt% of most of the sludge elements measured was dissolved after 2 hours contact; the exceptions were Ce and Zr. More than 80 wt% of Al, Cr, and Mg was found to be in solution after 2 hours, and more than 90 wt% of Fe, K, Mn, Na, and Sr was found to be in solution after two hours. The Ca results are believed to be due to a gross analytical error; one would expect Ca to exhibit similar behavior to Mg and Sr, which were observed to be >80 wt% dissolved and >90 wt% dissolved respectively, after 2 hours contact.

Table 4.8. Percentage and Average Percentage of Sludge Elements Dissolved in Sludge-Dissolution Repeatability Tests After 2 and 4 Hours Contact

Element	Hours	Test 2*	Test 7*	Test 8*	Test 16*	Average	Std Dev
Al	2	71	73	76	110	82	16
Al	4	73	68	75	71	72	2.8
Ca	2	600	260	280	440	390	140
Ca	4	580	260	280	400	380	130
Ce	2	15	2.8	4.5	1.7	5.9	5.1
Ce	4	10	4.8	6.8	1.5	5.9	3.2
Cr	2	99	83	82	86	87	7.0
Cr	4	91	74	79	81	81	6.4
Fe	2	93	95	96	92	94	1.6
Fe	4	93	91	110	96	97	7.4
K	2	88	110	120	71	96	18
K	4	92	96	100	68	90	14
Mg	2	74	87	89	84	84	5.8
Mg	4	73	93	98	82	86	9.7
Mn	2	88	97	95	95	94	3.4
Mn	4	91	87	91	94	91	2.2
Na	2	84	94	97	95	92	5.1
Na	4	82	95	91	93	90	4.9
Ni	2	70	89	87	37	71	21
Ni	4	72	86	77	64	75	8.0
Sr	2	100	100	99	79	96	10
Sr	4	95	95	120	88	99	11
Ti	2	82	72	67	63	71	7.0
Ti	4	82	74	75	64	74	6.5
Zr	2	1.1	1.2	1.2	0.78	1.1	0.2
Zr	4	1.1	1.3	1.3	0.79	1.1	0.2

* Values are ±10% except Ti (±15%) and Ca (±20%)

4.2.3 Long-Term Experiment

The third experiment examined the effects of long-term exposure to oxalic acid on sludge dissolution. One test was run using the optimum process conditions of 40 L 8 wt% oxalic acid/kg sludge, added at 25°C and held at 50°C for 11 days. The pH and temperature logs are contained in Appendix H. Sample calculations showing how raw ICP-MS data were converted to percent dissolved are shown in Appendix J. The data were corrected for sample removal, and this correction is reflected in the calculations shown in Appendix J.

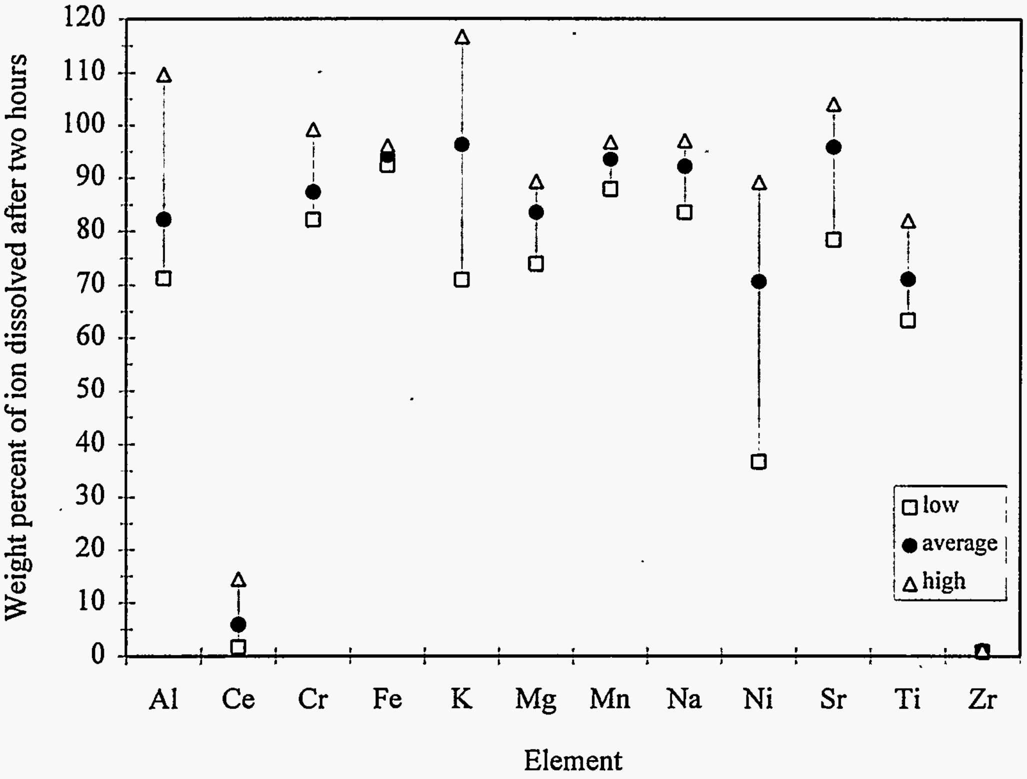

Figure 4.3. Range of Percentages of Sludge Elements Dissolved in Sludge-Dissolution Repeatability Tests After 2 Hours Contact

The results of this experiment are shown in Table 4.9 and the data are plotted in Figure 4.4 through Figure 4.7. The percent dissolved remains constant (i.e., observed changes are within experimental error) after 2 hours contact time for most of the sludge elements. A slight trend toward decreased solubility was observed for Na, Sr, and Mg after 96, 96, and 24 hours contact time, respectively. These results indicate that, as observed in the first experiment, contact time does not appear to play a major role in the dissolution of sludge simulant by oxalic acid. At 4 hours and 24 hours contact time, Ce appears to show an increase in dissolution. This may just be due to the ICP-MS technique not picking up all the of Ce in the sample, but it is also possible that the Ce begins to dissolve into the solution by 4 hours contact time, and is then tied up in some kind of oxalate complex (perhaps the one detected by XRD as described in section 4.2.7) which would explain the drop back down to 2.0 wt% at 96 hours contact time.

4.2.4 Multiple-Contact Experiment

The fourth experiment examined the effects of multiple contacts with oxalic acid on the dissolution of sludge. Sludge simulant was contacted with oxalic acid (at 40 L 8 wt% oxalic acid/kg sludge, 25°C acid addition) three times, for 2 hours. Two-hour contacts were chosen because the first experiment indicated that >70 wt% of most of the sludge elements had dissolved after 2 hours. Two tests were run: Test 10, in which the oxalic acid from each contact was removed from the reaction vessel before the next contact, and Test 11, in which the

Table 4.9. Percentage of Sludge Elements Dissolved in Long-Term Sludge-Dissolution Tests*

Sludge Element	Contact Time 2 Hours	4 Hours	24 Hours	96 Hours	168 Hours	264 Hours
Al	75	92	80	85	70	78
Ca**	390	410	410	730	270	520
Ce	2.9	21	8.0	2.3	2.9	2.3
Cr	92	98	100	95	98	84
Fe	93	97	100	160†	110	110
K	86	82	95	90	100†	84
Mg	95	94	94	86	80	85
Mn	96	100	98	110	100	99
Na	97	100	100	92	83	85
Ni	84	89	87	88	100†	70
Sr	100	100	110	100	65	72
Ti	65	68	76	73	80	68
Zr	1.0	1.1	1.1	1.0	1.3	1.1

* All values in table are ±10% except Ti (±15%) and Ca (±20%)

** These values are in error

† These values are "flyers", which fall somewhat outside the range of analytical error

fresh oxalic acid was added to the acid from previous contacts. The results of these experiments are shown in Table 4.10. The pH and temperature logs are located in Appendix H; sample calculations showing how percent dissolved was obtained from the raw ICP-MS data are shown in Appendix J. The data were corrected for sample removal, and these corrections are reflected in the calculations shown in Appendix J.

In Test 10, where the oxalic acid was removed after each contact, most of the dissolution occurred during the first two-hour contact. Additional contacts increased the total dissolution of sludge elements by only a few percent. In Test 11, where the "used" oxalic acid was left in the vessel, for many sludge elements, after the initial contact, total dissolution appeared to decrease with subsequent contacts.

The data indicate that multiple contacts with oxalic acid are not advantageous to the sludge-dissolution process. The amounts of sludge elements in solution remain constant within experimental error over the three contacts for both the case where the acid is removed after each contact and where fresh acid is added to that which is already present.

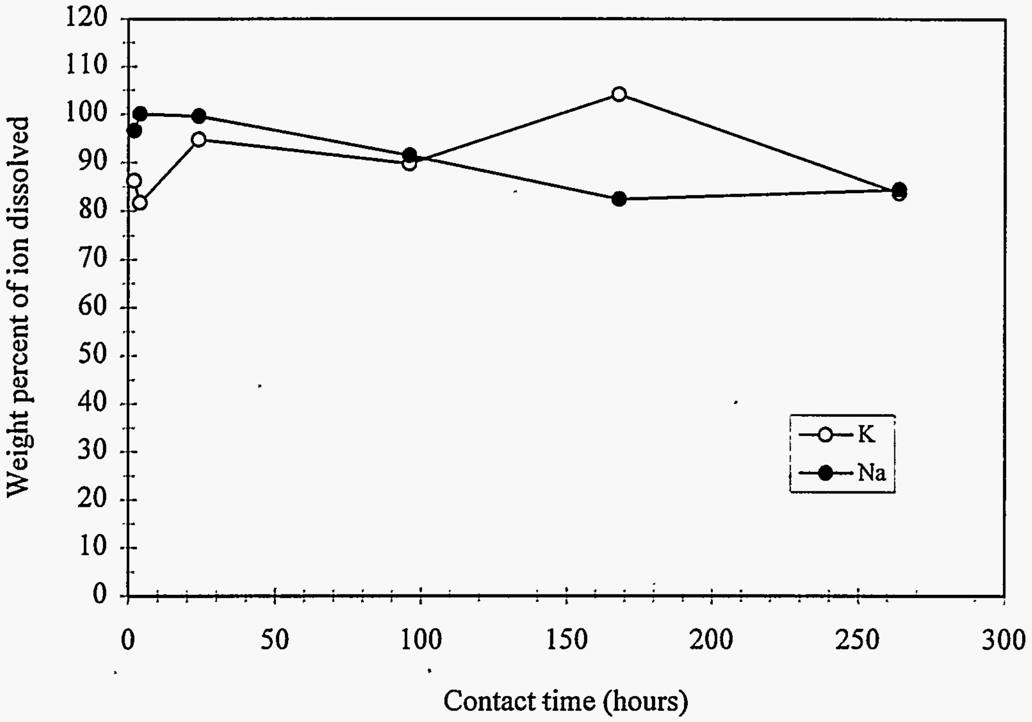

Figure 4.4. Wt% Na and K Dissolved in Long-Term Sludge-Dissolution Test

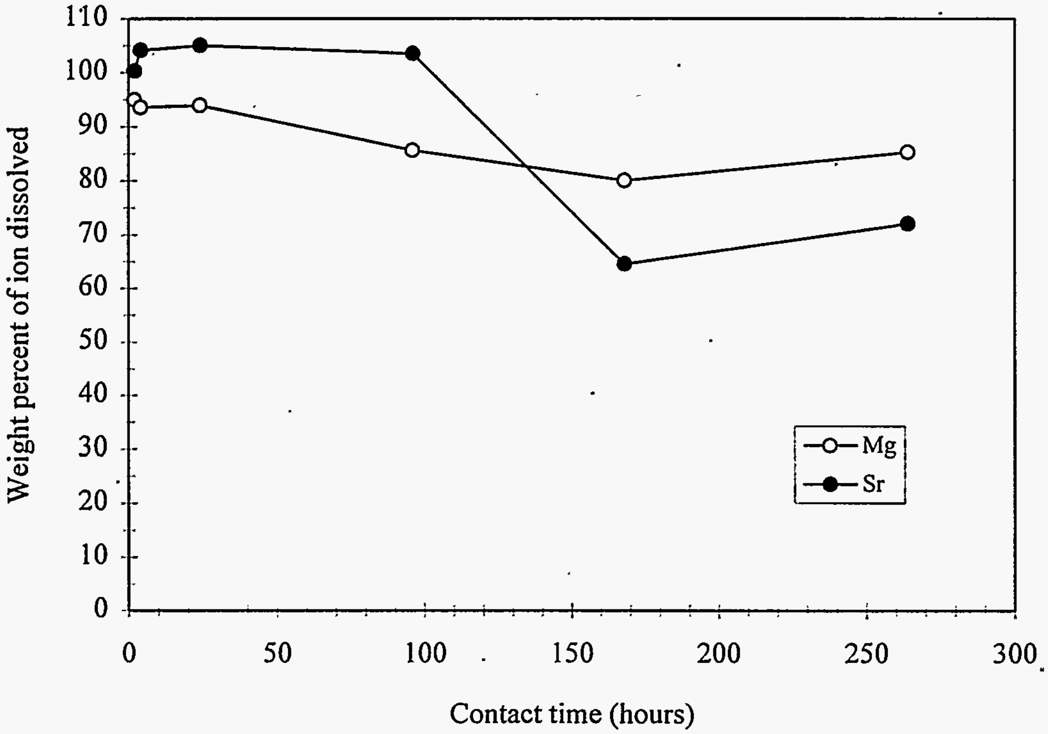

Figure 4.5. Wt% Mg and Sr Dissolved in Long-Term Sludge-Dissolution Test

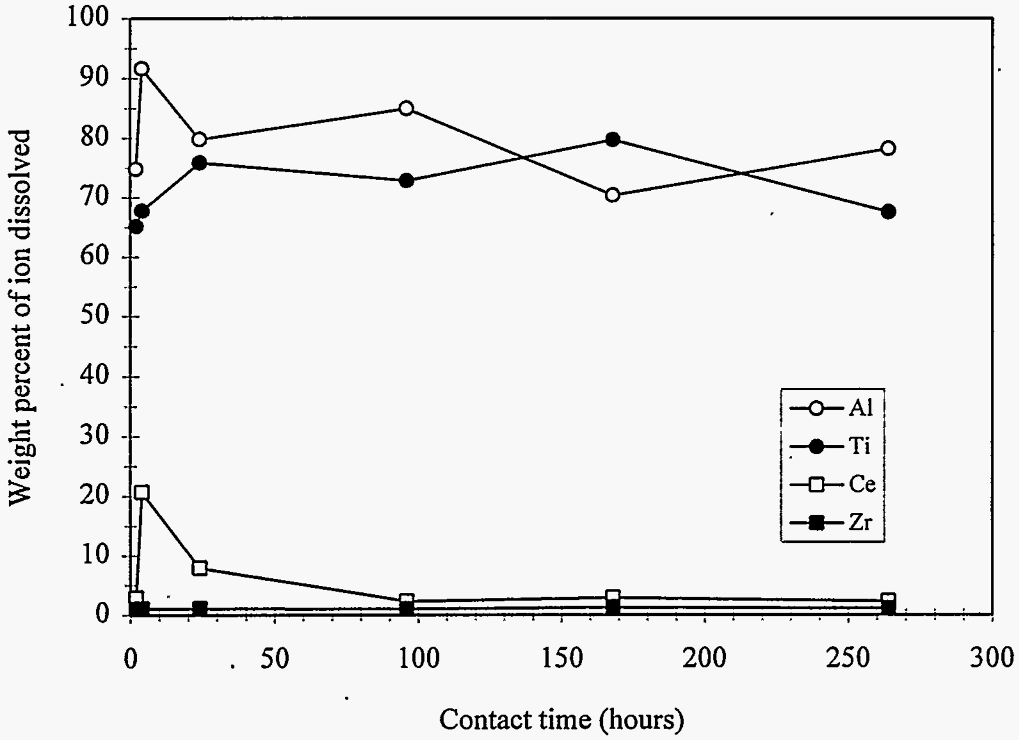

Figure 4.6. Wt% Al, Ti, Ce, and Zr Dissolved in Long-Term Sludge-Dissolution Test

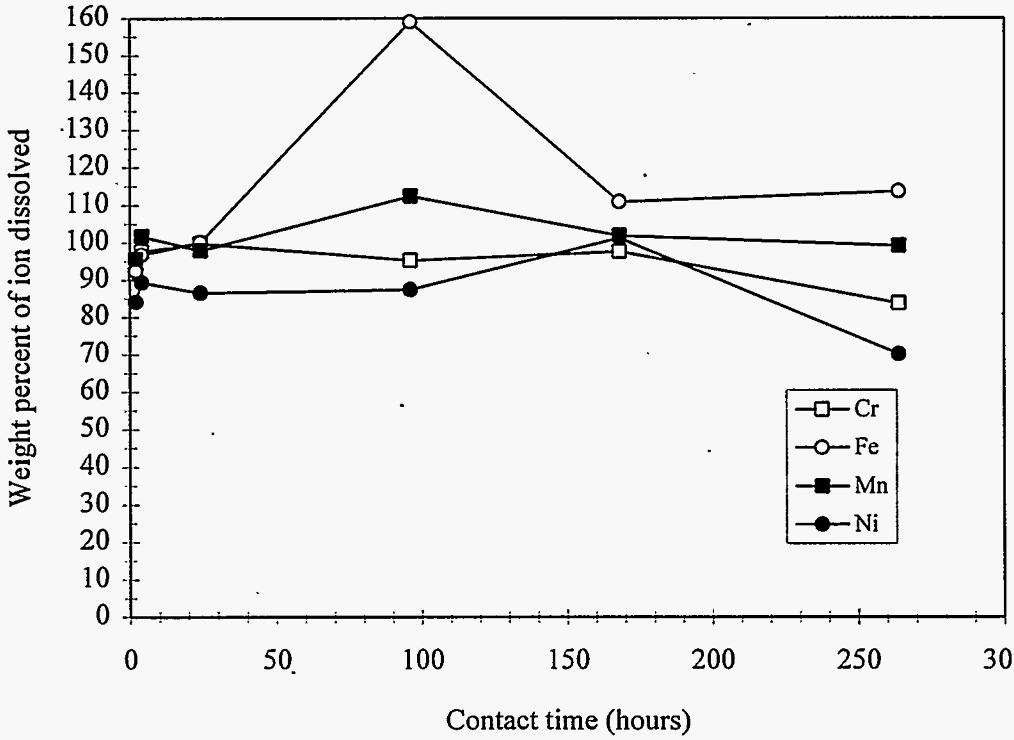

Figure 4.7. Wt% Cr, Fe, Mn, and Ni Dissolved in Long-Term Sludge-Dissolution Test

Table 4.10. Percentage of Sludge Elements Dissolved in Multiple-Contact Sludge-Dissolution Tests*

Sludge Element	Test 10 (acid removed w/ each contact) Contact 1	Contact 2	Contact 3	Test 11 (acid added w/ each contact) Contact 1	Contact 2	Contact 3
Al	73	75	76	79	75	73
Ca**	400	410	410	430	440	390
Ce	1.7	3.4	5.0	1.6	1.9	3.6
Cr	92	96	98	95	98	91
Fe	91	93	94	100	99	100
K	88	90	90	89	82	83
Mg	99	100	100	110	100	100
Mn	96	98	99	100	96	93
Na	94	95	96	110	99	90
Ni	88	90	91	88	90	83
Sr	99	100	100	110	100	100
Ti	67	69	70	70	77	72
Zr	1.0	1.1	1.2	1.1	1.1	1.1

* All values in table have a ±10% uncertainty due to analytical error, except Ti (±15%) and Ca (±20%)

** These values are in error

4.2.5 Effects of Condition Modifications

The fifth experiment examined the effects of several conditions on sludge dissolution at the optimum process conditions of 40 L 8 wt% oxalic acid/kg sludge added at 25°C. These additional conditions were: addition of Fe_2O_3 to simulate the presence of corrosion products (rust) in the tank; addition of carbon-steel coupons (one precorroded, one clean) to simulate the sides of the tank and to yield preliminary corrosion data; no agitation; and a 25°C bath temperature instead of 50°C.

At each sampling time, pH and temperature data were recorded for each reaction vessel. These data can be found in Appendix H. The concentration of each sludge element in solution was measured by ICP-MS after each contact time. These data, in the form of tables and plots of weight percent of each sludge element dissolved under the various experimental conditions, are contained in Appendix K. Sample calculations showing the method by which the raw data were converted to wt% sludge elements dissolved are shown in Appendix J. The data were corrected for sample removal; these corrections are reflected in the calculations shown in Appendix J.

The conditions in this experiment seemed to affect the dissolution of Sr, Ni, and Fe the most. Figure 4.8 through Figure 4.10 plot the data for these sludge elements. In the case of Sr, the 25°C bath decreases the Sr dissolution to well below the control level after 50 hours contact time (see Figure 4.8). Nickel dissolution is dramatically decreased by the presence of steel coupons, as shown in Figure 4.9. To a somewhat lesser extent, the dissolution of Ni is lower than the control at 50 hours under both the no-stir and the 25°C bath temperature conditions. Iron dissolution, shown in Figure 4.10, will be discussed in the following subsections.

The data indicate that these conditions had no apparent effect on the dissolution of Al and Na, and that for Cr, K, Mg, Mn (except at the 28 hour contact time), and Ti; almost all of the data points fell within analytical error of the control (see Appendix K). As has been the case throughout these experiments, Ce and Zr dissolutions were too low (< 5 wt%) to look for meaningful trends, and Ca dissolutions showed values >> 100 wt%, which is believed to be a gross analytical error.

Addition of Fe_2O_3

The addition of Fe_2O_3 to simulate corrosion products in the tank did not appear to have a significant effect on the dissolution of the sludge elements analyzed, as can be seen upon examination of the data in Appendix K. As was the case with Cs elution, the amount of oxalate in the system is in extreme excess, and it is therefore not surprising that there is no evidence of competition between the dissolution of sludge components and the dissolution of Fe. However,

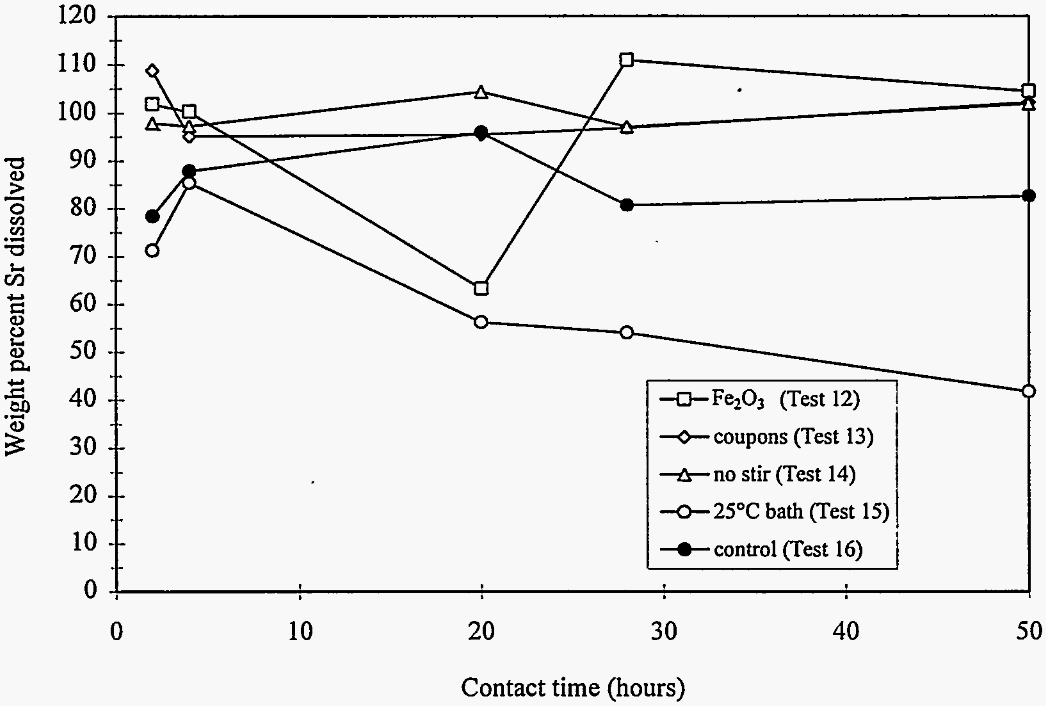

Figure 4.8. Weight Percent Sr Dissolved During Tests to Assess the Effects of Condition Modifications on Sludge Dissolution

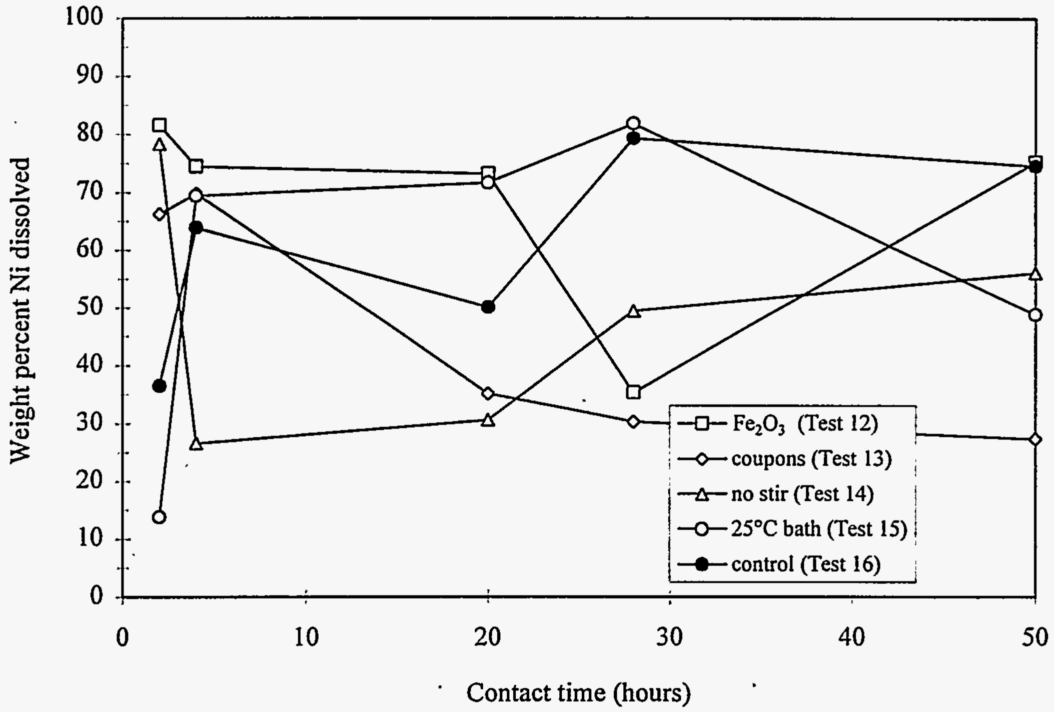

Figure 4.9. Weight Percent Ni Dissolved During Tests to Assess the Effects of Condition Modifications on Sludge Dissolution

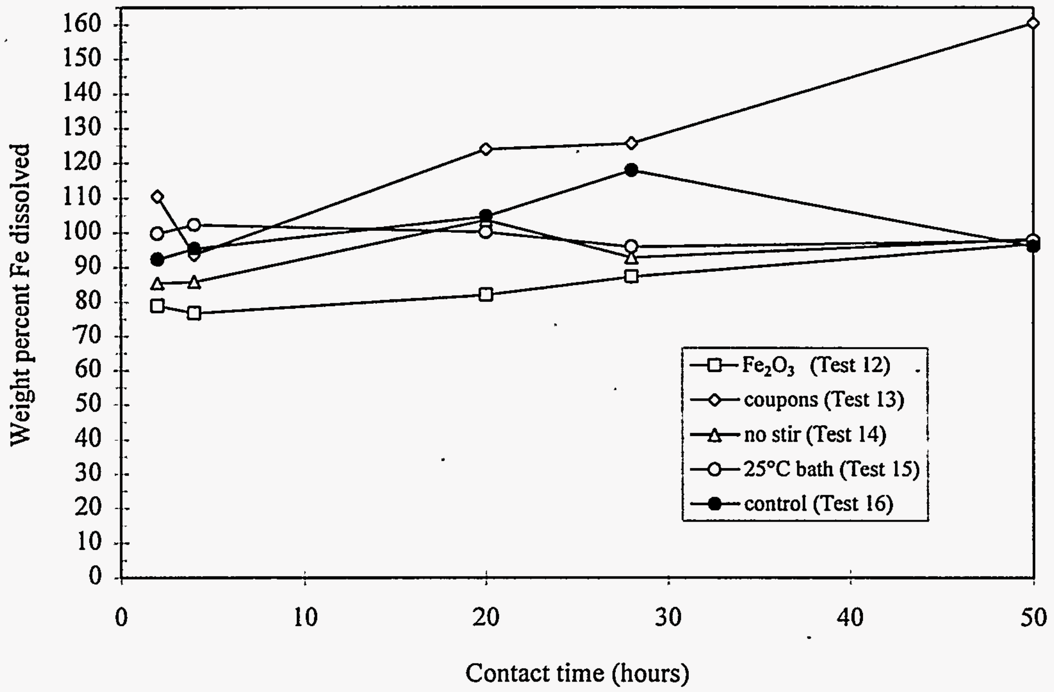

Figure 4.10. Weight Percent Fe Dissolved During Tests to Assess the Effects of Condition Modifications on Sludge Dissolution

it did have an effect on the amount of Fe in solution. In Figure 4.10, 100% Fe dissolved means 100% of the Fe added to the system. For Test 12, in which Fe_2O_3 was added, this includes Fe in the Fe_2O_3. Although Test 12 and Test 16, the control, both appear to have dissolved the same percentage of Fe, the total amount of Fe in the system is much higher for Test 12. The data indicate that 97 wt% of the Fe in the system, including loose rust at the bottom of the tank, is in solution at 50 hours contact time. This may increase the number of glass logs produced by vitrification.

It is important to note that since Fe is the major component of the sludge, any process that dissolves sludge efficiently is going to dissolve the rust and corrosion products as well. In fact, oxalic acid is used in the automobile industry as a cleaning compound for cooling systems by virtue of its efficacy in removing rust from iron (Meservey 1977). It appears, then, that maximum sludge dissolution and minimum Fe dissolution are mutually exclusive. In light of this information, it may be necessary to focus on minimizing base-metal (tank) corrosion rather than minimizing dissolution of Fe contained in rust in the tank, which might be done by lowering the acid:sludge ratio or the oxalic acid concentration.

Carbon-Steel Coupons

Figures 4.9 and 4.10 show that the presence of carbon-steel coupons in the oxalic-acid solution had an effect on the dissolution of both Ni and Fe in the sludge-dissolution experiments. The data indicate that the presence of the coupons did not have a significant effect on the dissolution of any of the other sludge elements present in the sludge simulant (see Appendix K).

The dissolution of Ni decreases dramatically with time (from 70 wt% after 4 hours contact to 30 wt% after 50 hours contact) when the coupons are present, as shown in Figure 4.9. A possible explanation for this is that Ni was plated out on the surface of the Fe. Analysis of the coupon surfaces and the stripping solution was beyond the scope of this study, but such experiments might confirm whether this is the cause of the dramatic decrease in Ni in solution when carbon-steel coupons are present. If this is the case, it is possible that once the oxalic acid cleans the oxide off of the tank wall, other metals will also be reduced and plate out on the tank surfaces. These other metals might include radionuclides, which would not be a desirable situation.

Amounts of Fe greater than 100 wt% are in solution at all contact times except 4 hours, as shown in Figure 4.10. In this test, 100% Fe is based on the amount of Fe in the sludge simulant alone; therefore, amounts greater than 100% would have to be attributed to Fe dissolved from the carbon-steel coupons. The control test indicates that at 50 hours, approximately 96 wt% of the Fe from the simulant is in solution. This suggests that amounts of Fe above 96 wt% ± 10% in the coupon tests are probably due to the dissolution of the coupons themselves.

The results of the coupon tests are summarized in Table 4.11. The reported corrosion rates are based on weight loss after cleaning in the inhibited acid cleaning solution, and assume that the weight loss, and therefore corrosion rate, is uniform over the whole surface of each coupon. For these tests, this is a reasonable assumption because no localized corrosion (i.e. pitting) was observed on any of the specimens. The calculated corrosion rates resulting from these tests are higher than anticipated. The rates may be unrealistically high because of the short duration of the tests. Corrosion rates commonly decrease with time as corrosion products accumulate on the specimen and passivate the surface, thereby inhibiting further corrosion. Longer tests would be

Table 4.11. Corrosion Rates for Coupons in Sludge-Dissolution Test 13

	Cleaned Coupon	Rusted Coupon
Coupon ID	X027C	X027R
Weight change, g	0.0717	0.0874
Surface area, cm^2	7.596	7.918
Corrosion rate, mpy*	83	97
Corrosion rate, um/yr	2100	2460

* mpy = mils (thousandths of an inch) per year

needed to determine the true long-term corrosion rate under these conditions more accurately; however, since the oxalic acid does remove the oxide coating of the carbon steel, the surface may not passivate under these conditions. If so, the corrosion rates determined from these short tests may be indicative of longer-term corrosion rates.

The results indicate that the weight losses, and therefore the corrosion rates, for the rusted coupons are higher than for the cleaned coupons. If true, the oxide coating is apparently dissolved at a higher rate than the base metal. However, with the limited number of coupons used in this testing, the significance of the differences in the corrosion rates cannot be determined. These results seem to suggest that the surface would not be passivated under these conditions, and that the longer-term corrosion rates may well be this high. Without further testing, one would expect that the corrosion rate for carbon steel under these conditions is between 80 and 100 mils per year (2000 and 2500 um/yr). This is somewhat lower than the corrosion rates observed under the Cs-elution conditions. In this test, the corrosion rates for clean vs. rusted coupons are nearly the same; in the Cs-elution test, the corrosion rate for the rusted coupon is nearly twice that of the cleaned coupon. This result would not be expected because the test conditions are very nearly the same. The effect on corrosion rates of small amounts of sludge vs. zeolite in the test solution should be negligible.

No Stirring

A lack of agitation of the reaction vessel contents did not appear to have a significant effect on the dissolution of any of the sludge elements analyzed in these tests except for Ni, which exhibited slightly lower dissolution than the control at longer contact times.

25°C Bath

A 25°C bath temperature appeared to affect the dissolution of both Sr and Ni, as is shown in Figures 4.8 and 4.9. Both of these sludge elements exhibit somewhat lower dissolution than the control in a 25°C bath.

4.2.6 Solids Dissolved in Sludge-Dissolution Experiments

The solids remaining at the end of most of the experiments were collected and weighed to determine the amount of solids that had been dissolved by treatment. A description of the

Table 4.12. Solids Dissolved in Sludge-Dissolution Experiments After 50 Hours Contact with Oxalic Acid

Experiment	Conditions	Wt% Solids Dissolved
Test 9	long-term	76.0
Test 10	multiple contact	76.8
Test 11	multiple contact	76.6
Test 12	Fe_2O_3 addition	71.7
Test 13	steel coupons	75.1
Test 14	no stir	75.2
Test 15	25°C bath	69.3
Test 16	control	74.2

methodology is provided in section 4.1.8. The results are shown in Table 4.12. An average of 74 wt% (with a standard deviation of 2.6 wt%) of the solids was dissolved in the tests listed in the table.

4.2.7 Characterization of Solids Remaining in Sludge-Dissolution Experiments

X-ray diffraction was performed on the remaining solid material from Test 2, in the first set of experiments. The material was found to be a complex mixture of phases. The main phases identified are listed in Table 4.13 along with a semi-quantitative weight %. Additional phases are likely to be present, but were not identifiable, and the semi-quantitative analysis ignores the presence of additional unidentified phases. The presence of SiO_2 and ZrO_2 is not surprising; since these are both difficult compounds to dissolve. Also, the large percentage of $C_6H_{20}Ce_2O_{22}$ (cerium oxalate hydroxide) present agrees with the ICP-MS analysis for Ce in the supernatants, which indicated that very little of the Ce initially present in the sludge had been dissolved. Although the presence of other unidentified phases makes it impossible to calculate exactly how much Ce (and hence how much U and Th) might be tied up in oxalate precipitates, calculations (see Appendix A) show that even if all of the Ce (U and Th) present precipitated out as $C_6H_{20}Ce_2O_{22}$, only 0.14% of the oxalate present would be tied up in the complex (at 40 L 8 wt% oxalic acid/kg sludge).

Table 4.13. Phases Identified in Solids Remaining After Sludge Dissolution at Optimum Process Conditions (50 Hour Contact Time)

Phase	Semi-Quantitative wt%
Quartz, SiO_2	35%
Baddeleyite, ZrO_2	15%
$C_6H_{20}Ce_2O_{22}$#	35%
Dawsonite, $NaAlCO_3(OH)_2$*	10%
$C_6Fe_2O_{12}$*	5%

cerium oxalate hydroxide

* tentative ID at best

5.0 Mild-Steel Corrosion Studies

5.1 Procedures

The overall approach to this testing was to expose steel corrosion specimens to environments simulating the anticipated conditions in the waste tank(s) during Cs elution from zeolite particles. The selected test temperature (50°C) was considered to be a conservative upper limit for in-tank processing. Two oxalic acid concentrations were selected (4 and 8 wt% acid), which represent the anticipated processing range. The test solutions in each vessel were removed and replaced with fresh acid solution after each week of specimen exposure, simulating proposed multiple contacts of the zeolite with batches of oxalic acid. Following exposures ranging from 1 to 3 weeks, the specimens were removed from the vessels, cleaned, and examined for corrosion. General corrosion rates were calculated from weight loss measurements. Pitting density and pit depths were determined optically.

5.1.1 Test Materials

The materials used for this testing included the oxalic acid solutions (4 and 8 wt%) and steel corrosion specimens (ASTM A-516 grade 55 mild steel). The oxalic acid solutions were prepared by dissolving crystalline oxalic acid in deionized water. Fresh 4 and 8 wt% acid solutions were prepared each week, replacing the previous week's test solutions to simulate a multiple-contact processing scenario where spent acid would be pumped out and fresh acid added back at frequent intervals.

The steel test specimens used for these corrosion tests were procured from Metal Samples Co., Munford, AL, and met the ASTM A-516, Grade 55 specification. Available records indicate that the ASTM designation for the type of steel used in fabricating tanks 8D-1 and 8D-2 was ASTM A-201A. This designation has since been replaced with the designation ASTM A-516 (Grade 55 for carbon <0.18%), which has essentially the same chemical composition and microstructure as material conforming to the earlier specification. (Microstructural and chemical analyses had been previously conducted verifying conformance of archived steel from Tank 8D-1 with the A-516 Grade 55 designation.)

The dimensions of each specimen were measured with digital calipers to the nearest 0.025 mm (1 mil). Typical specimen dimensions were 5 cm x 2 cm x 0.33 cm. A final thorough cleaning of the specimens was performed in a 28-g/l trisodium phosphate (TSP)/water solution, followed by deionized water-rinse and air-dry. The specimens were then weighed on a 4-place analytical balance to the nearest 0.0001 grams. Typical initial weights ranged from ~22 to ~23 grams.

5.1.2 Test Apparatus

The test apparatus used for this testing consisted of four ~4-liter Teflon®-lined vessels (similar to resin kettles) with removable lids. These vessels were partially immersed in and heated by a controlled temperature oil bath, and were fitted with reflux condensers to minimize evaporation. The vessel temperatures were monitored with thermocouples inserted through ports in the lids of the vessels. A sketch of the test setup is shown in Figure 5.1.

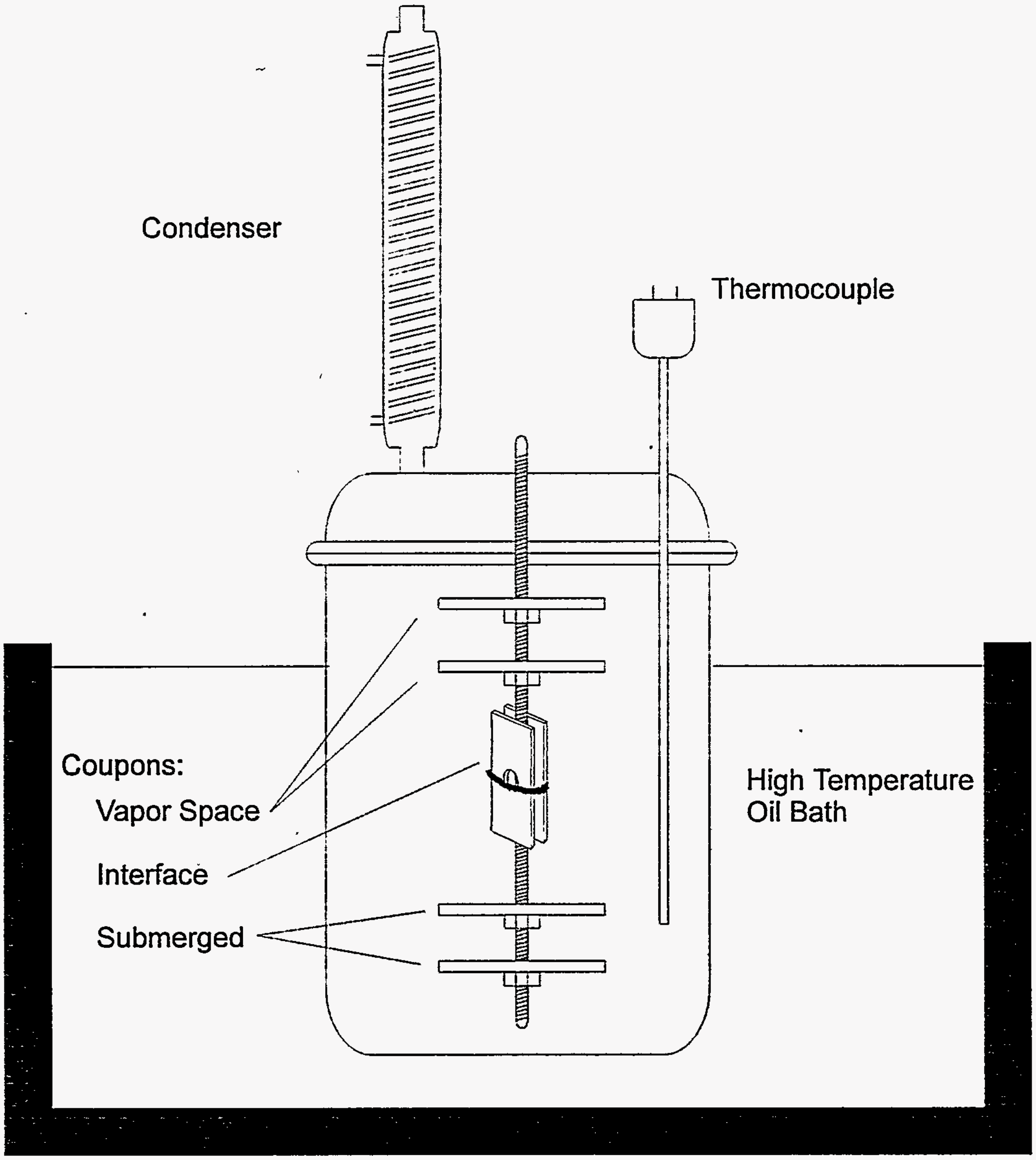

Figure 5.1. Corrosion Test Apparatus Showing Vapor Space, Interface, and Submerged Corrosion Specimens in Test Vessel

5.1.3 Test Procedure

Once the specimens were cleaned and weighed, they were placed in the test vessels, and were suspended on Teflon rods. Specimens were oriented, as shown in Figure 5.1, such that some of the specimens were exposed to the vapor space in the tops of the vessels, some were totally submerged in the solutions, and others (only the 3-week exposure sets) were suspended partially submerged at the vapor/liquid interface. The vessels were assembled and sealed, oxalic acid solution was added, and the oil bath was adjusted to maintain the vessels at 50°C. Throughout the test, the vessels were monitored to ensure that the temperature of the vessels was constant and that water did not evaporate from the vessels.

One set of specimens (duplicate vapor and submerged specimens) was removed from each vessel after one week exposure. The acid solution was replaced, the removed set of specimens was replaced with new specimens, and the test continued for two more weeks with acid solution replacement following the second week of exposure. In this way, one set of specimens was exposed for one week, a second set was exposed for two weeks (the final two weeks) and a third set of specimens was exposed for the entire three weeks of the test.

Examination of the corrosion specimens included: (1) observing the general appearance of the specimens as they were removed from the vessels; (2) cleaning the specimens in an inhibited acid cleaning solution to remove corrosion products; (3) weighing the cleaned specimens to then calculate corrosion rate from the weight loss; (4) microscopic examination to characterize the type(s) and extent of attack visually; and (5) measuring pitting depth with an optical micrometer.

5.2 Results and Discussion

The typical appearance of the specimens after exposure in the test, and before removing the corrosion products with the inhibited acid cleaning is illustrated by Figures 5.2 and 5.3. Figure 5.2 is a photograph of specimens removed from vessel 2 (4 wt% acid) at the end of the test following 2-week and 3-week exposures (3-week set has the additional pair of interfacial specimens). Note the heavy accumulation of precipitate (bright yellow) covering the submerged specimens. This precipitate had previously been identified by X-ray diffraction analysis as Humboldine, a common crystalline form of iron oxalate. In addition to the accumulation on the specimens, a large amount of precipitate was observed in the bottoms of the vessels at the end of each of the one-week intervals. The appearance of specimens taken from vessel 1 (duplicate vessel, also 4 wt% acid) was similar.

The corresponding set of samples after removal from vessel 3 (8 wt% oxalic acid) are shown in Figure 5.3. Note that the accumulation of iron oxalate on these specimens is significantly less than in the vessels with 4 wt% acid(a). An accumulation of a crystallized material is seen at the top of the interface coupons, which was identified as oxalic acid crystals by X-ray diffraction. The reason for the acid crystallizing around the specimens on the surface of the acid solution is unclear, but may be related to the changing solubility of oxalic acid at the test conditions.

A search for data on the solubility of oxalic acid under different conditions relevant to these tests revealed little information. The solubility in water at 25°C is about 10 wt%, and increases with temperature; however, the presence of other salts in even minor quantities may significantly depress the solubility of oxalic acid. For example, one study showed that the addition of ~1 wt% sodium oxalate decreased the solubility of oxalic acid from ~10 wt% to ~1 wt% (Linke 1965). Another study reported similar effects for the addition of ~1 wt% sodium hydroxide (i.e., decreased solubility of oxalic acid by an order of magnitude). It is conceivable that the appearance of iron in solution as steel corrodes may produce a similar effect on the acid solubility. Likewise, the solubility of iron species in oxalic acid solutions probably varies with acid concentration, although data on solubility in the iron-oxalic acid-water system were not located. The summation of these effects may explain, in part, the appearance of oxalic acid

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(a) The black and white photo makes it difficult to see, but these were the observations made in the lab.

crystals on the surface of the 8 wt% acid tests, and differences in corrosion rate and iron oxalate precipitate. These factors need to be further evaluated to determine the significance, if any, to corrosion of the steel and subsequently the proper processing conditions for Cs elution.

After cleaning the specimens in inhibited HCl to remove the corrosion products, the final specimen weights were recorded. Overall corrosion rates were calculated from weight loss determinations. The results of the weight-loss-derived overall corrosion rates, showing the range and average for measurements on duplicate specimens and duplicate vessels are summarized in Table 5.1. Individual specimen results are included in Appendix L.

Typical sets of specimens after final cleaning are shown in Figures 5.4 and 5.5. Figure 5.4 shows a set of specimens taken from vessel 2 (4 wt% acid) after 3 weeks exposure. The vapor-space specimens were only lightly attacked. (The dark appearance of the right-hand vapor-space specimens in Figure 5.4 is due to limitations of the camera flash. Its actual appearance is similar to the left-hand specimen. The same is true for both of the vapor-space specimens in Figure 5.5.) The submerged specimens in Figure 5.4 show the relatively deep and uneven attack characteristic of the specimens exposed to the 4 wt% acid solutions. The interface specimens (middle row) show marks across the width of the mounting hole. This is a crevice-type of attack that occurred under Teflon tape used to secure the specimens in an upright position to the holder. The actual liquid interface was on the upper half of these specimens, but is not readily apparent because the precipitating iron oxalate is believed to have wicked the acid up to the top of the specimen and corroded the entire specimen, including the portion sticking above the liquid surface.

The steel corrosion exhibited by some of the heavily corroded submerged specimens, as shown in Figure 5.4, was not a smooth uniform dissolution of the specimens' surfaces, but rather was characterized by rough irregular pits and crevices. Because the population densities of these pits were so high such that they effectively blended into one another, the approximation of penetration rate by overall weight loss is a reasonable estimate. Some of the deepest pits penetrated further than would be estimated from total weight loss, but the original surfaces on some of these specimens (a necessary reference point for pit-depth measurements) were obliterated by the heavy corrosion. This is illustrated in Figure 5.6 where the photograph shows a close-up of a specimen taken from vessel 2 (4 wt% acid) after 3 weeks exposure. Where reference to the original surface could still be made (which was true for most of the specimens), pit-depth measurements were performed with an optical micrometer. The depths of the deepest pits are included in Table 5.1. Pit depths and pit (population) densities for individual specimens are given in the table in Appendix L.

A set of specimens after 3 weeks exposure in vessel 3 (8 wt% acid) is shown in Figure 5.5. These specimens show much less attack than those exposed to the 4 wt% acid. On the middle pair of specimens (liquid interface) one can see the slight crevice attack that occurred under the Teflon tape. The position of the solution interface is also visible on the top halves of both of the interface specimens. These specimens had much less precipitate on their surfaces, which may have led to less wicking of the acid solution up the sides of the specimens when compared with the specimens in Figure 5.4.

Although Figures 5.4 and 5.5 are representative of the other sets of specimens, photographs were taken of all specimen sets after final cleaning. For completeness, a full set of photographs is included in Appendix M.

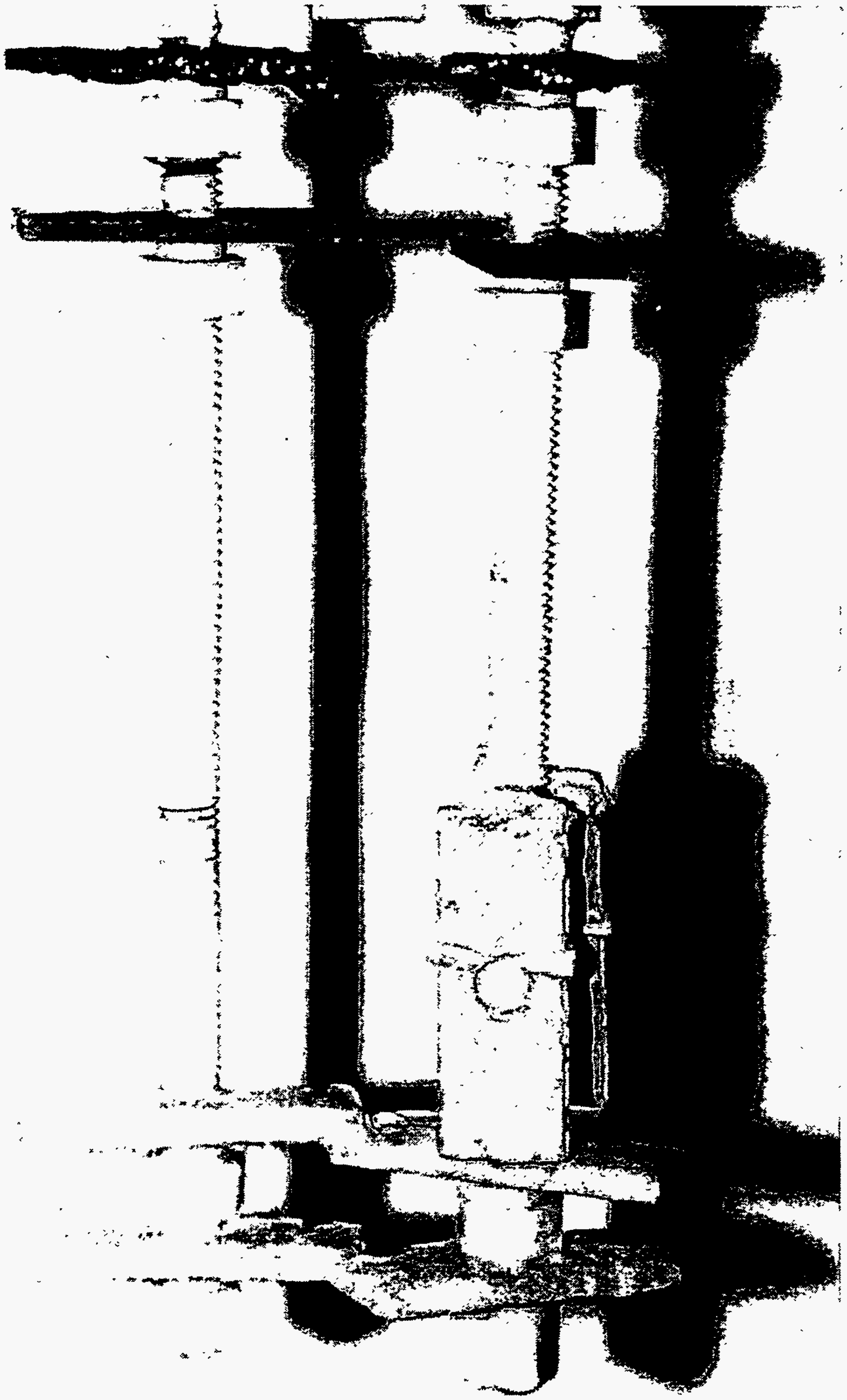

Figure 5.2. Corrosion Specimens Removed from Vessel 2 (4 wt% Oxalic Acid) at Conclusion of Test

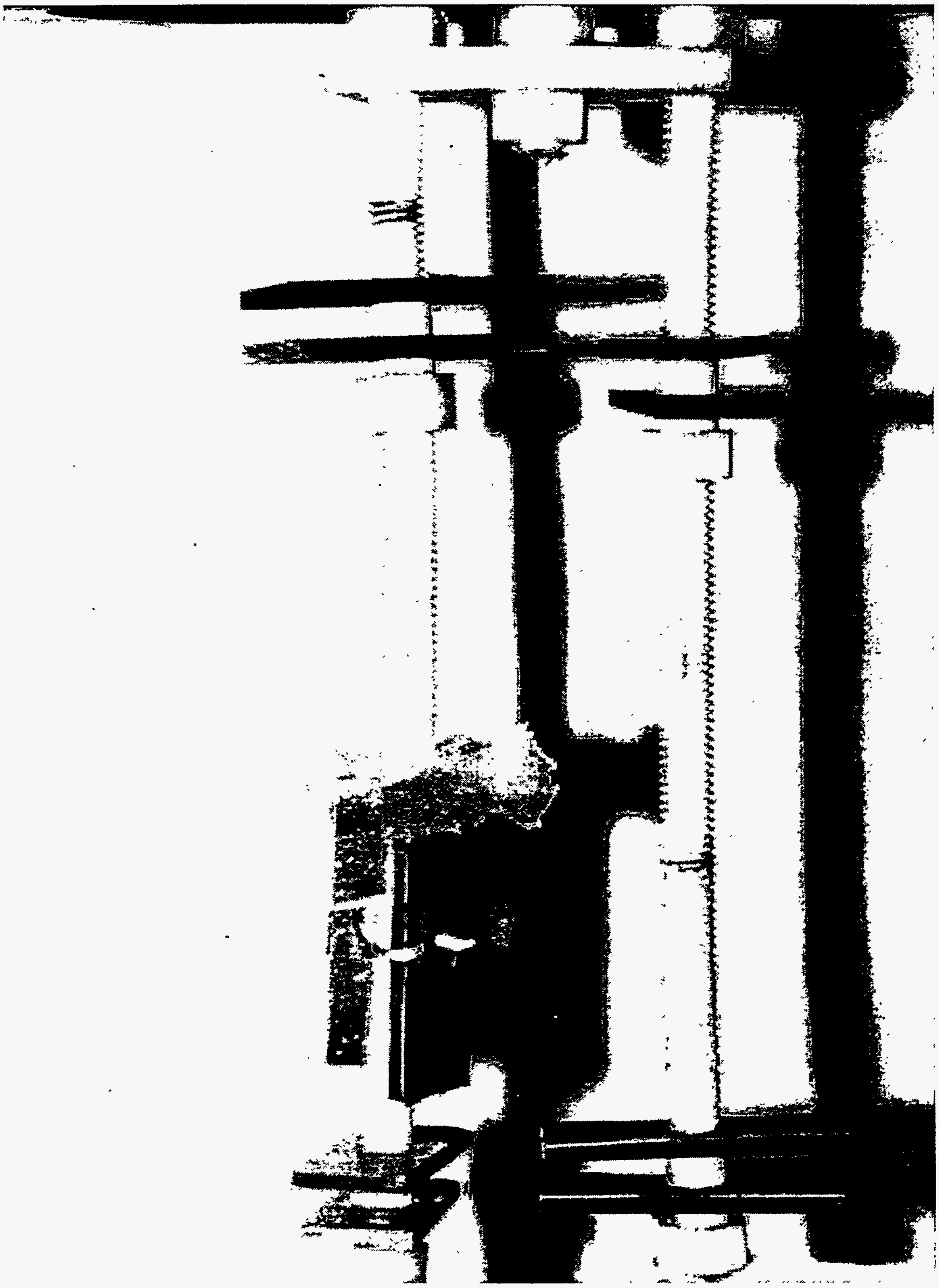

Figure 5.3. Corrosion Specimens Removed from Vessel 3
(8 wt% Oxalic Acid) at Conclusion of Test

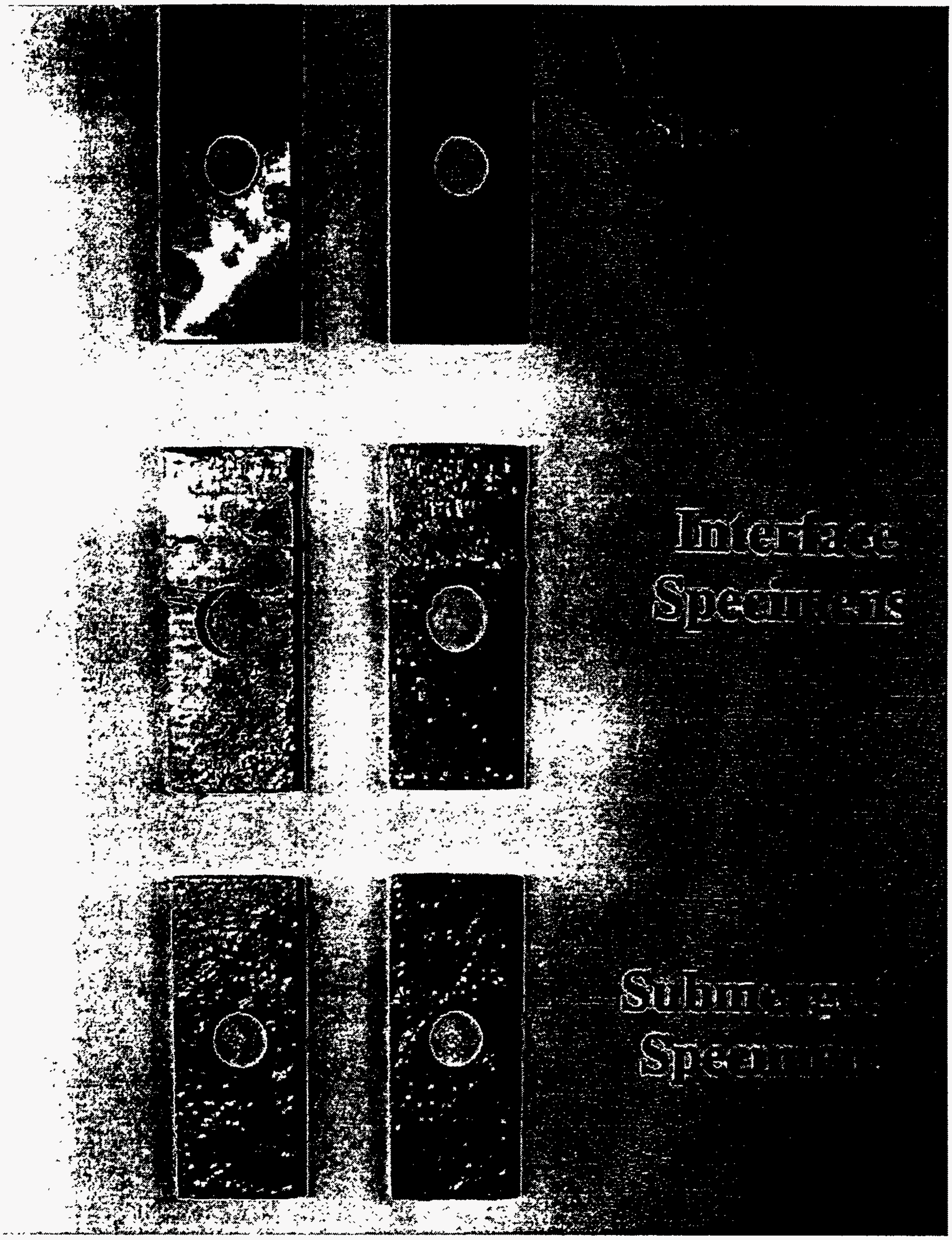

Figure 5.4. Corrosion Specimens Removed from Vessel 2 (4 wt% Oxalic Acid) After 3 Weeks Exposure

Figure 5.5. Corrosion Specimens Removed from Vessel 3 (8 wt% Oxalic Acid) After 3 Weeks Exposure

Table 5.1. Corrosion Test Results of 1-, 2-, and 3-Week Exposures of Mild-Steel Specimens to 4 and 8 wt% Oxalic Acid Solutions

Oxalic Acid	Exposure	Specimen	Corrosion Rate			Pit Depths	
Concentration	(weeks)	Position	(mpy)*	(mm/yr)	Pit Population	(mils)	(mm)
4 wt%	1	vapor space	< 1 - 8	< 0.02 - 0.2	none - few	0 - 4	0 - 0.1
4 wt%	1	submerged	37 - 118	0.9 - 3.0	many	2 - 6	0.05 - 0.15
4 wt%	2	vapor space	1 - 5	0.02 - 0.13	few	< 1 - 1	< 0.02 - 0.02
4 wt%	2	submerged	75 - 110	1.9 - 2.8	many	8 - 10	0.2 - 0.25
4 wt%	3	vapor space	1 - 2	0.02 - 0.05	few	< 1	< 0.02
4 wt%	3	submerged	126 - 165	3.2 - 4.2	many	8 - 15	0.2 - 0.38
8 wt%	1	vapor space	1 - 8	0.02 - 0.2	none - few	0 - 2	0 - 0.05
8 wt%	1	submerged	30 - 47	0.76 - 1.2	moderate	1 - 4	0.02 - 0.1
8 wt%	2	vapor space	< 1	< 0.02	none - few	< 1	< 0.02
8 wt%	2	submerged	16 - 20	0.41 - 0.51	many	< 1	< 0.02
8 wt%	3	vapor space	< 1 - 1	< 0.02 - 0.02	none - few	< 1	< 0.02
8 wt%	3	submerged	16 - 20	0.41 - 0.51	moderate	< 1	< 0.02

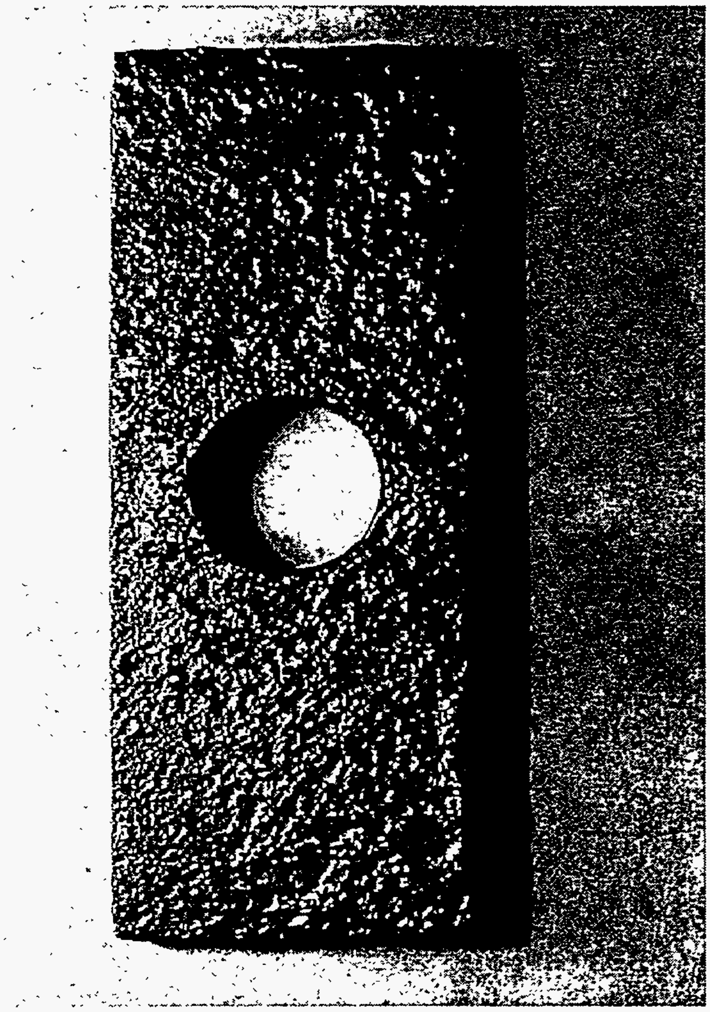

Figure 5.6. Close-up View of "Submerged" Specimen (Shown in Figure 5.4)

6.0 Modeling of Oxalate-Treated Sludge at Thermodynamic Equilibrium

6.1 Procedure

The modeling activities were designed to parallel the experimental work. Residual sludge was modeled based on the experimental procedure for preparing the sludge simulant(a)(b). The dissolution of the residual waste by adding oxalic acid was then modeled. The specific questions to be answered by the modeling work were as listed below:

- What fraction of each species (especially Fe, U, and Th) will be soluble after treatment with oxalic acid?
- How well does the Ce surrogate simulate the process chemistry of U and Th?
- What oxalate precipitates will form?
- What soluble oxalate complexes will form?

6.1.1 Modeling Approach

The WVNS sludge-oxalate system was modeled using an equilibrium thermodynamic computer model called the Environmental Simulation Program (ESP) (OLI Systems Inc., 1993). The ESP was developed to model aqueous, conventional, and complex chemical systems, including interphase equilibria, intraphase equilibria, and reaction kinetics. An extensive database of species physical and thermodynamic information is included with the ESP code. This database includes most of the species in the Design Institute for Physical Properties Project 801 data compilation, many of the species on the United States Environmental Protection Agency List of Lists, and the European Red, Grey, and Black Lists. The database provided with ESP contained most of the solution and solid species required to model the sludge-oxalate system, with the exception of some solid oxalate species. Additional data for these species was added to the ESP database to support this effort; this data is shown in Table 6.1.

Three dissolution cases were modeled using the three 8 wt% oxalic acid:sludge ratios examined in the laboratory experiments (20, 40, and 80 L acid/kg sludge).

Assumptions Made Regarding the Chemical System

Assumptions regarding the chemical system were required to allow modeling of the system. These assumptions were made based on the best technical judgment of the modelers and, where possible, were based on related observations or previously reported results. The assumptions, basis, and validity of the assumptions are discussed below.

(a) Patello, G.K. 1994. *Test Instruction for Preparation of West Valley HLW Simulant.* WTC-007-041-TI-1. Pacific Northwest Laboratory, Richland, Washington.

(b) Patello, G.K. 1994. *Preparation of High Level Waste Simulant for West Valley Melter Feed Studies.* WTC-007-041, Rev. 0. Pacific Northwest Laboratory, Richland, Washington.

Table 6.1. Additional Data Supplied to ESP Program

Species	Equilibrium Constant
$CaC_2O_4{\cdot}H_2O \leftrightarrow Ca(ion) + C_2O_4(ion) + H_2O$	4E-09*
$MnC_2O_4{\cdot}2H_2O \leftrightarrow Mn(ion) + C_2O_4(ion) + 2H_2O$	1.1E-15*
$NiC_2O_4 \leftrightarrow Ni(ion) + C_2O_4(ion)$	4E-10*
$SrC_2O_4{\cdot}H_2O \leftrightarrow Sr(ion) + C_2O_4(ion) + H_2O$	1.6E-07*
$ZnC_2O_4 \leftrightarrow Zn(ion) + C_2O_4(ion)$	2.7E-08*
$Th(C_2O_4)_2 \leftrightarrow Th(ion) + 2C_2O_4(ion)$	1E-22*
$UO_2C_2O_4{\cdot}3H_2O \leftrightarrow UO_2(ion) + C_2O_4(ion) + 3H_2O$	2E-04*
$Fe_2(C_2O_4)_3 \leftrightarrow 2Fe^{III}(ion) + 3C_2O_4(ion)$	**, †

* Dean, 1992

** $D_fH^o = -6845.7$ J/g; $D_fG^o = -5405.398$ J/g; $S^o = -1317.35$ J

† Wagman, et al., 1982

- Zeolite IE-96 was considered insoluble in this modeling and was not included in the modeling calculations; however, where applicable, results were corrected to include the zeolite. This assumption was necessary due to a lack of sufficient thermodynamic data on zeolite IE-96. Without sufficient data, it was not possible to model the zeolite. This assumption was believed to be reasonable, based on the small amount of zeolite present compared to the amount of oxalic acid. If this assumption were incorrect, then the model would under-predict the dissolution materials from the sludge.

- The amounts of uranium and thorium (not included in the experimental sludge-simulant recipe, but included in the model) were calculated based on the ratio of the amounts of uranium and thorium reported to be present in the sludge to the amount of silica in the sludge, and then using these ratios to calculate the amount of uranium and thorium to be added to the sludge simulant. This assumption was made to allow the modeling of uranium and thorium in the sludge. The ratios of these constituents to silica should be accurate, as the silica is largely insoluble at the tank conditions; therefore, if the amount of silica included in the sludge recipe is accurate, then the amounts of uranium and thorium used are accurate.

Assumptions Made to Allow Modeling of the System

Several assumptions were made to allow modeling of the physical system using the ESP computer code. These assumptions are described as follows:

- The experimental contact times (~50 hours) were assumed to be sufficiently long such that equilibrium was reached. This assumption is required due to the unavailability of sufficient kinetic data for all of the principal reactions in the sludge-oxalate system, and it should be reasonable, provided that there are no unexpected kinetic barriers to the dissolution of the waste. Such kinetic barriers should not be present in the simulated waste, but may be present in the actual waste if mineralization has taken place, or if the physical system prevents mixing of the oxalic acid with the sludge.

- The species shown in Appendix N, Table N.1, accurately represent those that would be present in the sludge and in the sludge-oxalate mixture. This assumption is a restatement of the scope limitations of any modeling effort. If a species of effect has not been considered in the model, then the model will not accurately reflect the physical system. For the sludge-oxalate system, it is the judgment of the modelers that all required species have been incorporated. However, if this assumption is incorrect and the species neglected is critical to the results, then the results involving this species will be incorrect, although other model results would not be significantly impacted. In the sludge-oxalate system, the biggest possibility for error in this area is the assumption of the wrong oxidation states for species used in the modeling. If the wrong oxidation state was assumed, then the wrong species would be included. Because the oxidation states assumed were based on the oxidation states present in the simulant, any errors of this type will not affect the comparability of the experimental and modeling results, but will affect their extrapolation to the actual tank system. Based on the fact that oxalic acid is neither a strong oxidizing agent nor a strong reductant, it is assumed that in this system, the oxidation states should not change during the sludge-dissolution process.

- The chemical reactions shown in Appendix N, Table N.2, accurately represent the physical system. This assumption is also a scope-limiting assumption inherent to any modeling effort. However, it should be noted that not all possible reactions must be included but only sufficient reactions to provide a conversion pathway from each form of each species to all other forms including a species. (For example, the reaction to convert calcium oxalate to calcium carbonate does not have to be listed if the reaction to convert calcium oxalate to calcium ion and oxalate ion and the reaction to convert calcium carbonate to calcium ion and carbonate ion are included.) The technique used to generate the reactions will generate sufficient reactions to ensure that all conversion paths are included in the model, but if a species has not been included, no reactions involving that species will be included. It is the judgment of the modelers that based on the simulant recipe and the tank chemistry, the correct species and reactions have been included.

6.1.2 Modeling Process

The modeling approach consisted of creating a sludge waste form by following the experimental procedure used for developing the sludge simulant. The form and amount of each chemical used in the modeling of the sludge are shown in Table 6.2. It should be noted that although in some cases different chemicals were used in the modeling than were used to prepare the simulant, as long as the oxidation states of the compounds added were the same, there is no impact on the modeling results. The species present determines the final system state at equilibrium.

The chemicals shown in Table 6.2 were mixed together and the equilibrium system conditions calculated. Based on this system state, the solid was separated from the liquid and recombined with sufficient supernate to result in a 20 wt% sludge slurry. Eight wt% oxalic acid was then added to this slurry and the system equilibrium state calculated. Calculations for adding 8 wt% oxalic acid up to 100 L/kg were completed.

Table 6.2. Compounds Used in Sludge-Dissolution Model

Compound	Amount Added, g
$Al(NO_3)_3 \cdot 9H_2O$	302.10
$Fe(NO_3)_3 \cdot 9H_2O$	2390.50
$Mn(NO_3)_2$	162.50
$Cr(NO_3)_3 \cdot 9H_2O$	26.78
$Ni(NO_3)_2 \cdot 6H_2O$	28.84
$Pd(NO_3)_3$	3.01
$Rh(NO_3)_3$	1.25
$Ru(NO)(NO_3)_3$	168.60
$Sr(OH)_2 \cdot 8H_2O$	315.45
$Al(OH)_3$	47.40
$NaH_2PO_4 \cdot H_2O$	49.78
SiO_2	29.94
$CaCO_3$	27.59
ZrO_2	39.39
KNO_3	89.16
$NaNO_3$	110.13
$NaNO_2$	52.11
Na_2SO_4	2.30
IE-96 Zeolite	480.00

6.2 Results and Discussion

The sludge simulant was modeled using an equilibrium thermodynamic computer model, the Environmental Simulation Program. Three dissolution cases were modeled: 20, 40, and 80 L 8 wt% oxalic acid/kg sludge. The composition of the modeled sludge (no zeolite present) calculated from the modeling process is shown in Appendix N, Table N.3. The compositions of the solid and liquid phases after oxalic acid addition at 20, 40, and 80 L/kg are shown in Appendix N, Table N.4. Compositions of solid and liquid phases after oxalic acid addition at 60 and 100 L/kg are shown in Table N.5 (also in Appendix N). Results of additional cases are shown graphically in Figure 6.1 through Figure 6.6 for the constituents of main concern: Fe, and the radionuclides U and Th. The gram-mols of these elements (not considering zeolite) as a function of the amount of acid added are demonstrated in Figure 6.1 through Figure 6.3. Figure 6.4 through Figure 6.6 show the percent of the original solid constituent dissolved as a function of the amount of oxalic acid added (corrected for zeolite). Figures showing the gram-mols remaining and the percentages dissolved for the other constituents of the sludge can be found in Appendix O.

The modeling results shown in Figure 6.1 through Figure 6.6 and in Appendix O indicate several items of interest. The first item is that essentially all of the dissolution of species happens at much lower ratios of oxalic acid:sludge than the lowest ratio examined in the laboratory (20 L/kg). In fact, essentially all of the dissolution that happens can be completed

Table 6.3. Comparison of Computer Modeling and Experimental Sludge-Dissolution Data

Constituent	20 L Acid/kg Sludge % Soluble		40 L Acid/kg Sludge % Soluble		80 L Acid/kg Sludge % Soluble	
	Model	Lab*	Model	Lab*	Model	Lab*
Al	100.00	75.66	100.00	78.88	100.00	80.06
Ca	50.83	477.56	100.00	568.33	64.65	490.56
Ce	n/a	17.01	n/a	34.86	n/a	7.71
Cr	100.00	97.95	100.00	87.84	100.00	87.27
Fe	48.97	88.54	100.00	94.75	100.00	92.74
K	100.00	79.67	100.00	86.33	100.00	83.37
Mg	n/a	91.38	n/a	73.20	n/a	35.94
Mn	100.00	82.70	100.00	90.13	100.00	90.02
Na	100.00	83.33	100.00	81.81	100.00	87.53
Ni	14.86	66.20	38.93	77.90	11.80	68.86
Sr	100.00	84.98	100.00	99.89	100.00	96.46
Ti	n/a	80.53	n/a	83.83	n/a	79.53
Zr	77.93	2.57	98.39	1.23	98.39	2.01
U	100.00	n/a	100.00	n/a	100.00	n/a
Th	63.82	n/a	100.00	n/a	100.00	n/a

* Laboratory results are taken from optimum process conditions experiment, fifty hour contact time

with a ratio as low as 10 L oxalic acid/kg sludge. In this region of lower acid:sludge ratios, the chemistry of aluminum is dominant, as Figure 6.7 shows. Initial additions of oxalic acid serve to neutralize any free caustic present in the sludge. Subsequent to this neutralization, the decrease in solution pH causes the aluminum in solution to precipitate and then ultimately redissolve as the pH of the solution is lowered further. As the buffering effect of the aluminum precipitation and redissolution is saturated, other species (e.g., Fe) are dissolved. This buffering effect can be seen clearly by the pH curves shown on any of the figures. The reason for the negative percent dissolved shown in Figure 6.7 is that percent dissolved refers to the amount of Al in solution beyond that which was initially in solution. As oxalic acid is added to the system, the pH drops and Al precipitates out as $Al(OH)_3$. The liquid then has a lower Al concentration than it did initially, hence the negative percent dissolved.

The only oxalate species ever seen to precipitate was calcium oxalate. This species is precipitated after the oxalate reduces the solution pH sufficiently to dissolve the calcium initially present as a solid in the sludge, and then reprecipitates it as an oxalate. This oxalate precipitate then redissolves as the pH of the solution was lowered further at higher acid:sludge ratios. In none of the cases investigated did significant concentrations of oxalate complexes in solution appear to be a significant factor in the solubility of any constituent.

The modeling results are compared with the experimental results in Table 6.3. As the table shows, the modeling results agree fairly well with the laboratory results. The only areas where significant differences are seen are in Al, Na, and Ce. The differences in aluminum and sodium could be explained by the assumption that the zeolite was completely insoluble. Sodium and Al are major constituents of the zeolite and the assumption that they are insoluble dominates the percent-dissolved results. The model predicts that all of the Al, Na, and Fe present in the initial sludge would be dissolved at the 20 L/kg level, and scoping calculations indicate that much larger quantities of Na, Al and Fe (~800 additional grams of iron oxide) would be soluble in the 20 L/kg solution. No solubility limit was determined for Na or Al, as test cases indicated very large solubilities. Thus, if any of the zeolite dissolved or leached Na or Al, it would have dissolved. Based on the experimental results, it is apparent that significant quantities of Na and Al in the zeolite are soluble. Further modeling work in this system should include the effect of zeolite dissolution, either by extrapolation of empirical data or by the use of thermodynamic data.

The results show that essentially all of the constituents associated with the sludge, except for Ce and Zr, are completely soluble in quantities of oxalic acid greater than 20 L/kg of sludge. The solubility of Ce and Zr appears to be relatively unaffected by the quantity of oxalic acid added, and both elements appear to be essentially insoluble in this system. This insolubility is in

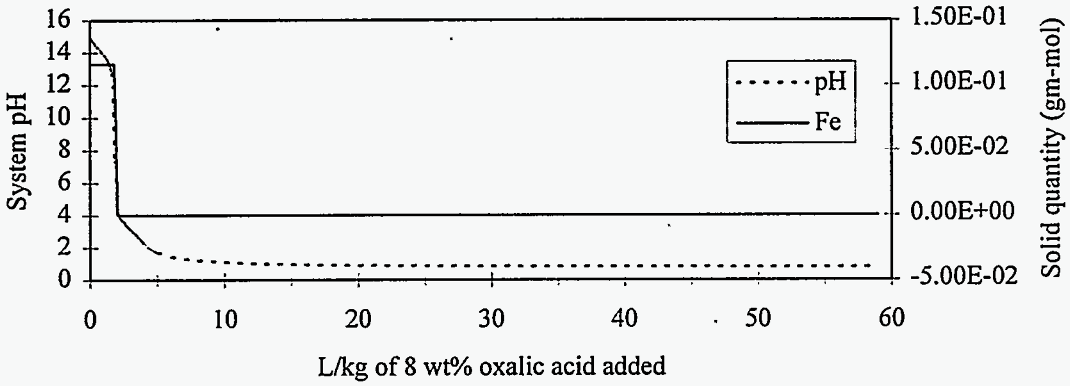

Figure 6.1(a). Iron Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

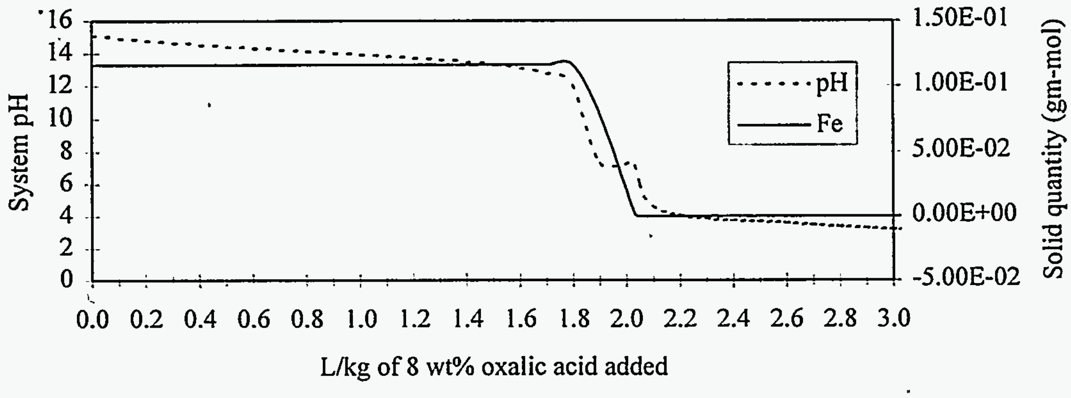

Figure 6.1(b). Iron Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

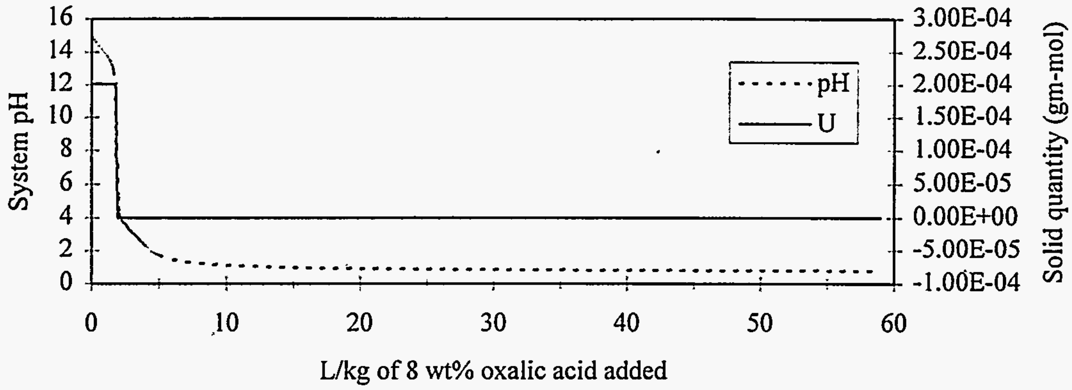

Figure 6.2(a). Uranium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

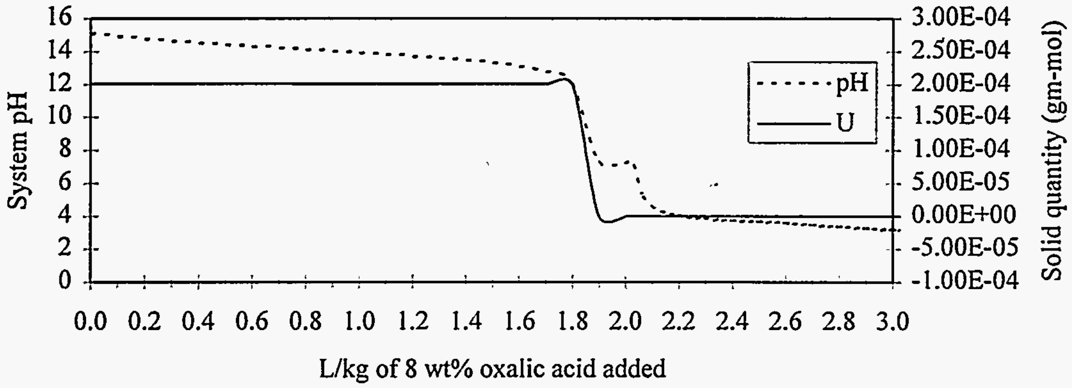

Figure 6.2(b). Uranium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

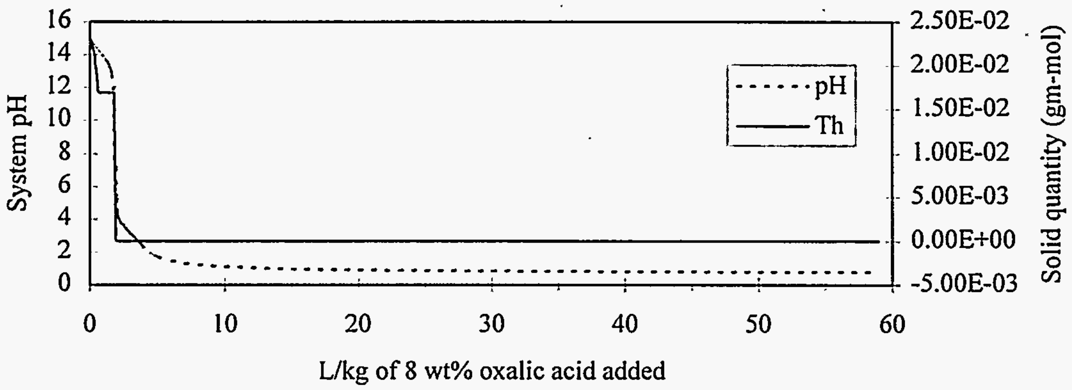

Figure 6.3(a). Thorium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

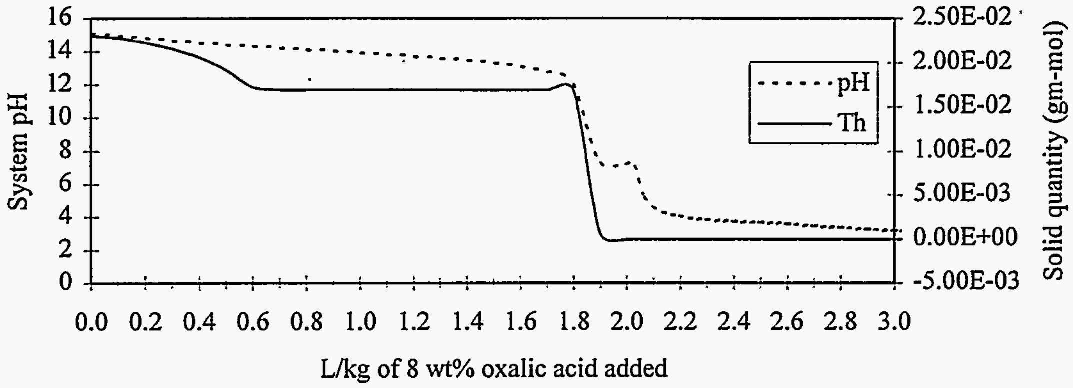

Figure 6.3(b). Thorium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

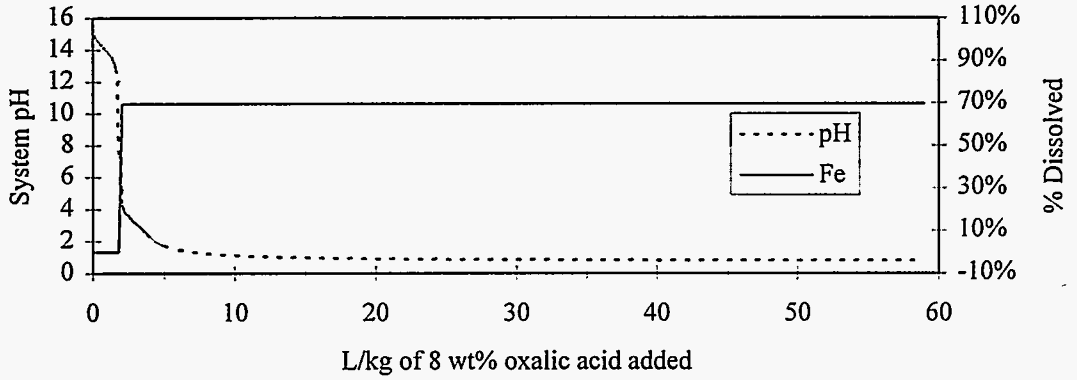

Figure 6.4(a). Percent Modeled Sludge Fe Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

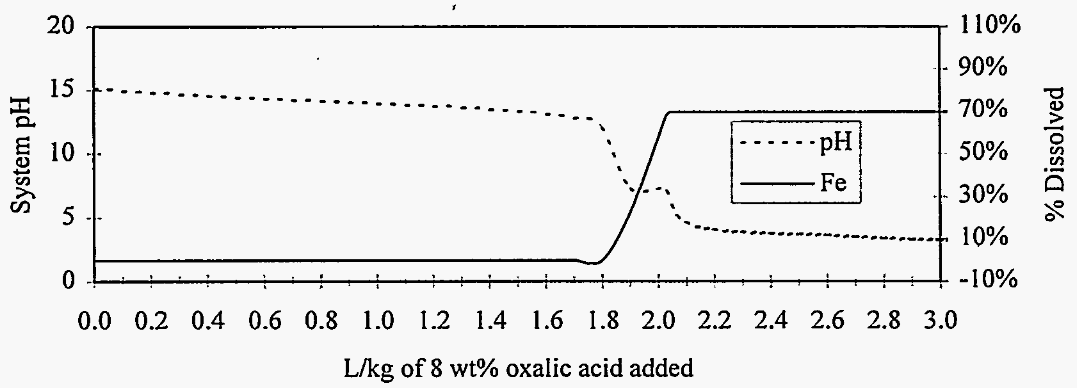

Figure 6.4(b). Percent of Modeled Sludge Fe Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

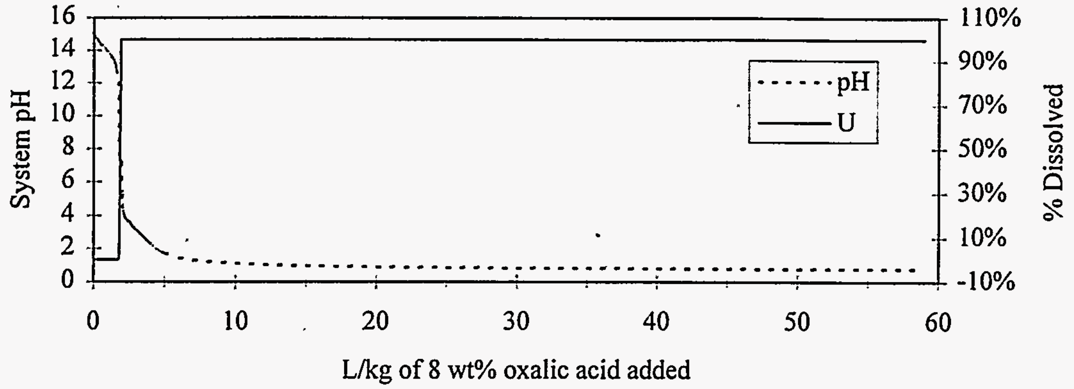

Figure 6.5(a). Percent of Modeled Sludge U Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

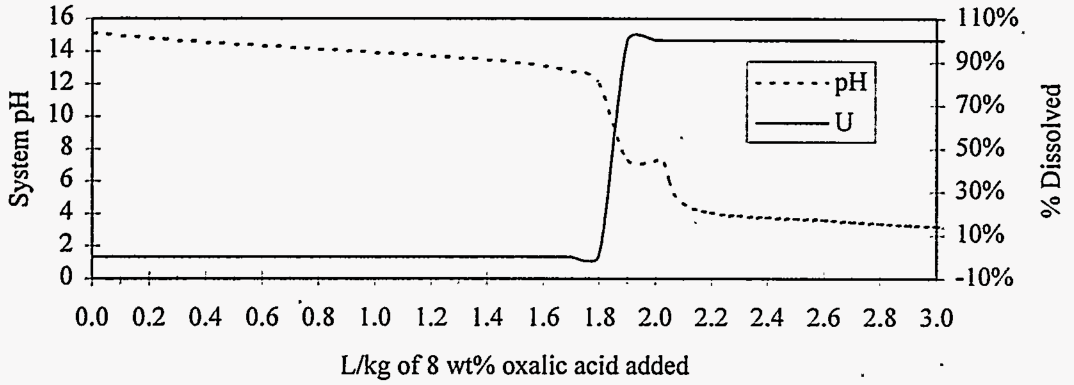

Figure 6.5(b). Percent of Modeled Sludge U Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

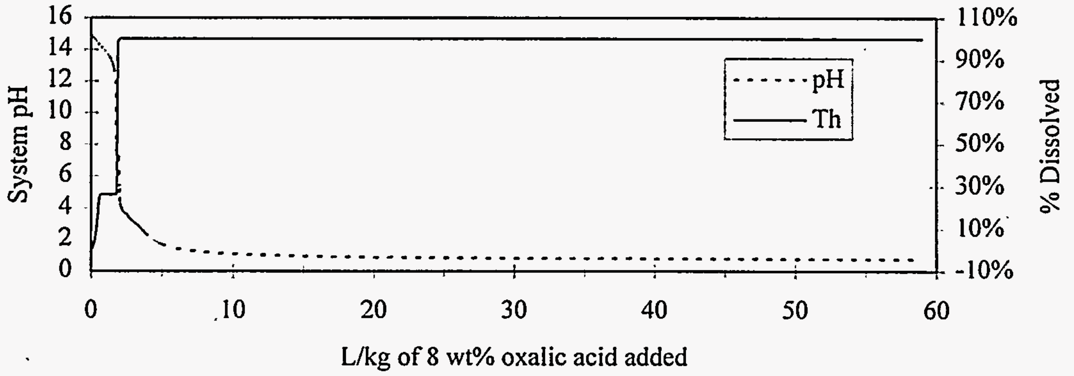

Figure 6.6(a). Percent of Modeled Sludge Th Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

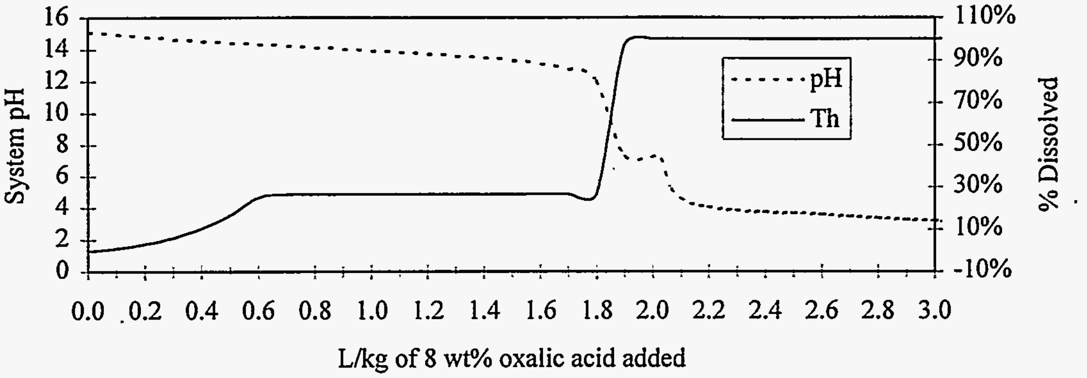

Figure 6.6(b). Percent of Modeled Sludge Th Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

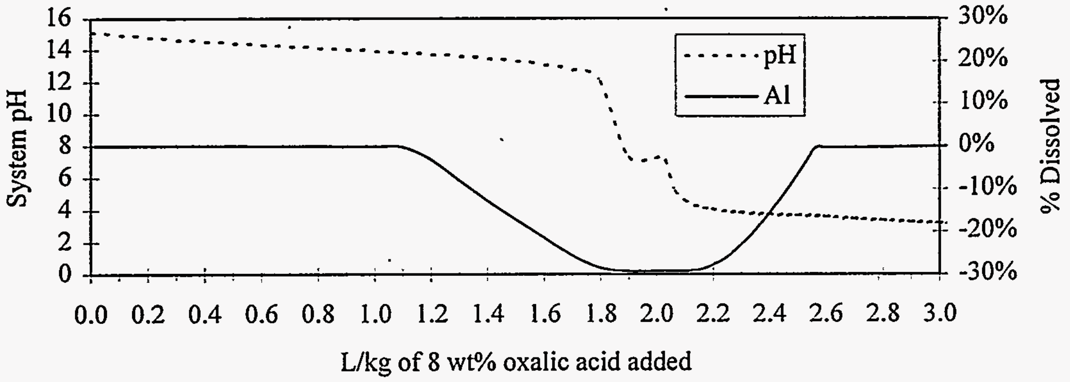

Figure 6.7. Percent of Modeled Sludge Al Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

sharp contrast to the behavior of U and Th, both of which are readily dissolved in this system, and very soluble. This would appear to indicate that Ce is a poor surrogate for U and Th in this system. For U, one possible explanation may involve the fact that U forms oxy-anions upon going into solution (UO_2, UO_2OH, etc.). In the case of Th, no readily apparent explanation of the difference could be postulated.

In summary, the modeling work indicates several important items with respect to the use of oxalic acid for the dissolution of sludge, as follows:

- The oxalic acid-sludge system is not solubility limited for any species investigated except Ce and Zr. This finding is important as it indicates that essentially all of the Fe present in the tank as iron compounds or iron oxides is capable of being dissolved. In addition, if a corresponding redox couple is available, it is possible that Fe from the walls of the tank could be dissolved into the system.

- Essentially all species from the sludge are readily soluble in this system. (It should be noted, however, that this is a thermodynamic model, and therefore does not take into account the effects of kinetics. Although the model predicts that essentially all species are.readily soluble, it does not predict how long it would take for these species to dissolve.)

- If further modeling of this system is to be conducted, additional data for zeolite IE-96 should be obtained to further improve the accuracy of the predictions.

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Appendix A

Calculations

Calculations for Loading Zeolite with Cs

Need to determine how much Cs needs to be loaded into zeolite in order to simulate Cs loaded zeolite in tank 8D-1. The following information was provided in a fax from SC Fahey to KD Wiemers on October 7, 1994.

- total estimated lbs zeolite in tank 8D-1 (at completion of Thorex transfer and third sludge wash) is 154,800 lbs (this is based on 43 cv and 3600 lbs (dry weight) zeolite/cv)
- total estimate of Curies of Cs in tank 8D-1 (at end of third sludge wash) is 5534 kCi Cs
- 5534 kCi / 154,800 lb = 35.75 Ci/lb zeolite

Calculations: goal is to calculate grams Cs required to give an activity level of 35.75 Ci/lb zeolite

$$\text{Activity} = c\lambda N = c\,\frac{-dN}{dt}$$

where c = detection coefficient, assumed to be 1 as measured Ci should have been normalized before it was reported

λ = decay constant = ln 2/ $\tau_{1/2}$

$\tau_{1/2}$ = half life

N = number of atoms

So need to find N, the number of Cs atoms giving rise to the activity.
Assume that all of the activity is from ^{137}Cs (99% of it will be)
Assume also that this represents all of the Cs present in the tank (i.e., no ^{133}Cs, etc.)

1 Ci = 3.700×10^{10} disintegrations/second (by definition)
$\tau_{1/2}$ = 30.17 yrs

$$\lambda = \frac{\ln 2}{30.17\text{ yrs}} = \frac{\ln 2}{(30.17\text{ yrs})(365\text{ days/yr})(24\text{ hrs/day})(3600\text{ s/hr})} = 7.2852 \times 10^{-10}\ \text{s}^{-1}$$

$$\text{Activity} = \frac{(35.75\text{ Ci})\,3.700 \times 10^{10}\text{ dis/s}}{\text{Ci}} = 1.3228 \times 10^{12}\text{ dis/s}$$

Then N can be calculated from

$$\text{Activity} = c\lambda N$$
$$1.3228 \times 10^{12}\text{ dis/s} = (1)(7.2852 \times 10^{-10}\text{s}^{-1})\,(N)$$
$$1.8157 \times 10^{21}\text{ atoms} = N$$

$$N = 1.8157 \times 10^{21}\text{ atoms} \times \frac{1\text{ mole}}{6.0225 \times 10^{23}\text{ atoms}} \times \frac{132.9\text{ g}}{\text{mole}} = 0.401\text{ g }^{133}\text{Cs / lb zeolite}$$

$$= 0.401\text{ g Cs / 0.4525 kg zeolite}$$
$$= \mathbf{0.8862\text{ g Cs / kg zeolite}}$$

To load zeolite on a lab scale

To load 1 kg zeolite IE-96 will require 0.8862 g Cs = 6.666×10^{-3} moles Cs
The Cs source to be used is $CsNO_3$ (formula wt = 194.909 g/mole)
6.666×10^{-3} moles $CsNO_3$ x 194.909 g/mole = **1.30 g $CsNO_3$ / kg zeolite**
when making up zeolite, remember that amounts of zeolite are based on **dry weight.**

Calculation of Conditions Used in Tests 22-29

Calculation of liters of oxalic acid per kg of zeolite required for 3 ft depth in tank 8D-1

This calculation assumes that 5% of the zeolite will remain in the tank for oxalic acid treatment.
Total estimated lbs zeolite in 8D-1 = 154,800 lbs
This is based on a total of 43 column volumes (cv) and 3600 lbs (dry weight) zeolite/cv
(this information comes from a fax from S.C. Fahey to K.D. Wiemers, dated 27 October, 1994)

154,800 lbs = 70,202 kg zeolite

5% of this is 3510 kg. This is a dry weight. Wet zeolite is going to weigh a little more. In our lab experiments, 5.63 g dry weight was equivalent to 5.93 g wet weight, a factor of 1.054 higher. This would give us 3700 kg zeolite wet weight. Since West Valley's estimate of the weight of zeolite is in terms of dry weight, that's what we'll use for this calculation.

Amount of zeolite in 8D-1, assuming 95% transfer = 0.05(70,202) = **3510 kg**

A minimum of 3 feet of liquid must be in 8D-1 in order for the contents to be agitated (M.A.Schiffhauer, conference call between M.A. Schiffhauer, C.King, G.K. Patello, and J.A. Sills, 4/20/95). At 109,000 L/ft, this means we need a minimum of 327,000 L of liquid (assumes that volume occupied by zeolite and rust is negligible in comparison)

With 3510 kg of zeolite in the tank, we have a minimum amount of liquid equaling

$$\frac{327{,}000\ \text{L}}{3510\ \text{kg}} = \mathbf{93\ L/kg}$$

For the laboratory experiments, we will use 100 L oxalic acid/kg zeolite to simulate the tank proportions.

Calculation of minimum contact time required based on pumping rate

The pumping rate into and out of 8D-1 was given as 100 gallons per minute (M.A. Schiffhauer, conference call between M.A. Schiffhauer, C.King, G.K. Patello, and J.A. Sills, 4/20/95).

$$\frac{100\ \text{gallons}}{\text{minute}} * \frac{1\ \text{liter}}{0.264\ \text{gallons}} = 379\ \text{liters/minute}$$

If we want to fill the tank to 3 feet, the minimum depth for agitation, this requires 327,000 liters (~ 93 L/kg). The time to pump in this much liquid, and the times to pump in the amount of liquid required for 80, 40, and 20 L/kg (covered by our previous testing), are shown in the table below:

Table A.1. Times Required to Pump Oxalic Acid into Tank 8D-1 Assuming 100 gal/min

L/kg Zeolite	Total Liters	Time to Pump In	
		Minutes	Hours
100	351,000	926	15.4
93	327,000	862	14.4
80	281,000	741	12.4
40	140,000	369	6.2
20	70,000	185	3.1

These times are just those necessary to pump the liquid *into* the tank. Pumping out will take the same amount of time, and there may be additional time that the liquid is left in the tank to be agitated (or not) with the tank contents.

Amount of Fe_2O_3 needed to simulate rust on bottom of tank and on internal tank structures

At 3 feet liquid level in tank with 1/32" rust there will be 3990 kg Fe_2O_3
At 4 feet liquid level in tank with 1/32" rust there will be 4139 kg Fe_2O_3
(This info comes from a fax from Connie King dated 6/1/95)

Scaled to 5 grams of zeolite:

$$\frac{0.005\text{ kg zeolite}}{3510\text{ kg zeolite}} = \frac{x\text{ kg }Fe_2O_3}{3990\text{ kg }Fe_2O_3} \Longrightarrow x = \frac{0.005(3990\text{ kg})(1000\text{ g/kg})}{3510} \Longrightarrow 5.68\text{ g }Fe_2O_3$$

$$\frac{0.005\text{ kg zeolite}}{3510\text{ kg zeolite}} = \frac{x\text{ kg }Fe_2O_3}{4139\text{ kf }Fe_2O_3} \Longrightarrow x = \frac{0.005(4139\text{ kg})(1000\text{ g/kg})}{3510} \Longrightarrow 5.90\text{ g }Fe_2O_3$$

At 100 L oxalic acid/kg zeolite, the tank level will be 3.22 feet (calculated from 109,000 L/ft)

Interpolation of the amounts above gives us 5.73 g Fe_2O_3 required.

In addition, there is 120 kg Fe_2O_3 loose on the bottom of the tank (5% of the 2400 kg estimated to be present in the form of corrosion products). Scaled to 5 g zeolite:

$$\frac{0.005\text{ kg zeolite}}{3510\text{ kg zeolite}} = \frac{x\text{ kg }Fe_2O_3}{120\text{ kg }Fe_2O_3} \Longrightarrow x = \frac{0.005(120)(1000\text{ g/kg})}{3510} = 0.17\text{ g}$$

Total Fe_2O_3 required for laboratory scale testing = 5.73 g + 0.17 g = **5.90 g Fe_2O_3**

Approximate coupon size needed to simulate internal surface area exposed to oxalic acid

From a fax from Connie King to Gert Patello dated 5-30-95, a three foot depth of oxalic acid in the tank will contact 10,583 ft^2 of tank surface. The volume of acid at a three foot depth is (109,000 L/ft)(3 ft) = 327,000 L.

The laboratory experiments are scaled to 5 grams zeolite.

$$5\text{ g} * \frac{100\text{ L}}{\text{kg}} * \frac{1\text{ kg}}{1000\text{ g}} * \frac{1000\text{ ml}}{\text{L}} * = 500\text{ ml} = 0.5\text{ L oxalic acid}$$

$$\frac{0.5\text{ L}}{327{,}000\text{ L}} = \frac{x}{10{,}583\text{ ft}^2} \implies x = 1.618 \times 10^{-2}\text{ ft}^2$$

$$1.618 \times 10^{-2}\text{ ft}^2 * (12\text{ in/ft})^2 = \mathbf{2.33\ in^2}$$

Amount of Oxalate Depleted by Fe_2O_3

Question: If all of the iron oxide (rust) in tank 8D-1 reacted with oxalic acid to form iron oxalate, how much of the oxalic acid would be consumed?

Assumptions:

1. 5% residual zeolite remains in tank 8D-1; this is 3510 kg.
2. 2400 kg rust in tank 8D-1 (MA Schiffhauer, FAX to KD Wiemers, 9/27/94); assume only 5% of this remains when oxalic acid is added to tank. This is 120 kg.
3. All of the "rust" is in the form of Fe_2O_3 rather than hydrated ferric oxide
4. Tank will be filled to a 3 foot depth. With 5% residual zeolite, this corresponds to 93 L/kg
5. At a 3 foot depth, and assuming that all of the 1/32" of rust on the walls ends up in solution, the maximum amount of Fe_2O_3 in solution would be 3990 kg (in addition to the 120 kg on bottom of tank)

Data:

formula weight oxalic acid = 90.04 g/mole
formula weight Fe_2O_3 = 159.69 g/mole
oxalic acid = $H_2C_2O_4$
iron (III) oxalate = $Fe_2(C_2O_4)_3 \cdot 5H_2O$

Calculations:

1 mole $Fe_2O_3 \rightarrow$ 1 mole $Fe_2(C_2O_4)_3 \cdot 5H_2O$, consuming 3 moles of oxalic acid

$$4110\text{ kg } Fe_2O_3 \times \frac{1000\text{ g}}{\text{kg}} \times \frac{1\text{ mole}}{159.69\text{ g}} = 25{,}737\text{ moles } Fe_2O_3$$

and since each mole of Fe_2O_3 consumes 3 moles oxalic acid, 77,212 moles of oxalic acid would be consumed.

$$77{,}212\text{ moles } H_2C_2O_4 \times \frac{90.04\text{ g}}{\text{mole}} = 6{,}952{,}177\text{ g oxalic acid}$$

The oxalic acid solution is at 8 weight % = 80 g oxalic acid/litre solution (assuming a density of 1)

$$\frac{6{,}952{,}177\text{ g oxalic acid}}{80\text{ g oxalic acid/L sol'n}} = \text{the oxalic acid in 86,902 litres of 8 wt\% solution would be consumed by 4110 kg of rust}$$

So for 93 L/kg

$3510 \text{ kg} \times \frac{93 \text{ L}}{\text{kg}} = 326{,}430$ L of 8 wt% oxalic acid added to the tank

$\frac{86{,}902 \text{ liters depleted}}{326{,}430 \text{ liters added}} \times 100 =$ **26.62% of the oxalate consumed by rust**

Amount of Oxalate Tied up in Ce Oxalate Precipitates

Considering amounts present in the vessel used for sludge-dissolution Test 2 (experiment for determination of optimum process conditions, 40 L 8 wt% oxalic acid/kg sludge, acid added at 25°C, 50 hours total contact time):

Total amount Ce initially present: 58656 ug
Total amount Ce dissolved at T5 (50 hours): 20450 ug
Amount of Ce remaining in solids: 38206 ug

moles Ce in solids: 38206×10^{-6} g = 0.0382 g
molecular weight of Ce is 140.12 g/mol
0.0382 g / 140.12 = 2.726×10^{-4} moles Ce in remaining solids

This implies 1.363×10^{-4} moles of $C_6H_{20}Ce_2O_{22}$ assuming that all of the Ce in the solids precipitated in this form. This would tie up 3 moles of oxalic acid per 2 moles of the cerium oxalate hydroxide precipitate.

At 40 L 8 wt% oxalic acid / kg sludge in the system, we have 0.286 moles of oxalate present.

$(4.09 \times 10^{-4} / 0.286) * 100 = 0.14\%$ of the oxalate would be tied up in the Ce oxalate hydroxide precipitate.

Appendix B

Sources of Experimental Error in Cs-Elution and Sludge-Dissolution Tests

Throughout this report, the term "experimental error" is used. Experimental error is composed of analytical error and other sources of error that may contribute to the size of the range of values in which valid data points may lie. For the experiments reported in this work, possible sources of error include the following:

Analytical error:	±10% for XRF on Cs-loaded zeolite ±15% for ICP-MS measurements of Ti ±20% for ICP-MS measurements of Ca ±10% for ICP-MS on other ions ±10% for ICP-ES on all ions
Weighing errors:	balances that weigh to 0.1 g have ±0.1 g error balances that weigh to 0.01 g have ±0.01 g error
Operator errors:	Samples were drawn and weighed by three different test operators. It is not possible to quantify operator error, some possible errors include transposing numbers when weighing, spilling a small amount of solution when transferring from reaction vessel to sample bottle, and (due to the large number of samples generated in each experiment) putting samples in the wrong sample bottle.

Table B.1 shows a sample calculation for one of the Cs-elution experiments in which possible sources of error are taken into account during the calculation. Three calculations are performed, one taking no error into account, one in which a minimum value is calculated by always subtracting the error of x% from the value, and one in which a maximum value is calculated by always adding the error of x% to the value. This table shows that the analytical error does not propagate throughout the calculation, and that, combined with possible error from the balances, the Cs-elution calculations still yield values that are within ±10% of the analytical values.

Table B.1. Sample Error Calculation for Cs-Elution Test

	Test 21	error	min error	max error	source
Time 0 : initial conditions					
initial amount of zeolite in reaction vessel, g	7.77	0.01	7.76	7.78	error on balance assumed +/- 0.01
F factor of zeoltie (= dry weight/ wet weight)	0.724		0.6835	0.7668	
amount of dry zeolite used, g	5.63	0.01	5.62	5.64	error on balance assumed +/- 0.01
ug Cs per gram dry zeolite	957	10%	861.3	1052.7	analytical error given as +/-10%
total amount Cs initially present in vessel, ug	5384		4837	5932	
weight of oxalic acid initially present, g	229.3	0.1	229.2	229.4	error on balance assumed +/- 0.1
weight of NaOH (pH ~ 11) initially present, g	2.76	0.01	2.75	2.77	error on balance assumed +/- 0.01
weight of Fe_2O_3 present, g	0		0	0	
total initial material weight, g	239.83		239.71	239.95	
weight loss per hour, g	0				
final vessel weight	2297.2	0.1	2297.1	2297.3	error on balance assumed +/- 0.1
empty vessel weight	2088.9	0.1	2088.8	2089.0	error on balance assumed +/- 0.1
total finish weight	208.3		208.3	208.3	

Table B.1, cont.

	Test 21	error	min error	max error	source
Time 1 = 2 hours	2		2	2	
weight of material present at T1, g	239.83		239.71	239.95	
weight of sample removed at T1, g	6.31	0.01	6.30	6.32	error on balance assumed +/- 0.1
ug Cs per gram sample -- run 1	17.3	10%	15.6	19.0	analytical error given as +/-10%
ug Cs per gram sample -- run 2	19.1	10%	17.2	21.0	analytical error given as +/-10%
average ug Cs per gram sample	18.2		16.4	20.0	
amount Cs in solution at T1, ug	4365		3926	4804	
percentage Cs dissolved at T1	**81.08**		**81.18**	**80.97**	
Time 2 = 4 hours	4		4	4	
weight of material present at T2, g	233.52		233.41	233.63	
weight of sample removed at T2, g	5.29	0.01	5.28	5.30	error on balance assumed +/- 0.01
ug Cs per gram sample -- run 1	20.2	10%	18.2	22.2	analytical error given as +/-10%
ug Cs per gram sample -- run 2	19.9	10%	17.9	21.9	analytical error given as +/-10%
average ug Cs per gram sample	20.1		18.0	22.1	
amount of Cs in solution at T2, ug	4682		4212	5153	
amount of Cs removed during T1 sampling, ug	115		103	127	
total amount of Cs dissolved, ug	4797		4315	5279	
percentage of Cs dissolved at T2	**89.10**		**89.22**	**88.99**	
Time 3 = 20 hours	20		22	23	
weight of material present at T3, g	228.23		228.13	228.33	
weight of sample removed at T3, g	5.89	0.01	5.88	5.90	error on balance assumed +/- 0.01
ug Cs per gram sample -- run 1	20.4	10%	18.4	22.4	analytical error given as +/-10%
ug Cs per gram sample -- run 2	20.0	10%	18.0	22.0	analytical error given as +/-10%
average ug Cs per gram sample	20.2		18.18	22.22	
amount of Cs in solution at T3, ug	4610		4147	5073	
amount of Cs removed during T1 sampling, ug	115		103	127	
amount of Cs removed during T2 sampling, ug	106		95	117	
total amount of Cs removed during sampling, ug	221		198	243	
total amount of Cs dissolved at T3, ug	4831		4346	5317	
percentage of Cs dissolved at T3	**89.74**		**89.85**	**89.62**	
Time 4 = 28 hours	28		28	28	
weight of material present at T4, g	222.34		222.25	222.43	
weight of sample removed at T4, g	5.35	0.01	5.34	5.36	error on balance assumed +/- 0.01
ug Cs per gram sample -- run 1	20.5	10%	18.5	22.6	analytical error given as +/-10%
ug Cs per gram sample -- run 2	22.0	10%	19.8	24.2	analytical error given as +/-10%
average ug Cs per gram sample	21.3		19.1	23.4	
amount of Cs in solution at T4, ug	4725		4251	5199	
amount of Cs removed during T1 sampling, ug	115		103	127	
amount of Cs removed during T2 sampling, ug	106		95	117	
amount of Cs removed during T3 sampling, ug	119		107	131	
total amount of Cs removed during sampling, ug	340		305	375	
total amount of Cs dissolved at T4, ug	5065		4556	5574	
percentage of Cs dissolved at T4	**94.08**		**94.20**	**93.95**	

Table B.1, cont.

	Test 21	error	min error	max error	source
Time 5 = 50 hours	50		50	50	
weight of material present at T5, g	216.99		216.91	217.07	
weight of sample removed at T5, g	6.56	0.01	6.55	6.57	error on balance assumed +/- 0.01
ug Cs per gram sample -- run 1	19.3	10%	17.4	21.2	analytical error given as +/-10%
ug Cs per gram sample -- run 2	21.8	10%	19.6	24.0	analytical error given as +/-10%
average ug Cs per gram sample	20.6		18.5	22.6	
amount of Cs in solution at T5, ug	4459		4012	4907	
amount of Cs removed during T1 sampling, ug	115		103	127	
amount of Cs removed during T2 sampling, ug	106		95	117	
amount of Cs removed during T3 sampling, ug	119		107	131	
amount of Cs removed during T4 sampling, ug	114		102	125	
total amount of Cs removed during sampling, ug	454		407	500	
total amount of Cs dissolved at T5, ug	4913		4419	5407	Note values are within ±10% of 4913
percentage of Cs dissolved at T5	**91.25**		**91.37**	**91.14**	

Appendix C

Cs-Elution and Sludge-Dissolution Test Parameters

Table C.1. Experimental Parameters for Cs-Elution Tests

test #	concentration	L/kg	add. T	bath T	agitation?	# contacts	contact time	other conditions	experiment
1	8 wt%	20	25	50	y	1	50 hrs		optimum process
2	8 wt%	40	25	50	y	1	50 hrs		optimum process
3	8 wt%	80	25	50	y	1	50 hrs		optimum process
4	8 wt%	20	80	50	y	1	50 hrs		optimum process
5	8 wt%	40	80	50	y	1	50 hrs		optimum process
6	8 wt%	80	80	50	y	1	50 hrs		optimum process
7	8 wt%	40	25	50	y	1	50 hrs		repeatability
8	8 wt%	40	25	50	y	1	50 hrs		repeatability
9	8 wt%	40	25	50	y	1	50 hrs		repeatability
10	8 wt%	40	25	50	y	1	50 hrs		repeatability
11	8 wt%	40	25	50	y	1	11 days		long term
12	8 wt%	20	25	70	y	1	11 days		long term
13	8 wt%	40	25	50	y	3	6 hrs	3-2 hr contacts, no rinse between contacts	multiple contacts
14	8 wt%	40	25	50	y	3	6 hrs	3-2 hr contacts, rinse between contacts	multiple contacts
15	8 wt%	40	25	50	y	3	6 hrs	3-2 hr contacts, no rinse between contacts	multiple contacts
16	8 wt%	40	25	50	y	3	6 hrs	3-2 hr contacts, rinse between contacts	multiple contacts
17	8 wt%	40	25	50	y	1	50 hrs	Fe_2O_3 added	condition modifications
18	8 wt%	40	25	50	y	1	50 hrs	steel coupons added	condition modifications
19	8 wt%	40	25	50	n	1	50 hrs	(no agitation)	condition modifications
20	8 wt%	40	25	25	y	1	50 hrs	(25°C bath)	condition modifications
21	8 wt%	40	25	50	y	1	50 hrs	(control)	condition modifications
22	4 wt%	100	25	50	y	3	141 hrs	control	corrosion/concentration
23	4 wt%	100	25	50	y	3	141 hrs	Fe_2O_3 added	corrosion/concentration
24	4 wt%	100	25	50	y	3	141 hrs	steel coupons added	corrosion/concentration
25	4 wt%	100	25	50	y	3	141 hrs	steel coupons, no zeolite	corrosion/concentration
26	1 wt%	100	25	50	y	3	141 hrs	control	corrosion/concentration
27	1 wt%	100	25	50	y	3	141 hrs	Fe_2O_3 added	corrosion/concentration
28	1 wt%	100	25	50	y	3	141 hrs	steel coupons added	corrosion/concentration
29	1 wt%	100	25	50	y	3	141 hrs	steel coupons, no zeolite	corrosion/concentration

Table C.2. Experimental Parameters for Sludge-Dissolution Tests

test #	concentration	L/kg	add. T	bath T	agitation?	# contacts	contact time	other conditions	experiment
1	8 wt%	20	25	50	y	1	50 hrs		optimum process
2	8 wt%	40	25	50	y	1	50 hrs		optimum process
3	8 wt%	80	25	50	y	1	50 hrs		optimum process
4	8 wt%	20	80	50	y	1	50 hrs		optimum process
5	8 wt%	40	80	50	y	1	50 hrs		optimum process
6	8 wt%	80	80	50	y	1	50 hrs		optimum process
7	8 wt%	40	25	50	y	1	50 hrs		repeatability
8	8 wt%	40	25	50	y	1	50 hrs		repeatability
9	8 wt%	40	25	50	y	1	11 days		long term
10	8 wt%	40	25	50	y	3	6 hrs	acid removed w/ each contact	multiple contacts
11	8 wt%	40	25	50	y	3	6 hrs	acid added w/ each contact	multiple contacts
12	8 wt%	40	25	50	y	1	50 hrs	Fe_2O_3 added	condition modifications
13	8 wt%	40	25	50	y	1	50 hrs	steel coupons added	condition modifications
14	8 wt%	40	25	50	n	1	50 hrs	(no agitation)	condition modifications
15	8 wt%	40	25	25	y	1	50 hrs	(25°C bath)	condition modifications
16	8 wt%	40	25	50	y	1	50 hrs	(control)	condition modifications

Appendix D

Temperature and pH Data Collected During Cs-Elution Experiments

Table D.1. Temperature and pH Data Collected During Determination of Optimum Process Conditions for Cs-Elution Experiment

	Test 1		Test 2		Test 3	
	20 L/kg, 25°C		40 L/kg, 25°C		80 L/kg, 25°C	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)
NaOH addition	10.729	--	10.776	--	10.776	--
8 wt% oxalic acid addition	1.272	24.0	0.778	23.4	0.791	23.4
pH at bath temperature	0.898	50.0	0.735	45.2	0.797	48.7
2-hour sampling	0.880	45.8	0.759	50.1	0.797	48.7
4-hour sampling	0.912	49.2	0.827	49.4	0.860	53.4
20-hour sampling	0.750	37.3	0.739	49.4	0.832	48.4
28-hour sampling	0.579	48.2	0.682	49.5	0.706	49.5
50-hour sampling	0.120	48.5	0.602	47.1	0.668	51.0

	Test 4		Test 5		Test 6	
	20 L/kg, 80°C		40 L/kg, 80°C		80 L/kg, 80°C	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)
NaOH addition	10.776	--	10.729	--	10.776	--
8 wt% oxalic acid addition	0.249	79.6	0.953	80.0	0.833	80.7
pH at bath temperature	0.732	45.4	0.841	50.0	0.778	50.9
2-hour sampling	0.775	48.0	0.946	50.3	0.778	50.9
4-hour sampling	0.821	47.5	0.846	50.6	0.846	48.1
20-hour sampling	0.732	45.1	0.972	49.2	0.834	49.3
28-hour sampling	0.716	49.1	0.630	50.0	0.724	50.1
50-hour sampling	0.650	48.9	0.202	49.1	0.718	49.3

Table D.2. Temperature and pH Data Collected During Cs-Elution Repeatability Experiment

	Test 7		Test 8		Test 9		Test 10	
	40 L/kg, 25°C		40 L/kg, 25°C		40 L/kg, 25°C		40 L/kg, 25°C	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
NaOH addition	10.729	--	10.729	--	10.729	--	10.729	--
8 wt% oxalic acid addition	1.037	24.0	1.070	24.0	1.016	24.0	0.953	24.0
pH at bath temperature	0.886	50.0	0.827	50.0	0.946	50.0	0.789	50.0
2-hour sampling	0.886	51.3	0.896	49.0	0.800	49.2	0.977	47.4
4-hour sampling	0.904	50.2	0.868	49.3	0.879	47.7	0.837	49.3
20-hour sampling	1.045	50.5	1.069	49.4	1.049	53.1	1.035	49.3
28-hour sampling	0.713	49.9	0.622	50.2	0.624	48.5	0.631	48.7
50-hour sampling	0.222	50.0	0.239	50.7	0.221	50.0	0.221	49.0

Table D.3. Temperature and pH Data Collected During Long-Term Cs-Elution Experiment

	Test 11		Test 12	
	40 L/kg, 50°C Bath		20 L/kg, 70°C Bath	
Event	pH	T (°C)	pH	T (°C)
NaOH addition	12.319	--	11.675	--
8 wt% oxalic acid addition	0.942	23.0	--	25.0
pH at bath temperature	0.525	50.0	1.035	70.0
2-hour sampling	0.651	51.1	0.900	70.5
4-hour sampling	0.684	50.0	0.988	67.8
1 day sampling	0.745	49.2	0.940	65.9
4 days sampling	0.838	50.7	1.930	67.1
7 days sampling	0.869	49.8	0.289	66.4
11 days sampling	1.212	49.7	0.777	66.9

Table D.4. Temperature and pH Data Collected During Multiple-Contact Cs-Elution Experiment

	Test 13		Test 14		Test 15		Test 16	
	No Rinse		Rinse		No Rinse		Rinse	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
NaOH addition	11.614	--	11.614	---	11.699	--	11.699	--
8 wt% oxalic acid additio	1.674	23.0	1.811	23.0	1.167	24.0	1.004	24.0
pH at bath temperature	1.149	50.0	1.233	50.0	0.948	50.0	0.928	50.0
End of contact 1	1.134	51.4	1.128	55.2	0.977	48.6	0.918	50.4
Oxalic acid addition	1.031	25.0	1.147	25.0	0.998	24.0	1.016	24.0
pH at bath temperature	1.046	50.0	1.145	50.0	0.913	50.0	0.954	50.0
End of contact 2	1.141	48.1	1.160	49.9	0.950	47.5	0.934	47.6
Oxalic acid addition	1.183	25.0	1.257	25.0	1.037	24.0	1.017	24.0
pH at bath temperature	1.061	50.0	1.178	50.0	0.935	50.0	0.969	50.0
End of contact 3	1.155	48.1	1.169	47.7	0.889	48.6	0.923	49.4

Table D.5. Temperature and pH Data Collected During Experiment to Assess the Effects of Condition Modifications on Cs Elution

	Test 17		Test 18		Test 19	
	Fe_2O_3		Coupons		No Stir	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)
NaOH addition	11.454		11.454		11.454	
8 wt% oxalic acid addition	1.326	24.0	1.295	24.0	1.288	24.0
pH at bath temperature	1.123	50.0	1.254	50.0	1.308	50.0
2-hour sampling	1.239	48.2	1.257	50.4	1.217	49.6
4-hour sampling	1.184	48.6	1.278	47.6	1.381	49.5
20-hour sampling	1.007	47.9	1.083	47.0	1.046	50.0
28-hour sampling	1.070	48.4	1.144	46.8	1.152	49.7
50-hour sampling	0.984	47.9	1.032	48.6	1.043	51.1

	Test 20		Test 21	
	25°C Bath		Control	
Event	pH	T (°C)	pH	T (°C)
NaOH addition	11.542		11.454	
8 wt% oxalic acid addition	1.116	24.0	1.238	24.0
pH at bath temperature			1.257	50.0
2-hour sampling	1.040	23.8	1.294	48.4
4-hour sampling	1.031	23.6	1.266	49.1
20-hour sampling	1.158	24.0	1.067	48.4
28-hour sampling	1.197	23.0	1.114	49.0
50-hour sampling	1.045	24.1	1.064	48.3

Table D.6. Temperature and pH Data Collected During Experiment to Determine the Effects of Varying Oxalic Acid Concentration on Cs Elution

	Test 22 4%, Control		Test 23 4%, Fe_2O_3		Test 24 4%, Coupon		Test 25 4%, No Zeolite	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
Contact 1, initial	1.175	22.0	1.298	22.0	1.092	22.0	1.023	22.0
At temperature	0.980	48.0	0.993	48.0	1.258	49.0	0.978	49.0
Contact 1, T1 = 2hours	0.927	49.1	1.022	48.1	0.963	47.3	0.946	47.9
Contact 1, T2 = 4 hours	0.899	49.2	1.233	49.3	0.954	47.5	0.932	48.2
Contact 1, T3 = 20 hours	1.150	48.6	1.094	50.7	0.919	49.8	0.896	48.5
Contact 1, T4 = 28 hours	0.969	49.4	0.957	50.2	0.891	47.7	0.948	47.5
Contact 1, T5 = 47 hours	1.016	49.3	0.936	50.3	0.963	49.0	1.004	48.0
Contact 2, initial	1.036	24.0	1.056	24.0	1.064	23.0	1.046	23.0
At temperature	1.019	48.0	0.988	49.0	0.956	51.0	0.985	48.0
Contact 2, T1 = 2 hours	1.015	50.1	1.027	49.3	0.973	48.9	0.957	48.6
Contact 2, T2 = 4 hours	1.006	48.8	0.999	49.6	1.048	49.7	0.997	48.0
Contact 2, T3 = 20 hours	1.035	49.3	0.987	49.0	1.081	48.1	1.095	48.0
Contact 2, T4 = 28 hours	0.966	49.2	0.926	48.7	1.070	48.3	1.084	47.6
Contact 2, T5 = 47 hours	0.949	49.9	0.851	49.4	1.117	48.8	1.143	48.0
Contact 3, initial	0.972	22.0	0.990	22.0	1.266	23.0	1.240	23.0
At temperature	0.677	49.0	1.015	54.0	1.122	49.0	1.161	47.0
Contact 3, T1 = 2 hours	0.914	47.0	0.975	50.1	1.120	48.2	1.174	47.2
Contact 3, T2 = 4 hours	0.933	49.3	0.937	49.4	1.237	47.5	1.180	48.3
Contact 3, T3 = 20 hours	0.932	47.4	0.930	47.9	1.055	48.2	1.049	47.7
Contact 3, T4 = 28 hours	0.933	48.7	0.958	48.1	1.016	49.1	1.065	49.1
Contact 3, T5 = 47 hours	0.919	48.3	0.889	48.4	1.078	47.7	1.010	49.0

Table D.6, continued

	Test 26		Test 27		Test 28		Test 29	
	1%, Control		1%, Fe_2O_3		1%, Coupon		1%, No Zeolite	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
Contact 1, initial	1.605	22.0	1.798	22.0	1.308	22.0	1.313	22.0
At temperature	1.491	50.0	2.282	51.0	1.478	51.0	1.295	53.0
Contact 1, T1 = 2 hours	1.384	49.7	1.563	49.0	1.323	50.3	1.269	51.2
Contact 1, T2 = 4 hours	1.320	51.3	1.782	50.6	1.555	47.9	1.264	47.6
Contact 1, T3 = 20 hours	1.562	51.3	1.509	50.1	1.352	49.4	1.301	46.3
Contact 1, T4 = 28 hours	1.348	49.2	1.457	49.4	1.340	49.3	1.255	47.0
Contact 1, T5 = 47 hours	1.449	51.2	1.525	50.1	1.444	48.2	1.307	47.1
Contact 2, initial	1.308	24.0	1.286	24.0	1.296	23.0	1.314	23.0
At temperature	1.295	48.0	1.303	48.0	1.269	49.0	1.297	51.0
Contact 2, T1 = 2 hours	1.313	47.9	1.274	49.9	1.285	49.4	1.283	53.1
Contact 2, T2 = 4 hours	1.312	50.8	1.280	50.6	1.287	48.6	1.298	48.5
Contact 2, T3 = 20 hours	1.328	50.7	1.256	50.1	1.392	48.8	1.466	48.1
Contact 2, T4 = 28 hours	1.281	47.7	1.236	49.7	1.363	48.3	1.391	49.0
Contact 2, T5 = 47 hours	1.231	48.2	1.232	50.0	1.435	48.9	1.450	46.5
Contact 3, initial	1.263	22.0	1.204	22.0	1.468	23.0	1.474	23.0
At temperature	1.245	51.0	1.303	52.0	1.410	48.0	1.461	50.0
Contact 3, T1 = 2 hours	1.257	51.5	1.210	46.4	1.442	48.3	1.463	47.8
Contact 3, T2 = 4 hours	1.282	49.8	1.204	48.0	1.527	48.2	1.473	47.7
Contact 3, T3 = 20 hours	1.250	46.4	1.160	48.4	1.353	48.7	1.390	47.8
Contact 3, T4 = 28 hours	1.300	47.7	1.200	48.0	1.366	48.4	1.389	48.4
Contact 3, T5 = 47 hours	1.232	47.1	1.203	48.0	1.336	48.7	1.404	47.9

Appendix E

Sample Calculations and Formulas Used in Determination of Percent Cs Eluted in Cs-Elution Experiments

Table E.1. Sample Calculations and Formulas Used in Determination of Percent Cs Eluted in Experiment to Determine Optimum Process Conditions for Cs Elution, Repeatability Experiment, and Long-Term Experiment

	A	B	C
1			
2		Test 1	source of number
3	**Time 0 : initial conditions**		
4	initial amount of zeolite in reaction vessel, g	5.93	measured in laboratory
5	F factor of zeoltie (= dry weight/ wet weight)	0.724	measured in laboratory
6	amount of dry zeolite used, g	4.29	=B4*B5
7	ug Cs per gram dry zeolite	957	XRF measurment
8	total amount Cs initially present in vessel, ug	4109	=B6*B7
9	weight of oxalic acid initially present, g	114.6	measured in laboratory
10	weight of NaOH (pH ~ 11) initially present, g	2.75	measured in laboratory
11	total initial material weight, g	123.28	=B4+B9+B10
12	weight loss per hour, g	0.0	not measured in this test
13	final vessel weight	0.0	not measured in this test
14	empty vessel weight	0.0	not measured in this test
15	total finish weight	0.0	=B13-B14
16	**Time 1 = 2 hours**	2	measured in laboratory
17	weight of material present at T1, g	123.28	=B11-(B16*B12)
18	weight of sample removed at T1, g	6.93	measured in laboratory
19	ug Cs per gram sample -- run 1	20.1	ICP-MS measurement
20	ug Cs per gram sample -- run 2		not run in duplicate
21	average ug Cs per gram sample	20.1	=B19
22	amount Cs in solution at T1, ug	2478	=B17*B21
23	**percentage Cs eluted at T1**	**60.3**	**=(B22/B8)*100**
24	**Time 2 = 4 hours**	4	measured in laboratory
25	weight of material present at T2, g	116.35	=B17-B18-(B24-B16)*B12
26	weight of sample removed at T2, g	6.35	measured in laboratory
27	ug Cs per gram sample -- run 1	19.1	ICP-MS measurement
28	ug Cs per gram sample -- run 2		not run in duplicate
29	average ug Cs per gram sample	19.1	=B27
30	amount of Cs in solution at T2, ug	2222	=B25*B29
31	amount of Cs removed during T1 sampling, ug	139	=B18*B21
32	total amount of Cs eluted, ug	2362	=B30+B31
33	**percentage of Cs eluted at T2**	**57.5**	**=(B32/B8)*100**
34	**Time 3 = 20 hours**	20	measured in laboratory
35	weight of material present at T3, g	110.00	=B25-B26-(B34-B24)*B12
36	weight of sample removed at T3, g	7.36	measured in laboratory
37	ug Cs per gram sample -- run 1	23.3	ICP-MS measurement
38	ug Cs per gram sample -- run 2		not run in duplicate
39	average ug Cs per gram sample	23.3	=B37
40	amount of Cs in solution at T3, ug	2563	=B35*B39

Table E.1, continued

	A	B	C
41	amount of Cs removed during T1 sampling, ug	139	=B31
42	amount of Cs removed during T2 sampling, ug	121	=B26*B29
43	total amount of Cs removed during sampling, ug	261	=SUM(B41:B42)
44	total amount of Cs eluted at T3, ug	2824	=B40+B43
45	**percentage of Cs eluted at T3**	**68.7**	**=(B44/B8)*100**
46	**Time 4 = 28 hours**	28	measured in laboratory
47	weight of material present at T4, g	102.64	=B35-B36-(B46-B34)*B12
48	weight of sample removed at T4, g	6.02	measured in laboratory
49	ug Cs per gram sample -- run 1	22.2	ICP-MS measurement
50	ug Cs per gram sample -- run 2		not run in duplicate
51	average ug Cs per gram sample	22.2	=B49
52	amount of Cs in solution at T4, ug	2279	=B47*B51
53	amount of Cs removed during T1 sampling, ug	139	=B31
54	amount of Cs removed during T2 sampling, ug	121	=B42
55	amount of Cs removed during T3 sampling, ug	171	=B36*B39
56	total amount of Cs removed during sampling, ug	432	=SUM(B53:B55)
57	total amount of Cs eluted at T4, ug	2711	=B52+B56
58	**percentage of Cs eluted at T4**	**66.0**	**=(B57/B8)*100**
59	**Time 5 = 50 hours**	50	measured in laboratory
60	weight of material present at T5, g	96.62	=B47-B48-(B59-B46)*B12
61	weight of sample removed at T5, g	6.69	measured in laboratory
62	ug Cs per gram sample -- run 1	24.1	ICP-MS measurement
63	ug Cs per gram sample -- run 2		not run in duplicate
64	average ug Cs per gram sample	24.1	=B62
65	amount of Cs in solution at T5, ug	2329	=B60*B64
66	amount of Cs removed during T1 sampling, ug	139	=B31
67	amount of Cs removed during T2 sampling, ug	121	=B42
68	amount of Cs removed during T3 sampling, ug	171	=B55
69	amount of Cs removed during T4 sampling, ug	134	=B48*B51
70	total amount of Cs removed during sampling, ug	566	=SUM(B66:B69)
71	total amount of Cs eluted at T5, ug	2894	=B65+B70
72	**percentage of Cs eluted at T5**	**70.4**	**=(B71/B8)*100**
73	**percent error**	**10%**	**given by analytical**
74	density of oxalic acid	1.02	measured in laboratory
75	actual liters per kg	26.2	=(B9/B74)/B6

Table E.2. Sample Calculations and Formulas Used in Determination of Percent Cs Eluted in Multiple-Contact Cs-Elution Experiment

	A	B	C	D	E
1					
2		Test 13		Test 14	
3	**Time 0 : initial conditions**				
4	initial amount of zeolite in reaction vessel, g	7.77	measured in laboratory	7.77	measured in laboratory
5	F factor of zeolite (= dry weight/wet weight)	0.724	measured in laboratory	0.724	measured in laboratory
6	amount of dry zeolite used, g	5.63	=B4*B5	5.63	=D4*D5
7	ug Cs per gram dry zeolite	957	XRF measurment	957	XRF measurement
8	total amount Cs initially present in vessel, ug	5384	=B6*B7	5384	=D6*D7
9	weight of oxalic acid initially present, g	229.3	measured in laboratory	229.3	measured in laboratory
10	weight of NaOH (pH ~ 11) present, g	2.76	measured in laboratory	2.77	measured in laboratory
11	total initial material weight, g	239.83	=B4+B9+B10	239.84	=D4+D9+D10
12	weight of empty vessel	2001.1	measured in laboratory	2054.00	measured in laboratory
13	weight of loaded vessel	2240.93	=B11+B12	2293.84	=D11+D12
14	**Contact 1**				
15	amount of liquid present at T1 sampling	239.8	=B11	239.8	=D11
16	ug Cs per gram sample -- run 1	19.8	ICP-MS measurement	20.1	ICP-MS measurement
17	ug Cs per gram sample -- run 2		not run in duplicate		not run in duplicate
18	average ug Cs per gram sample	19.8	=B16	20.1	=D16
19	amount Cs in solution at T1, ug	4749	=B15*B18	4821	=D15*D18
20	**percentage Cs eluted at T1**	**88.21**	**=(B19/B8)*100**	**89.55**	**=(D19/D8)*100**
21	**Rinse 1**				
22	weight of assembly after removal of supernate	2012.7	measured in laboratory	2065.9	measured in laboratory
23	material remaining in vessel after contact 1	11.6	=B22-B12	11.9	=D22-D12
24	amount of deionized rinse water added	0.00	no rinse performed	7.76	measured in laboratory
25	amount of material removed after rinse	0.00	no rinse performed	6.76	measured in laboratory
26	material remaining in vessel after rinse 1	11.60	=B23	12.90	=D23+D24-D25
27	ug Cs per gram sample -- run 1	0.00	no rinse	6.82	ICP-MS measurement
28	ug Cs per gram sample -- run 2		no rinse	6.66	ICP-MS measurement
29	average ug Cs per gram sample	0	no rinse	6.74	=(D27+D28)/2
30	amount of Cs in rinse 1 solution, ug	0	no rinse	46	=D25*D29
31	**percentage of Cs eluted in rinse 1**	**0.00**	**no rinse**	**0.85**	**=(D30/D8)*100**
32	**Contact 2**				
33	amount of oxalic acid added to vessel	229.3	measured in laboratory	229.3	measured in laboratory
34	amount of liquid present at T2 sampling, g	240.9	=B33+B26	242.2	=D33+D26
35	ug Cs per gram sample -- run 1	3.12	ICP-MS measurement	2.30	ICP-MS measurement
36	ug Cs per gram sample -- run 2		not run in duplicate	2.14	ICP-MS measurement
37	average ug Cs per gram sample	3.12	=B35	2.22	=(D35+D36)/2
38	amount of Cs in solution at T2, ug	752	=B34*B37	538	=D34*D37
39	**percentage of Cs eluted at T2**	**13.96**	**=(B38/B8)*100**	**9.99**	**=(D38/D8)*100**

Table E.2, continued

	A	B	C	D	E
40	**Rinse 2**				
41	weight of assembly after removal of supernate	2012.5	measured in laboratory	2065.5	measured in laboratory
42	material remaining in vessel after contact 2	11.4	=B41-B12	11.5	=D41-D12
43	amount of deionized rinse water added	0.00	no rinse	7.77	measured in laboratory
44	amount of material removed after rinse	0.00	no rinse	6.91	measured in laboratory
45	material remaining in vessel after rinse 2	11.4	=B42	12.4	=D42+D43-D44
46	ug Cs per gram sample -- run 1		no rinse	1.01	ICP-MS measurement
47	ug Cs per gram sample -- run 2				not run in duplicate
48	average ug Cs per gram sample	0.000	no rinse	1.01	=D46
49	amount of Cs in rinse 1 solution, ug	0	=B44*B48	7	=D44*D48
50	**percentage of Cs eluted in rinse 2**	**0.00**	**=(B49/B8)*100**	**0.13**	**=(D49/D8)*100**
51	**Contact 3**				
52	amount of oxalic acid added to vessel	229.3	measured in laboratory	229.3	measured in laboratory
53	amount of liquid present at T3 sampling, g	240.7	=B45+B52	241.7	=D45+D52
54	ug Cs per gram sample -- run 1	0.962	ICP-MS measurement	0.853	ICP-MS measurement
55	ug Cs per gram sample -- run 2		not run in duplicate		not run in duplicate
56	average ug Cs per gram sample	0.962	=B54	0.853	=D54
57	amount of Cs in solution at T3, ug	232	=B53*B56	206	=D53*D56
58	**percentage of Cs eluted at T3**	**4.30**	**=(B57/B8)*100**	**3.83**	**=(D57/D8)*100**
59	**cumulative percentages eluted**				
60	contact 1	88.21	=B20	89.55	=D20
61	rinse 1			90.39	=D60+D31
62	contact 2	102.17	=B60+B31+B39	100.38	=D61+D39
63	rinse 2			100.51	=D62+D50
64	contact 3	106.47	=B62+B50+B58	104.34	=D63+D58

Table E.3. Sample Calculations and Formulas Used in Determination of Percent Cs Eluted in Effects of Condition-Modifications Experiment

	A	B	C
1			
2		Test 17	
3	**Time 0 : initial conditions**		
4	initial amount of zeolite in reaction vessel, g	7.76	measured in laboratory
5	F factor of zeolite (= dry weight/ wet weight)	0.724	measured in laboratory
6	amount of dry zeolite used, g	5.62	=B4*B5
7	ug Cs per gram dry zeolite	957	XRF measurement
8	total amount Cs initially present in vessel, ug	5377	=B6*B7
9	weight of oxalic acid initially present, g	229.3	measured in laboratory
10	weight of NaOH (pH ~ 11) initially present, g	2.76	measured in laboratory
11	weight of Fe_2O_3 present, g	4.13	measured in laboratory
12	total initial material weight, g	243.95	=B4+B9+B10+B11
13	weight loss per hour, g	0.1	measured in laboratory
14	final vessel weight	2209.0	measured in laboratory
15	empty vessel weight	1997.7	measured in laboratory
16	total finish weight	211.3	=B14-B15
17	**Time 1 = 2 hours**	2	measured in laboratory
18	weight of material present at T1, g	243.75	=B12-(B17*B13)
19	weight of sample removed at T1, g	6.15	measured in laboratory
20	ug Cs per gram sample -- run 1	14.5	ICP-MS measurement
21	ug Cs per gram sample -- run 2	15.5	ICP-MS measurement
22	average ug Cs per gram sample	15.0	=(B20+B21)/2
23	amount Cs in solution at T1, ug	3656	=B18*B22
24	**percentage Cs eluted at T1**	**68.00**	**=(B23/B8)*100**
25	**Time 2 = 4 hours**	4	measured in laboratory
26	weight of material present at T2, g	237.40	=B18-B19-(B25-B17)*B13
27	weight of sample removed at T2, g	5.17	measured in laboratory
28	ug Cs per gram sample -- run 1	18.2	ICP-MS measurement
29	ug Cs per gram sample -- run 2		not run in duplicate
30	average ug Cs per gram sample	18.2	=B28
31	amount of Cs in solution at T2, ug	4321	=B26*B30
32	amount of Cs removed during T1 sampling, ug	92	=B19*B22
33	total amount of Cs eluted, ug	4413	=B31+B32
34	**percentage of Cs eluted at T2**	**82.08**	**=(B33/B8)*100**

Table E.3, continued

	A	B	C
35	**Time 3 = 20 hours**	20	measured in laboratory
36	weight of material present at T3, g	230.63	=B26-B27-(B35-B25)*B13
37	weight of sample removed at T3, g	7.26	measured in laboratory
38	ug Cs per gram sample -- run 1	23.0	ICP-MS measurement
39	ug Cs per gram sample -- run 2	22.9	ICP-MS measurement
40	average ug Cs per gram sample	23.0	=(B38+B39)/2
41	amount of Cs in solution at T3, ug	5293	=B36*B40
42	amount of Cs removed during T1 sampling, ug	92	=B32
43	amount of Cs removed during T2 sampling, ug	94	=B27*B30
44	total amount of Cs removed during sampling, ug	186	=SUM(B42:B43)
45	total amount of Cs eluted at T3, ug	5479	=B41+B44
46	**percentage of Cs eluted at T3**	**101.91**	**=(B45/B8)*100**
47	**Time 4 = 28 hours**	28	measured in laboratory
48	weight of material present at T4, g	222.57	=B36-B37-(B47-B35)*B13
49	weight of sample removed at T4, g	5.26	measured in laboratory
50	ug Cs per gram sample -- run 1	20.5	ICP-MS measurement
51	ug Cs per gram sample -- run 2		not run in duplicate
52	average ug Cs per gram sample	20.5	=B50
53	amount of Cs in solution at T4, ug	4563	=B48*B52
54	amount of Cs removed during T1 sampling, ug	92	=B32
55	amount of Cs removed during T2 sampling, ug	94	=B43
56	amount of Cs removed during T3 sampling, ug	167	=B37*B40
57	total amount of Cs removed during sampling, ug	353	=SUM(B54:B56)
58	total amount of Cs eluted at T4, ug	4916	=B53+B57
59	**percentage of Cs eluted at T4**	**91.43**	**=(B58/B8)*100**
60	**Time 5 = 50 hours**	50	measured in laboratory
61	weight of material present at T5, g	215.11	=B48-B49-(B60-B47)*B13
62	weight of sample removed at T5, g	6.04	measured in laboratory
63	ug Cs per gram sample -- run 1	21.3	ICP-MS measurement
64	ug Cs per gram sample -- run 2		not run in duplicate
65	average ug Cs per gram sample	21.3	=B63
66	amount of Cs in solution at T5, ug	4582	=B61*B65
67	amount of Cs removed during T1 sampling, ug	92	=B32
68	amount of Cs removed during T2 sampling, ug	94	=B43
69	amount of Cs removed during T3 sampling, ug	167	=B56
70	amount of Cs removed during T4 sampling, ug	108	=B49*B52
71	total amount of Cs removed during sampling, ug	461	=SUM(B67:B70)
72	total amount of Cs eluted at T5, ug	5043	=B66+B71
73	**percentage of Cs eluted at T5**	**93.79**	**=(B72/B8)*100**
74	density of oxalic acid	1.02	measured in laboratory
75	actual liters per kg	40.0	=(B9/B74)/B6

Table E.4. Sample Calculations and Formulas Used in Determination of Percent Cs Eluted in Experiment to Determine the Effects of Varying Oxalic Acid Concentration

	A	B	C
1			
2		Test 23	source of number
3	**Contact 1, time 0: initial conditions**		
4	initial amount of zeolite in reaction vessel, g	5.40	measured in laboratory
5	F factor of zeolite (=dry weight/wet weight)	0.9261	measured in laboratory
6	amount of dry zeolite used, g	5.00	=B4*B5
7	ug Cs/g dry zeolite	899	XRF measurement
8	total amount Cs initially present in vessel, ug	4496	=B6*B7
9	weight of oxalic acid initially present, g	508.7	measured in laboratory
10	weight of NaOH (pH ~ 11) initially present, g	2.47	measured in laboratory
11	weight of Fe2O3 initially present, g	5.90	measured in laboratory
12	total initial material weight, g	517.07	=SUM(B9:B11)
13	weight of empty vessel, g	1872.5	measured in laboratory
14	weight of loaded vessel, g	2389.57	measured in laboratory
15	weight loss per hour, g	0.05	measured in laboratory
16	**Contact 1, time 1 = 2 hours**	2	
17	Weight of material present at C1T1, g	516.97	=B12-(B16*B15)
18	Weight of sample removed at C1T1, g	5.70	measured in laboratory
19	Cs concentration, run 1, ug Cs/g sample	1.84	ICP-MS measurement
20	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
21	average Cs concentration, ug/g	1.84	=B19
22	amount of Cs in solution at C1T1, ug	951	=B17*B21
23	percentage Cs dissolved at C1T1	**21.16**	=(B22/B8)*100
24	Contact 1, time 2 = 4 hours	4	
25	Weight of material present at C1T2, g	511.17	=B17-B18-(B24-B16)*B15
26	Weight of sample removed at C1T2, g	5.76	measured in laboratory
27	Cs concentration, run 1, ug Cs/g sample	2.9	ICP-MS measurement
28	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
29	average Cs concentration, ug/g	2.90	=B27
30	amount of Cs in solution at C1T2, ug	1482	=B25*B29
31	amount of Cs removed during C1T1 sampling, ug	10.49	=B18*B21
32	total amount of Cs dissolved at C1T2, ug	1492.88	=B30+B31
33	percentage of Cs dissolved at C1T2	**33.21**	=(B32/B8)*100
34	**Contact 1, time 3 = 20 hours**	20	
35	weight of material present at C1T3, g	504.61	=B25-B26-(B34-B24)*B15
36	weight of sample removed at C1T3, g	5.95	measured in laboratory
37	Cs concentration, run 1, ug Cs/g sample	5.08	ICP-MS measurement
38	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
39	average Cs concentration, ug/g	5.08	=B37
40	amount of Cs in solution at C1T3, ug	2563	=B35*B39

Table E.4, continued

	A	B	C
41	amount of Cs removed during C1T1 sampling, ug	10.49	=B31
42	amount of Cs removed during C1T2 sampling, ug	16.70	=B26*B29
43	total amount of Cs removed during sampling, ug	27.19	=SUM(B41:B42)
44	total amount of Cs dissolved at C1T3, ug	2591	=B40+B43
45	percentage of Cs dissolved at C1T3	**57.62**	=(B44/B8)*100
46	**Contact 1, time 4 = 28 hours**	28	
47	weight of material present at C1T4, g	498.26	=B35-B36-(B46-B34)*B15
48	weight of sample removed at C1T4, g	5.47	measured in laboratory
49	Cs concentration, run 1, ug Cs/g sample	5.80	ICP-MS measurement
50	Cs concentration, run 2, ug Cs/g sample	6.33	ICP-MS measurement
51	average Cs concentration, ug/g	6.07	=(B49+B50)/2
52	amount of Cs in solution at C1T4, ug	3022	=B47*B51
53	amount of Cs removed during C1T1 sampling, ug	10.49	=B31
54	amount of Cs removed during C1T2 sampling, ug	16.70	=B42
55	amount of Cs removed during C1T3 sampling, ug	30.23	=B36*B39
56	total amount of Cs removed during sampling, ug	57.42	=SUM(B53:B55)
57	total amount of Cs dissolved at C1T4, ug	3079	=B52+B56
58	percentage of Cs dissolved at C1T4	**68.49**	=(B57/B8)*100
59	**Contact 1, time 5 = 50 hours**	50	
60	weight of material present at C1T5, g	491.69	=B47-B48-(B59-B46)*B15
61	weight of sample removed at C1T5, g	5.60	measured in laboratory
62	Cs concentration, run 1, ug Cs/g sample	6.73	ICP-MS measurement
63	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
64	average Cs concentration, ug/g	6.73	=B62
65	amount of Cs in solution at C1T5, ug	3309.07	=B60*B64
66	amount of Cs removed during C1T1 sampling, ug	10.49	=B31
67	amount of Cs removed during C1T2 sampling, ug	16.70	=B42
68	amount of Cs removed during C1T3 sampling, ug	30.23	=B55
69	amount of Cs removed during C1T4 sampling, ug	33.18	=B48*B51
70	total amount of Cs removed during sampling, ug	90.59	=SUM(B66:B69)
71	total amount of Cs dissolved at C1T5, ug	3399.67	=B65+B70
72	percentage of Cs dissolved at C1T5	**75.62**	=(B71/B8)
73	**Contact 2, time 0: changeover**		
74	weight of vessel after C1T5 sampling, g	2364.0	measured in laboratory
75	weight of vessel after contact 1 removal, g	1888.2	measured in laboratory
76	weight of contact 1 supernatant removed, g	475.8	=B74-B75
77	amount of Cs removed from system, ug	3202.13	=B76*B64
78	total amount of Cs removed during contact 1, ug	3330.42	=B77+B70+(B61*B64)
79	amount of Cs remaining in system at C2T0, ug	1165.43	=B8-B78
80	amount of material remaining in system at C2T0, g	10.29	=B60-B61-B76

Table E.4, continued

	A	B	C
81	weight of oxalic acid added for contact 2, g	508.7	measured in laboratory
82	total initial material weight, g	518.99	=B80+B81
83	weight loss per hour, g	0.05	measured in laboratory
84	**Contact 2, time 1 = 2 hours**	2	
85	weight of material present at C2T1, g	518.90	=B82-(B84*B83)
86	weight of sample removed at C2T1, g	6.03	measured in laboratory
87	Cs concentration, run 1, ug Cs/g sample	0.2	ICP-MS measurement
88	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
89	average Cs concentration, ug/g	0.2	=B87
90	amount of Cs in solution at C2T1, ug	104	=B85*B89
91	percentage Cs dissolved at C2T1	76.39	=((B78+B90)/B8)*100
92	**Contact 2, time 2 = 4 hours**	4	
93	weight of material present at C2T2, g	512.77	
94	weight of sample removed at C2T2,g	5.02	measured in laboratory
95	Cs concentration, run 1, ug Cs/g sample	0.354	ICP-MS measurement
96	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
97	average Cs concentration, ug/g	0.35	=B95
98	amount of Cs in solution at C2T2, ug	181.52	=B93*B97
99	amount of Cs removed during C2T1 sampling, ug	1.21	=B86*B89
100	total amount of Cs dissolved at C2T2, ug	183	=B98*B99
101	percentage Cs dissolved at C2T2	78.14	=((B78+B100)/B8)*100
102	**Contact 2, time 3 = 20 hours**	20	
103	weight of material present at C2T3, g	507.01	=B93-B94-(B102-B92)*B83
104	weight of sample removed at C2T3, g	5.42	measured in laboratory
105	Cs concentration, run 1, ug Cs/g sample	0.540	ICP-MS measurement
106	Cs concentration, run 2, ug Cs/g sample	0.563	ICP-MS measurement
107	average Cs concentration, ug/g	0.55	=(B105+B106)/2
108	amount of Cs in solution at C2T3, ug	279.62	=B103*B107
109	amount of Cs removed during C2T1 sampling, ug	1.21	=B99
110	amount of Cs removed during C2T2 sampling, ug	1.78	=B94*B97
111	total amount of Cs removed during sampling, ug	2.98	=B109+B110
112	total amount of Cs dissolved at C2T3, ug	283	=B108+B111
113	percentage Cs dissolved at C2T3	80.36	=((B78+B112)/B8)*100
114	**Contact 2, time 4 = 28 hours**	28	
115	weight of material present at C2T4, g	501.22	=B103-B104-(B114-B102)*B83
116	weight of sample removed at C2T4, g	6.18	measured in laboratory
117	Cs concentration, run 1, ug Cs/g sample	0.386	ICP-MS measurement
118	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
119	average Cs concentration, ug/g	0.386	=B117
120	amount of Cs in solution at C2T4, ug	193.47	=B115*B119

Table E.4, continued

	A	B	C
121	amount of Cs removed during C2T1 sampling, ug	1.21	=B99
122	amount of Cs removed during C2T2 sampling, ug	1.78	=B110
123	amount of Cs removed during C2T3 sampling, ug	2.99	=B104*B107
124	total amount of Cs removed during sampling, ug	5.97	=SUM(B121:B123)
125	total amount of Cs dissolved at C2T4, ug	199	=B120+B124
126	percentage Cs dissolved at C2T4	**78.51**	=((B78+B125)/B8)*100
127	**Contact 2, time 5 = 50 hours**	50	
128	weight of material present at C2T5, g	494.02	=B115-B116-(B127-B114)*B83
129	weight of sample removed at C2T5, g	5.87	measured in laboratory
130	Cs concentration, run 1, ug Cs/g sample	0.526	ICP-MS measurement
131	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
132	average Cs concentration, ug/g	0.526	=B130
133	amount of Cs in solution at C2T5, ug	259.85	=B128*B132
134	amount of Cs removed during C2T1 sampling, ug	1.21	=B99
135	amount of Cs removed during C2T2 sampling, ug	1.78	=B110
136	amount of Cs removed during C2T3 sampling, ug	2.99	=B123
137	amount of Cs removed during C2T4 sampling, ug	2.39	=B116*B119
138	total amount of Cs removed during sampling, ug	8.36	=SUM(B134:B137)
139	total amount of Cs dissolved at C2T5, ug	268	=B133+B138
140	percentage Cs dissolved at C2T5	**80.04**	=((B78+B139)/B8)*100
141	**Contact 3, time 0: changeover**		
142	weight of vessel after C2T5 sampling, g	2366.2	measured in laboratory
143	weight of vessel after contact 2 removal, g	1885.4	measured in laboratory
144	weight of contact 2 supernatant removed, g	480.8	=B142-B143
145	amount of Cs removed from system, ug	253	=B144*B132
146	total amount of Cs removed during contact 2, ug	264	=B145+B138+(B129*B132)
147	amount of Cs remaining in system at C3T0, ug	901	=B8-B78-B146
148	amount of material remaining in system at C3T0, g	7.35	=B128-B129-B144
149	weight of oxalic acid added for contact 3, g	508.70	measured in laboratory
150	total initial material weight, g	516.05	=B148+B149
151	weight loss per hour, g	0.07	measured in laboratory
152	**Contact 3, time 1 = 2 hours**	2	
153	weight of material present at C3T1, g	515.91	=B150-(B151*B152)
154	weight of sample removed at C3T1, g	5.93	measured in laboratory
155	Cs concentration, run 1, ug Cs/g sample	0.105	ICP-MS measurement
156	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
157	average Cs concentration, ug/g	0.11	=B155
158	amount of Cs in solution at C3T1, ug	54.17	=B153*B157
159	percentage of Cs dissolved at C3T1	**81.16**	=((B78+B146+B158)/B8)*100

Table E.4, continued

	A	B	C
160	**Contact 3, time 2 = 4 hours**	4	
161	weight of material present at C3T2, g	509.85	=B153-B154-(B160-B152)*B151
162	weight of sample removed at C3T2, g	6.05	measured in laboratory
163	Cs concentration, run 1, ug Cs/g sample	0.0558	ICP-MS measurement
164	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
165	average Cs concentration, ug/g	0.06	=B163
166	amount of Cs in solution at C3T2, ug	28.45	=B161*B165
167	amount of Cs removed during C3T1 sampling, ug	0.62	=B154*B157
168	total amount of Cs dissolved at C3T2, ug	29	=B166+B167
169	percentage of Cs dissolved at C3T2	80.60	=((B78+B146+B168)/B8)*100
170	**Contact 3, time 3 = 20 hours**	20	
171	weight of material present at C3T3, g	502.69	=B161-B162-(B170-B160)*B151
172	weight of sample removed at C3T3, g	6.22	measured in laboratory
173	Cs concentration, run 1, ug Cs/g sample	0.0681	ICP-MS measurement
174	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
175	average Cs concentration, ug/g	0.07	=B173
176	amount of Cs in solution at C3T3, ug	34.23	=B171*B175
177	amount of Cs removed during C3T1 sampling, ug	0.62	=B167
178	amount of Cs removed during C3T2 sampling, ug	0.34	=B162*B165
179	total amount of Cs removed during sampling, ug	0.96	=B177+B178
180	total amount of Cs dissolved at C3T3, ug	35	=B176+B179
181	percentage Cs dissolved at C3T3	80.74	=((B78+B146+B180)/B8)*100
182	**Contact 3, time 4 = 28 hours**	28	
183	weight of material present at C3T4, g	495.92	=B171-B172-(B182-B170)*B151
184	weight of sample removed at C3T4, g	5.23	measured in laboratory
185	Cs concentration, run 1, ug Cs/g sample	0.0709	ICP-MS measurement
186	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
187	average Cs concentration, ug/g	0.07	=B185
188	amount of Cs in solution at C3T4, ug	35.16	=B183*B187
189	amount of Cs removed during C3T1 sampling, ug	0.62	=B167
190	amount of Cs removed during C3T2 sampling, ug	0.34	=B178
191	amount of Cs removed during C3T3 sampling, ug	212.93	=B172*B176
192	total amount of Cs removed during sampling, ug	213.89	=SUM(B189:B191)
193	total amount of Cs dissolved at C3T4, ug	249	=B188+B192
194	percentage Cs dissolved at C3T4	85.50	=((B78+B146+B193)/B8)*100
195	**Contact 3, time 5 = 50 hours**	50	
196	weight of material present at C3T5, g	489.17	=B183-B184-(B195-B182)*B151
197	weight of sample removed at C3T5, g	6.53	measured in laboratory
198	Cs concentration, run 1, ug Cs/g sample	0.114	ICP-MS measurement
199	Cs concentration, run 2, ug Cs/g sample		ICP-MS measurement
200	average Cs concentration, ug/g	0.114	=B198

Table E.4, continued

	A	B	C
201	amount of Cs in solution at C3T5, ug	55.77	=B196*B200
202	amount of Cs removed during C3T1 sampling, ug	0.62	=B167
203	amount of Cs removed during C3T2 sampling, ug	0.34	=B178
204	amount of Cs removed during C3T3 sampling, ug	212.93	=B191
205	amount of Cs removed during C3T4 sampling, ug	0.37	=B184*B187
206	total amount of Cs removed during sampling, ug	214.26	=SUM(B202:B205)
207	total amount of Cs dissolved at C3T5, ug	270	=B201+B206
208	percentage Cs dissolved at C3T5	85.96	=((B78+B146+B207)/B8)*100
209	**Summary**		
210	contact 1, 2 hours	21.16	=B23
211	contact 1, 4 hours	33.21	=B33
212	contact 1, 20 hours	57.62	=B45
213	contact 1, 28 hours	68.49	=B58
214	contact 1, 50 hours	75.62	=B72
215	contact 2, 2 hours	76.39	=B91
216	contact 2, 4 hours	78.14	=B101
217	contact 2, 20 hours	80.36	=B113
218	contact 2, 28 hours	78.51	=B126
219	contact 2, 50 hours	80.04	=B140
220	contact 3, 2 hours	81.16	=B159
221	contact 3, 4 hours	80.60	=B169
222	contact 3, 20 hours	80.74	=B181
223	contact 3, 28 hours	85.50	=B194
224	contact 3, 50 hours	85.96	=B208

Table E.5. Sample Calculations and Formulas Used in Determination of Percent Fe Dissolved in Experiment to Determine the Effects of Varying Oxalic Acid Concentration

	A	B	C
1			
2		Test 23	
3	**Contact 1, time 0: initial conditions**		
4	initial amount of zeolite in reaction vessel, g	5.40	measured in laboratory
5	F factor of zeolite (=dry weight/wet weight)	0.9261	measured in laboratory
6	amount of dry zeolite used, g	5.00	=B4*B5
7	ug Fe/g dry zeolite	24480	ICP-MS measurement
8	total amount Fe initially present in vessel, ug	4252423	=(B6*B7)+4130000; 413000=Fe in added Fe2O3
9	weight of oxalic acid initially present, g	508.7	measured in laboratory
10	weight of NaOH (pH ~ 11) initially present, g	2.47	measured in laboratory
11	weight of Fe2O3 initially present, g	5.90	measured in laboratory
12	total initial material weight, g	517.07	=SUM(B9:B11)
13	weight of empty vessel, g	1872.5	measured in laboratory
14	weight of loaded vessel, g	2389.57	=B12+B13
15	weight loss per hour, g	0.05	measured in laboratory
16	**Contact 1, time 1 = 2 hours**	2	
17	Weight of material present at C1T1, g	516.97	=B12-(B16*B15)
18	Weight of sample removed at C1T1, g	5.70	measured in laboratory
19	Fe concentration, run 1, ug Fe/g sample	90	ICP-MS measurement
20	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
21	average Fe concentration, ug/g	90	=B19
22	amount of Fe in solution at C1T1, ug	46527	=B17*B21
23	**percentage Fe dissolved at C1T1**	1.09	=(B22/B8)*100
24	**Contact 1, time 2 = 4 hours**	4	
25	Weight of material present at C1T2, g	511.17	=B17-B18-(B24-B16)*B15
26	Weight of sample removed at C1T2, g	5.76	measured in laboratory
27	Fe concentration, run 1, ug Fe/g sample	196	ICP-MS measurement
28	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
29	average Fe concentration, ug/g	196	=B27
30	amount of Fe in solution at C1T2, ug	100189	=B25*B29
31	amount of Fe removed during C1T1 sampling, ug	513	=B18*B21
32	total amount of Fe dissolved at C1T2, ug	100702	=B30+B31
33	**percentage of Fe dissolved at C1T2**	2.37	=(B32/B8)*100
34	**Contact 1, time 3 = 20 hours**	20	
35	weight of material present at C1T3, g	504.61	=B25-B26-B(34-B24)*B15
36	weight of sample removed at C1T3, g	5.95	measured in laboratory
37	Fe concentration, run 1, ug Fe/g sample	540	ICP-MS measurement
38	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
39	average Fe concentration, ug/g	540	=B37
40	amount of Fe in solution at C1T3, ug	272489	=B35*B39

Table E.5, continued

	A	B	C
41	amount of Fe removed during C1T1 sampling, ug	513	=B31
42	amount of Fe removed during C1T2 sampling, ug	1129	=B26*B29
43	total amount of Fe removed during sampling, ug	1642	=SUM(B41:B42)
44	total amount of Fe dissolved at C1T3, ug	274131	=B40+B43
45	**percentage of Fe dissolved at C1T3**	**6.45**	=(B44/B8)*100
46	**Contact 1, time 4 = 28 hours**	28	
47	weight of material present at C1T4, g	498.26	=B35-B36-(B46-B34)*B15
48	weight of sample removed at C1T4, g	5.47	measured in laboratory
49	Fe concentration, run 1, ug Fe/g sample	656	ICP-MS measurement
50	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
51	average Fe concentration, ug/g	656	=B49
52	amount of Fe in solution at C1T4, ug	326859	=B47*B51
53	amount of Fe removed during C1T1 sampling, ug	513	=B31
54	amount of Fe removed during C1T2 sampling, ug	1129	=B42
55	amount of Fe removed during C1T3 sampling, ug	3213	=B36*B39
56	total amount of Fe removed during sampling, ug	4855	=SUM(B53:B55)
57	total amount of Fe dissolved at C1T4, ug	331714	=B52+B56
58	**percentage of Fe dissolved at C1T4**	**7.80**	=(B57/B8)*100
59	**Contact 1, time 5 = 50 hours**	50	
60	weight of material present at C1T5, g	491.69	=B47-B48-(B59-B46)*B15
61	weight of sample removed at C1T5, g	5.60	measured in laboratory
62	Fe concentration, run 1, ug Fe/g sample	890	ICP-MS measurement
63	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
64	average Fe concentration, ug/g	890	=B62
65	amount of Fe in solution at C1T5, ug	437604	=B60*B64
66	amount of Fe removed during C1T1 sampling, ug	513	=B31
67	amount of Fe removed during C1T2 sampling, ug	1129	=B42
68	amount of Fe removed during C1T3 sampling, ug	3213	=B55
69	amount of Fe removed during C1T4 sampling, ug	3588	=B48*B51
70	total amount of Fe removed during sampling, ug	8443	=SUM(B66:B69)
71	total amount of Fe dissolved at C1T5, ug	446047	=B65+B70
72	**percentage of Fe dissolved at C1T5**	**10.49**	=(B71/B8)*100
73	**Contact 2, time 0: changeover**		
74	weight of vessel after C1T5 sampling, g	2364.0	measured in laboratory
75	weight of vessel after contact 1 removal, g	1888.2	measured in laboratory
76	weight of contact 1 supernatant removed, g	475.8	=B74-B75
77	amount of Fe removed from system, ug	423462	=B76*B64
78	total amount of Fe removed during contact 1, ug	436889	=B77+B70+(B61*B64)
79	amount of Fe remaining in system at C2T0, ug	3815534	=B8-B78
80	amount of material remaining in system at C2T0, g	10.29	=B60-B61-B76

Table E.5, continued

	A	B	C
81	weight of oxalic acid added for contact 2, g	508.7	measured in laboratory
82	total initial material weight, g	518.99	=B80+B81
83	weight loss per hour, g	0.05	measured in laboratory
84	**Contact 2, time 1 = 2 hours**	2	
85	weight of material present at C2T1, g	518.90	=B82-(B84*B83)
86	weight of sample removed at C2T1, g	6.03	measured in laboratory
87	Fe concentration, run 1, ug Fe/g sample	302	ICP-MS measurement
88	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
89	average Fe concentration, ug/g	302	=B87
90	amount of Fe in solution at C2T1, ug	156707	=B85*B89
91	**percentage Fe dissolved at C2T1**	13.96	=((B78+B90)/B8)*100
92	**Contact 2, time 2 = 4 hours**	4	
93	weight of material present at C2T2, g	512.78	=B85-B86-(B92-B84)*B83
94	weight of sample removed at C2T2,g	5.02	measured in laboratory
95	Fe concentration, run 1, ug Fe/g sample	316	ICP-MS measurement
96	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
97	average Fe concentration, ug/g	316	=B95
98	amount of Fe in solution at C2T2, ug	162037	=B93*B97
99	amount of Fe removed during C2T1 sampling, ug	1821	=B86*B89
100	total amount of Fe dissolved at C2T2, ug	163858	=B98+B99
101	**percentage Fe dissolved at C2T2**	14.13	=((B78+B100)/B8)*100
102	**Contact 2, time 3 = 20 hours**	20	
103	weight of material present at C2T3, g	507.02	=B93-B94-(B102-B92)*B83
104	weight of sample removed at C2T3, g	5.42	measured in laboratory
105	Fe concentration, run 1, ug Fe/g sample	507	ICP-MS measurement
106	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
107	average Fe concentration, ug/g	507	=B105
108	amount of Fe in solution at C2T3, ug	257059	=B103*B107
109	amount of Fe removed during C2T1 sampling, ug	1821	=B99
110	amount of Fe removed during C2T2 sampling, ug	1586	=B94*B97
111	total amount of Fe removed during sampling, ug	3407	=B109+B110
112	total amount of Fe dissolved at C2T3, ug	260467	=B108+B111
113	**percentage Fe dissolved at C2T3**	16.40	=((B78+B112)/B8)*100
114	**Contact 2, time 4 = 28 hours**	28	
115	weight of material present at C2T4, g	501.23	=B103-B104-(B114-B102)*B83
116	weight of sample removed at C2T4, g	6.18	measured in laboratory
117	Fe concentration, run 1, ug Fe/g sample	399	ICP-MS measurement
118	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
119	average Fe concentration, ug/g	399	=B117
120	amount of Fe in solution at C2T4, ug	199992	=B115*B119

Table E.5, continued

	A	B	C
121	amount of Fe removed during C2T1 sampling, ug	1821	=B99
122	amount of Fe removed during C2T2 sampling, ug	1586	=B110
123	amount of Fe removed during C2T3 sampling, ug	2748	=B104*B107
124	total amount of Fe removed during sampling, ug	6155	=SUM(B121:B123)
125	total amount of Fe dissolved at C2T4, ug	206147	=B120+B124
126	**percentage Fe dissolved at C2T4**	**15.12**	=((B78+B125)/B8)*100
127	**Contact 2, time 5 = 50 hours**	50	
128	weight of material present at C2T5, g	494.04	=B115-B116-(B127-B114)*B83
129	weight of sample removed at C2T5, g	5.87	measured in laboratory
130	Fe concentration, run 1, ug Fe/g sample	250	ICP-MS measurement
131	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
132	average Fe concentration, ug/g	250	=B130
133	amount of Fe in solution at C2T5, ug	123510	=B128*B132
134	amount of Fe removed during C2T1 sampling, ug	1821	=B99
135	amount of Fe removed during C2T2 sampling, ug	1586	=B110
136	amount of Fe removed during C2T3 sampling, ug	2748	=B123
137	amount of Fe removed during C2T4 sampling, ug	2466	=B116*B119
138	total amount of Fe removed during sampling, ug	8621	=SUM(B134:B137)
139	total amount of Fe dissolved at C2T5, ug	132131	=B133+B138
140	**percentage Fe dissolved at C2T5**	**13.38**	=((B78+B139)/B8)*B100
141	**Contact 3, time 0: changeover**		
142	weight of vessel after C2T5 sampling, g	2366.2	measured in laboratory
143	weight of vessel after contact 2 removal, g	1885.4	measured in laboratory
144	weight of contact 2 supernatant removed, g	480.8	=B142-B143
145	amount of Fe removed from system, ug	120200	=B144*B132
146	total amount of Fe removed during contact 2, ug	130289	=B145+B138+(B129*B132)
147	amount of Fe remaining in system at C3T0, ug	3685245	=B8-B78-B146
148	amount of material remaining in system at C3T0, g	7.37	=B128-B129-B144
149	weight of oxalic acid added for contact 3, g	508.7	measured in laboratory
150	total initial material weight, g	516.07	=B148+B149
151	weight loss per hour, g	0.07	measured in laboratory
152	**Contact 3, time 1 = 2 hours**	2	
153	weight of material present at C3T1, g	515.93	=B150-(B151*B152)
154	weight of sample removed at C3T1, g	5.93	measured in laboratory
155	Fe concentration, run 1, ug Fe/g sample	158	ICP-MS measurement
156	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
157	average Fe concentration, ug/g	158	=B155
158	amount of Fe in solution at C3T1, ug	81517	=B153*B157
159	**percentage of Fe dissolved at C3T1**	**15.25**	=((B78+B146+B158)/B8)*100

Table E.5, continued

	A	B	C
160	**Contact 3, time 2 = 4 hours**	4	
161	weight of material present at C3T2, g	509.86	=B153-B154-(B160-B152)*B151
162	weight of sample removed at C3T2, g	6.05	measured in laboratory
163	Fe concentration, run 1, ug Fe/g sample	113	ICP-MS measurement
164	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
165	average Fe concentration, ug/g	113	=B163
166	amount of Fe in solution at C3T2, ug	57615	=B161*B165
167	amount of Fe removed during C3T1 sampling, ug	937	=B154*B157
168	total amount of Fe dissolved at C3T2, ug	58552	=B166+B167
169	**percentage of Fe dissolved at C3T2**	**14.71**	=((B78+B146+B168)/B8)*100
170	**Contact 3, time 3 = 20 hours**	20	
171	weight of material present at C3T3, g	502.71	=B161-B162-(B170-B160)*B151
172	weight of sample removed at C3T3, g	6.22	measured in laboratory
173	Fe concentration, run 1, ug Fe/g sample	172	ICP-MS measurement
174	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
175	average Fe concentration, ug/g	172	=B173
176	amount of Fe in solution at C3T3, ug	86466	=B171*B175
177	amount of Fe removed during C3T1 sampling, ug	937	=B167
178	amount of Fe removed during C3T2 sampling, ug	684	=B162*B165
179	total amount of Fe removed during sampling, ug	1621	=B177+B178
180	total amount of Fe dissolved at C3T3, ug	88087	=B176+B179
181	**percentage Fe dissolved at C3T3**	**15.41**	=((B78+B146+B180)/B8)*100
182	**Contact 3, time 4 = 28 hours**	28	
183	weight of material present at C3T4, g	495.94	=B171-B172-(B182-B170)*B151
184	weight of sample removed at C3T4, g	5.23	measured in laboratory
185	Fe concentration, run 1, ug Fe/g sample	156	ICP-MS measurement
186	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
187	average Fe concentration, ug/g	156	=B185
188	amount of Fe in solution at C3T4, ug	77366	=B183*B187
189	amount of Fe removed during C3T1 sampling, ug	937	=B167
190	amount of Fe removed during C3T2 sampling, ug	684	=B178
191	amount of Fe removed during C3T3 sampling, ug	1070	=B172*B175
192	total amount of Fe removed during sampling, ug	2690	=SUM(B189:B191)
193	total amount of Fe dissolved at C3T4, ug	80057	=B188+B192
194	**percentage Fe dissolved at C3T4**	**15.22**	=((B78+B146+B193)/B8)*100
195	**Contact 3, time 5 = 50 hours**	50	
196	weight of material present at C3T5, g	489.19	=B183-B184-(B195-B182)*B151
197	weight of sample removed at C3T5, g	6.53	measured in laboratory
198	Fe concentration, run 1, ug Fe/g sample	172	ICP-MS measurement
199	Fe concentration, run 2, ug Fe/g sample		ICP-MS measurement
200	average Fe concentration, ug/g	172	=B198

Table E.5, continued

	A	B	C
201	amount of Fe in solution at C3T5, ug	84141	=B196*B200
202	amount of Fe removed during C3T1 sampling, ug	937	=B167
203	amount of Fe removed during C3T2 sampling, ug	684	=B178
204	amount of Fe removed during C3T3 sampling, ug	1070	=B191
205	amount of Fe removed during C3T4 sampling, ug	816	=B184*B187
206	total amount of Fe removed during sampling, ug	3506	=SUM(B202:B205)
207	total amount of Fe dissolved at C3T5, ug	87647	=B201+B206
208	**percentage Fe dissolved at C3T5**	**15.40**	=((B78+B146+B207)/B8)*100
209	**Summary**		
210	contact 1, 2 hours	1.09	=B23
211	contact 1, 4 hours	2.37	=B33
212	contact 1, 20 hours	6.45	=B45
213	contact 1, 28 hours	7.80	=B58
214	contact 1, 50 hours	10.49	=B72
215	contact 2, 2 hours	13.96	=B91
216	contact 2, 4 hours	14.13	=B101
217	contact 2, 20 hours	16.40	=B113
218	contact 2, 28 hours	15.12	=B126
219	contact 2, 50 hours	13.38	=B140
220	contact 3, 2 hours	15.25	=B159
221	contact 3, 4 hours	14.71	=B169
222	contact 3, 20 hours	15.41	=B181
223	contact 3, 28 hours	15.22	=B194
224	contact 3, 50 hours	15.40	=B208

Appendix F

Zeolite Dissolution Data From Cs-Elution Experiments: Amounts of Fe and Al Dissolved from Zeolite

Table F.1. Weight Percent Fe Dissolved from Zeolite During Experiment to Assess the Effects of Condition Modifications on Cs Elution

Time (hours)	Test 17 Fe_2O_3	Test 18 Coupons	Test 19 No Stir	Test 21 Control
2	2.8	61	4.0	34
4	6.2	97	63	40
20	5.1	110	100	79
28	63	120	73	65
50	92	160	76	91

Table F.2. Weight Percent Al Dissolved from Zeolite During Experiment to Assess the Effects of Condition Modifications on Cs Elution

Time (hours)	Test 17 Fe_2O_3	Test 18 Coupons	Test 19 No Stir	Test 21 Control
2	68	71	18	76
4	66	80	100	77
20	81	83	83	80
28	80	83	77	75
50	81	80	76	79

Appendix G

Data From Cs-Elution Tests Using Varying Concentrations of Oxalic Acid: Cs-Elution and Fe Dissolution

Table G.1. Weight Percent Cs Eluted in Cs-Elution Tests of Varying Oxalic Acid Concentration: Tests Using 4 wt% Oxalic Acid

Contact	Total Time (hours)	Test 22 Control	Test 23 Fe_2O_3	Test 24 Coupon
1	2	54	21	29
1	4	63	33	49
1	20	76	58	75
1	28	69	68	72
1	47	72	76	68
2	49	75	76	73
2	51	75	78	73
2	67	73	80	73
2	75	76	79	73
2	94	75	80	73
3	96	75	81	73
3	98	75	81	73
3	114	75	81	73
3	122	75	86	77
3	141	75	86	77

Table G.2. Weight Percent Cs Eluted in Cs-Elution Tests of Varying Oxalic Acid Concentration: Tests Using 1 wt% Oxalic Acid

Contact	Total Time (hours)	Test 26 Control	Test 27 Fe_2O_3	Test 28 Coupon
1	2	19	8.0	17
1	4	32	20	26
1	20	69	43	64
1	28	60	42	62
1	47	63	53	63
2	49	68	57	66
2	51	69	58	68
2	67	73	65	72
2	75	62	67	71
2	94	73	68	73
3	96	86	70	75
3	98	76	71	75
3	114	76	74	74
3	122	95	120	84
3	141	95	120	81

Table G.3. Weight Percent Fe Dissolved in Cs-Elution Tests of Varying Oxalic Acid Concentration: Tests Using 4 wt% Oxalic Acid

Contact	Total Time (hours)	Test 22 Control	Test 23 Fe_2O_3	Test 24 Coupon
1	2	72	1.1	49
1	4	70	2.4	84
1	20	81	6.5	98
1	28	100	7.8	110
1	47	79	10	100
2	49	81	14	120
2	51	79	14	130
2	67	77	16	170
2	75	79	15	180
2	94	85	13	200
3	96	85	15	260
3	98	88	15	250
3	114	88	15	280
3	122	90	15	270
3	141	90	15	300

Table G.4. Weight Percent Fe Dissolved in Cs-Elution Tests of Varying Oxalic Acid Concentration: Tests Using 1 wt% Oxalic Acid

Contact	Total Time (hours)	Test 26 Control	Test 27 Fe_2O_3	Test 28 Coupon
1	2	31	0.68	48
1	4	45	1.3	58
1	20	81	3.8	100
1	28	60	9.0	120
1	47	75	6.8	110
2	49	77	8.6	140
2	51	78	8.8	140
2	67	79	12	150
2	75	75	20	160
2	94	80	13	170
3	96	88	15	200
3	98	83	15	200
3	114	82	17	210
3	122	85	17	230
3	141	82	22	240

Table G.5. Amount Fe Dissolved (in ug) in Cs-Elution Tests of Varying Oxalic Acid Concentration: Tests Using 4 wt% Oxalic Acid

Contact	Total Time (hours)	Test 22 Control	Test 23 Fe_2O_3	Test 24 Coupon	Test 25 No Zeolite
1	2	88410	46527	59798	9816
1	4	85358	100702	102707	18051
1	20	98627	274131	120522	34756
1	28	122119	331714	129306	61362
1	47	96115	446047	125680	100756
2	49	99408	593596	150554	183344
2	51	97031	600747	159643	177718
2	67	94762	697352	205474	207271
2	75	97088	643032	224346	206184
2	94	104009	569016	240800	270127
3	96	103899	648692	318026	362178
3	98	107303	625727	311081	338906
3	114	107426	655262	340525	376585
3	122	110410	647232	335166	409820
3	141	109805	654822	369888	431457

Table G.6. Amount Fe Dissolved (in ug) in Cs-Elution Tests of Varying Oxalic Acid Concentration: Tests Using 1 wt% Oxalic Acid

Contact	Total Time (hours)	Test 26 Control	Test 27 Fe_2O_3	Test 28 Coupon	Test 29 No Zeolite
1	2	37586	28788	58953	7520
1	4	55213	56716	70891	14402
1	20	99517	162729	125301	39396
1	28	73492	382868	146048	42280
1	47	91660	287251	134012	61412
2	49	94030	363742	165740	88265
2	51	95470	374964	166780	93904
2	67	97055	518649	187468	116407
2	75	92374	866298	192753	121067
2	94	98235	534274	213619	151800
3	96	107167	637054	244079	214531
3	98	101355	640013	250439	214496
3	114	99832	720975	260396	249786
3	122	103691	702217	279870	274936
3	141	100263	929931	290268	307759

Appendix H

Temperature and pH Data Collected During Sludge-Dissolution Experiments

Table H.1. Temperature and pH Data Collected During Determination of Optimum Process Conditions for Sludge Dissolution Experiment

	Test 1		Test 2		Test 3	
	20 L/kg, 25°C		40 L/kg, 25°C		80 L/kg, 25°C	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)
Oxalic acid addition	0.929	24.0	0.920	24.0	0.913	24.0
pH at bath temperature	1.020	50.3	1.025	50.9	1.056	51.5
2 hour sampling	1.043	48.9	1.056	49.1	1.066	48.7
4 hour sampling	1.074	50.5	1.081	48.0	1.084	50.7
20 hour sampling	1.009	49.1	0.962	46.7	1.006	48.0
28 hour sampling	0.922	48.6	0.918	50.1	0.922	49.0
50 hour sampling	0.965	49.3	0.908	48.8	0.926	49.4

	Test 4		Test 5		Test 6	
	20 L/kg, 80°C		40 L/kg, 80°C		80 L/kg, 80°C	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)
Oxalic acid addition	0.705	79.0	0.944	81.0	1.015	80.0
pH at bath temperature	0.986	50.8	1.013	47.3	1.036	51.5
2 hour sampling	1.085	48.7	1.064	47.5	1.069	50.8
4 hour sampling	1.079	49.3	1.081	49.0	1.085	50.5
20 hour sampling	1.001	48.4	0.989	49.3	0.993	50.7
28 hour sampling	0.923	48.7	0.914	48.7	0.913	50.2
50 hour sampling	0.938	48.8	0.909	50.0	0.913	50.4

Table H.2. Temperature and pH Data Collected During Sludge-Dissolution Repeatability Experiment

	Test 2		Test 7		Test 8		Test 16	
	40 L/kg, 25°C		40 L/kg, 25°C		40 L/kg, 25°C		40 L/kg, 25°C	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
Oxalic acid addition	0.920	24.0	0.994	20.0	0.985	20.0	1.266	24.0
pH at bath temperature	1.025	50.9	0.815	50.0	0.820	50.0	1.183	50.0
2 hour sampling	1.056	49.1	0.833	49.1	0.845	49.1	1.272	47.3
4 hour sampling	1.081	48.0	0.866	48.8	0.863	48.4	1.224	48.2

Table H.3. Temperature and pH Data Collected During Long-Term Sludge-Dissolution Experiment

	Test 9 40 L/kg, 25°C	
Event	pH	T (°C)
Oxalic acid addition	1.263	22.0
pH at bath temperature	1.101	50.0
2 hour sampling	1.142	48.4
4 hour sampling	1.134	47.4
1 day sampling	1.082	49.3
4 day sampling	0.944	49.0
7 day sampling	1.218	47.4
11 day sampling	1.219	47.2

Table H.4. pH and Temeprature Data Collected During Multiple-Contact Sludge-Dissolution Experiment

	Test 10 Replaced Acid		Test 11 Added Acid	
Event	pH	T (°C)	pH	T (°C)
Oxalic acid addition	1.263	22.0	1.24	22.0
pH at bath temperature	1.067	50.0	0.992	55.5
End of contact 1	1.132	47.8	1.146	47.8
Oxalic acid addition	1.236	23.0	1.225	23.0
pH at bath temperature	1.136	50.0	1.048	50.0
End of contact 2	1.162	48.9	1.196	46.8
Oxalic acid addition	1.231	23.0	1.247	23.0
pH at bath temperature	1.158	50.0	1.163	50.0
End of contact 3	1.207	49.7	1.209	48.5

Table H.5. Temperature and pH Data Collected During Experiment to Assess the Effects of Condition Modifications on Sludge Dissolution

	Test 12 Fe_2O_3		Test 13 Coupons		Test 14 No Stir	
Event	pH	T (°C)	pH	T (°C)	pH	T (°C)
Oxalic acid addition	1.290	24.0	1.273	24.0	1.281	24.0
pH at bath temperature	1.240	50.0	1.185	50.0	1.208	50.0
2 hour sampling	1.223	47.9	1.246	48.0	1.149	47.1
4 hour sampling	1.210	49.0	1.213	48.9	1.220	48.7
20 hour sampling	1.075	50.0	1.080	50.6	1.111	49.1
28 hour sampling	0.867	50.2	0.872	49.8	0.884	48.5
50 hour sampling	1.015	49.7	0.968	50.1	0.980	49.0

	Test 15 25°C Bath		Test 16 Control	
Event	pH	T (°C)	pH	T (°C)
Oxalic acid addition	1.000	24.0	1.266	24.0
pH at bath temperature			1.183	50.0
2 hour sampling	1.014	23.4	1.272	47.3
4 hour sampling	1.016	23.4	1.224	48.2
20 hour sampling	1.144	24.2	1.145	48.5
28 hour sampling	1.184	23.0	0.893	48.9
50 hour sampling	1.052	24.2	0.994	48.4

Appendix I

Sludge-Dissolution Tests to Determine Optimum Process Conditions: Data and Plots of Percent Sludge Elements Dissolved

Table I.1. Weight Percent of Sludge Al Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	58	71	74	74	75	74
4	63	73	76	71	70	80
20	70	76	75	79	78	78
28	75	80	82	76	78	85
50	76	79	80	82	79	81

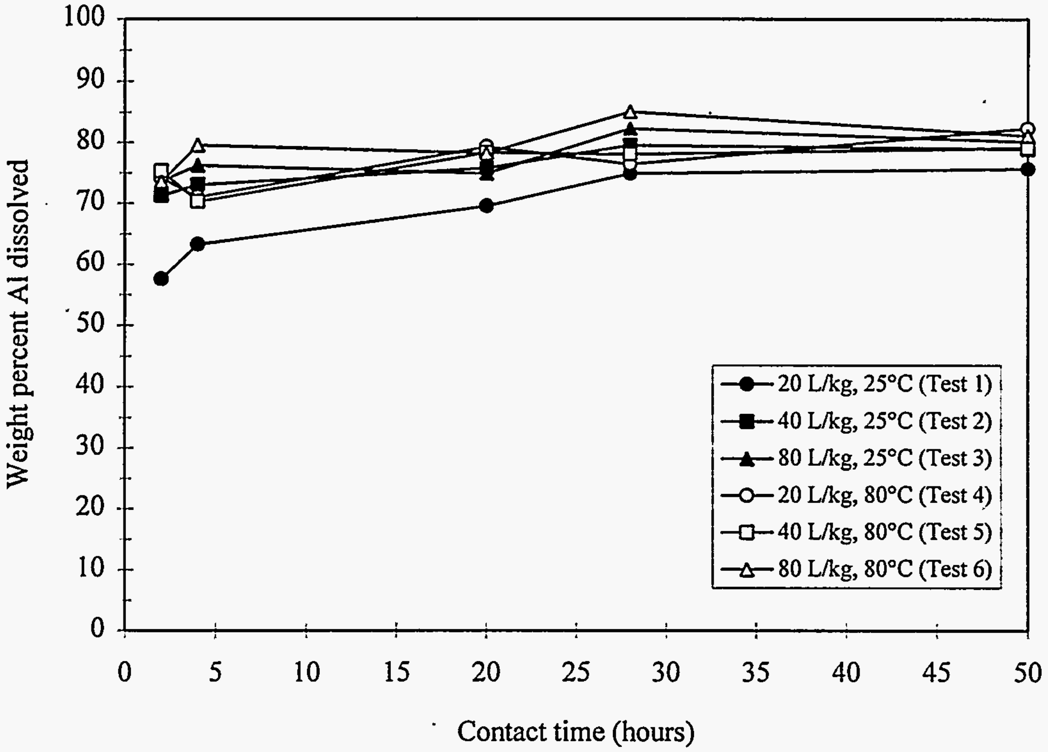

Figure I.1. Weight Percent of Sludge Al Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.2. Weight Percent of Sludge Ca Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	380	600	540	640	660	580
4	460	580	540	490	600	590
20	400	580	500	640	630	580
28	480	570	520	460	630	600
50	480	570	490	600	620	540

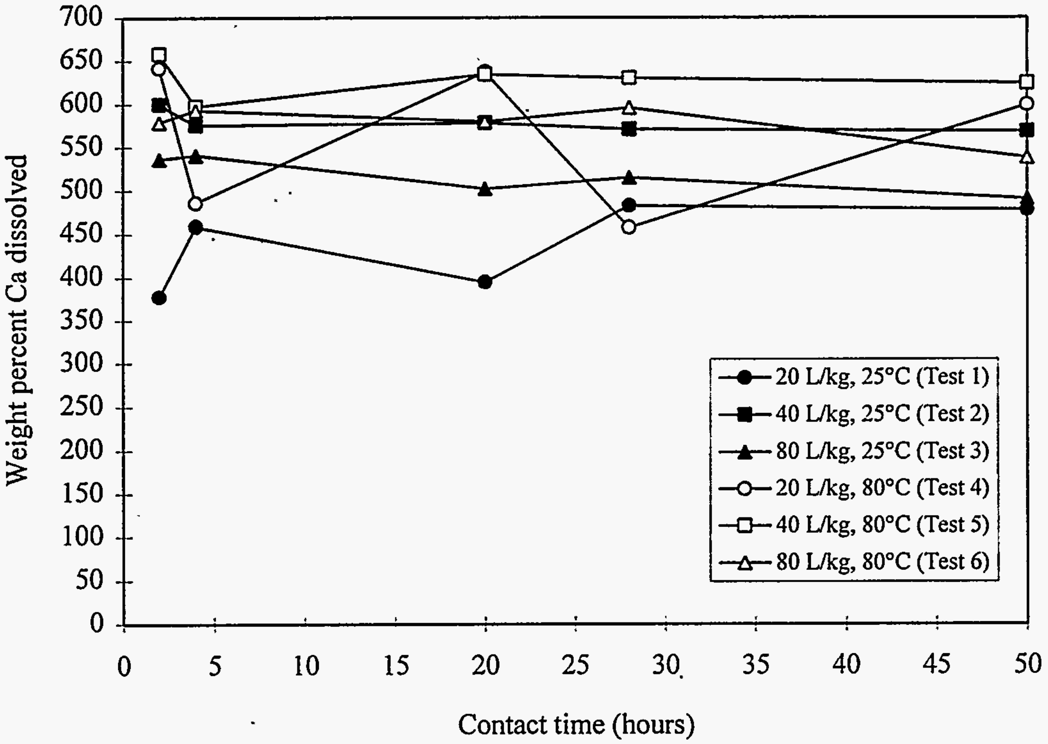

Figure I.2. Weight Percent of Sludge Ca Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.3. Weight Percent of Sludge Ce Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	20	15	14	7.5	7.7	6.3
4	15	10	9.0	9.6	11	12
20	7.9	7.0	15	4.8	10	16
28	15	61	9.3	7.1	8.0	11
50	17	35	7.7	4.6	21	7.7

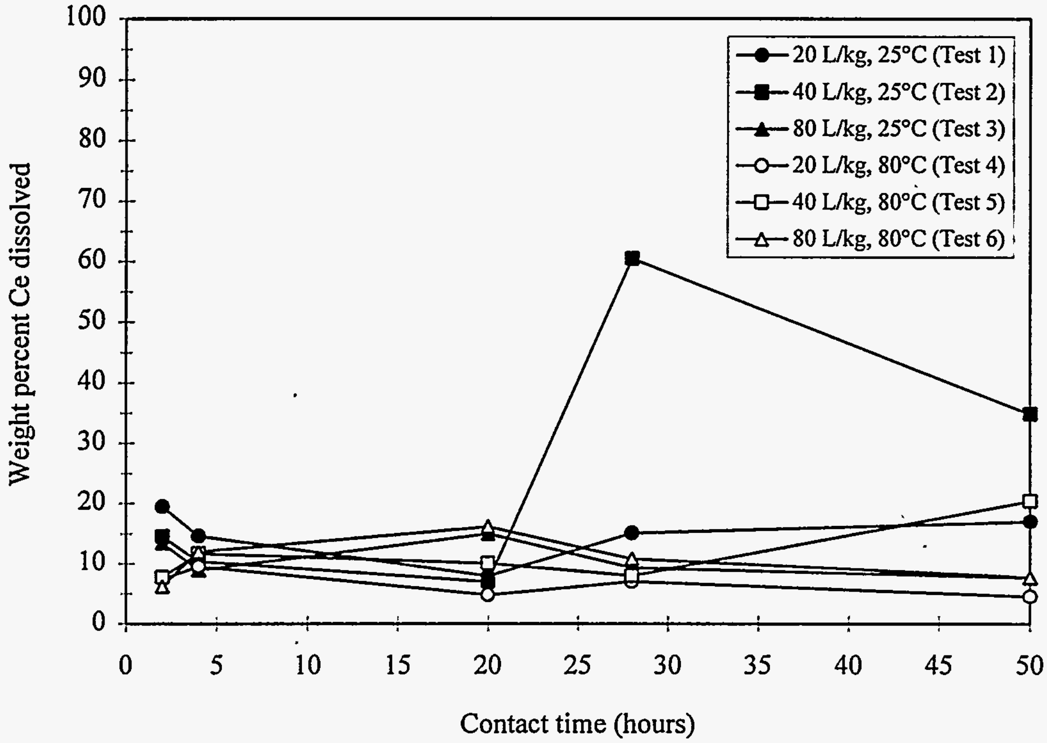

Figure I.3. Weight Percent of Sludge Ce Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.4. Weight Percent of Sludge Cr Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	85	99	95	94	94	88
4	90	91	96	87	90	93
20	98	91	120	93	93	90
28	100	92	92	88	91	92
50	98	88	87	88	90	90

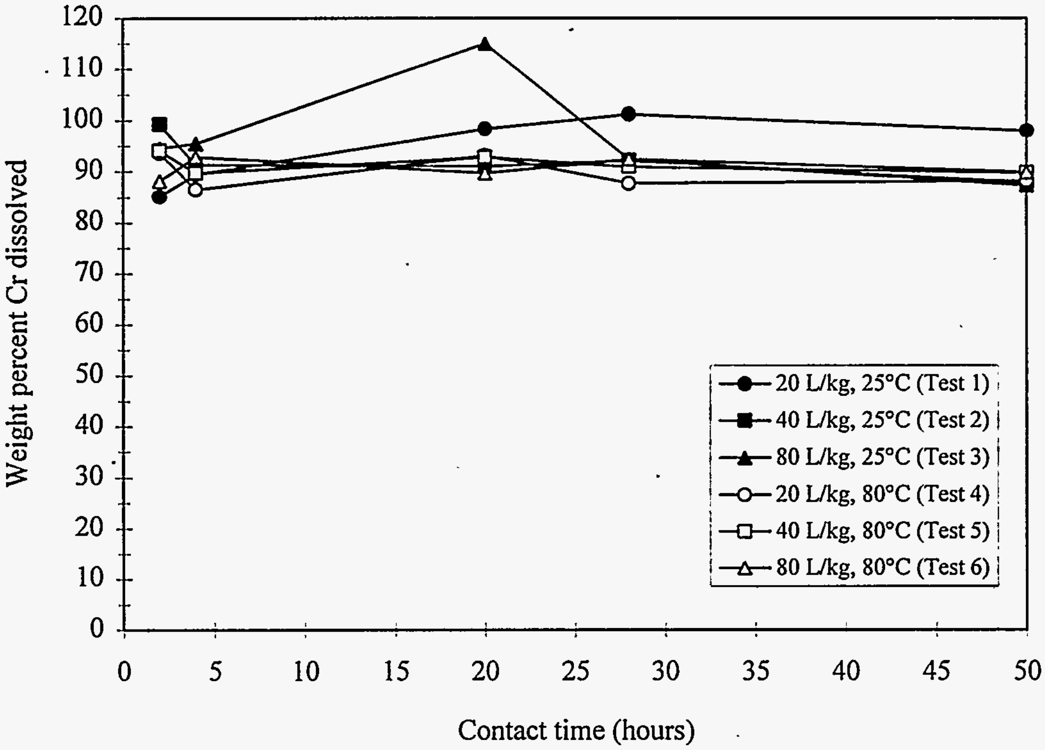

Figure I.4. Weight Percent of Sludge Cr Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.5. Weight Percent of Sludge Fe Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	88	93	99	96	100	92
4	93	93	98	90	97	99
20	87	95	98	98	99	99
28	90	97	90	92	95	100
50	89	95	93	89	96	95

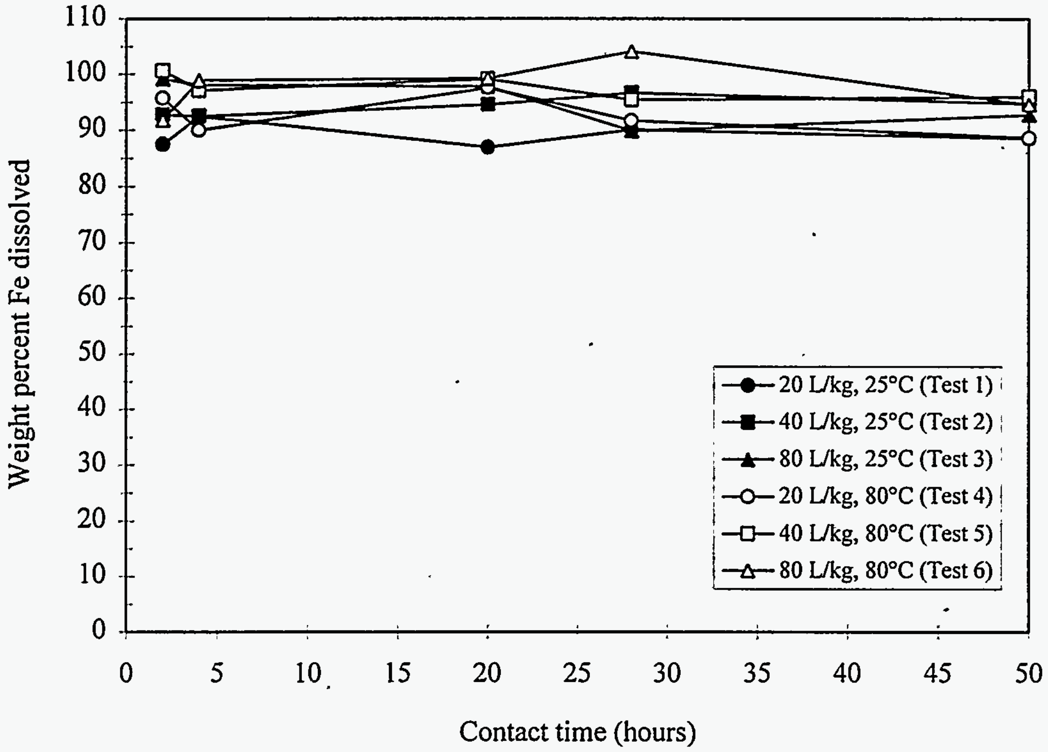

Figure I.5. Weight Percent of Sludge Fe Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.6. Weight Percent of Sludge K Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	79	88	89	95	97	90
4	82	92	93	86	90	100
20	81	88	85	93	95	90
28	84	92	95	86	91	99
50	80	86	83	86	92	85

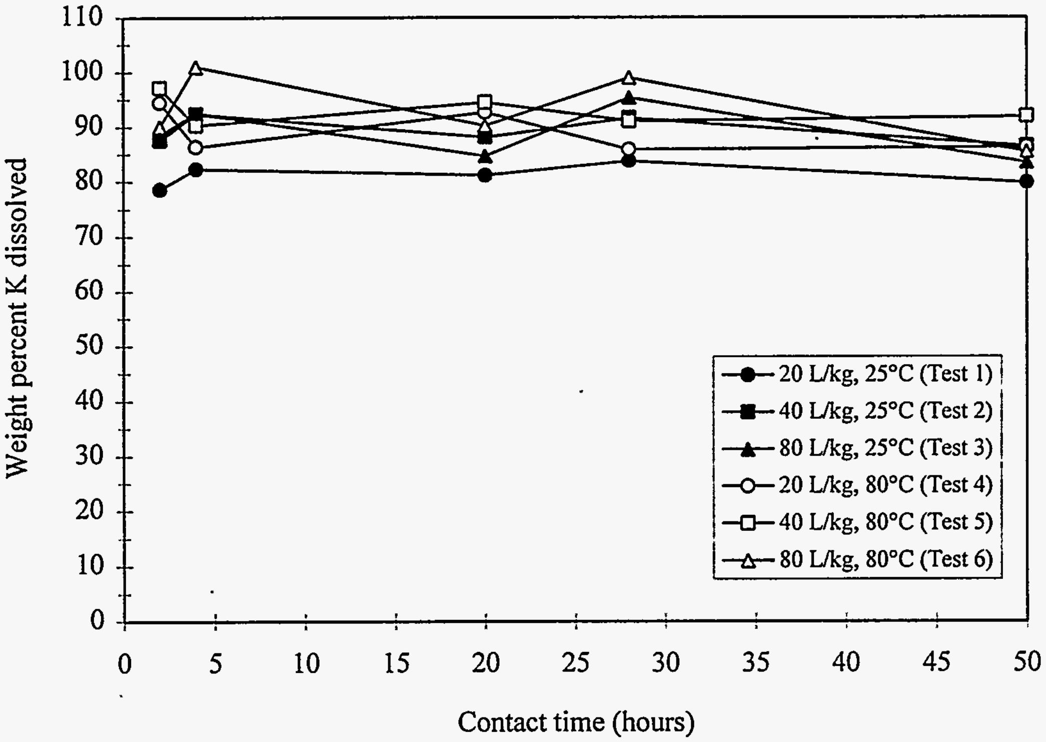

Figure I.6. Weight Percent of Sludge K Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.7. Weight Percent of Sludge Mg Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	82	74	38	98	110	32
4	84	73	38	99	86	47
20	89	79	30	100	86	40
28	94	78	46	94	85	52
50	91	73	36	97	79	35

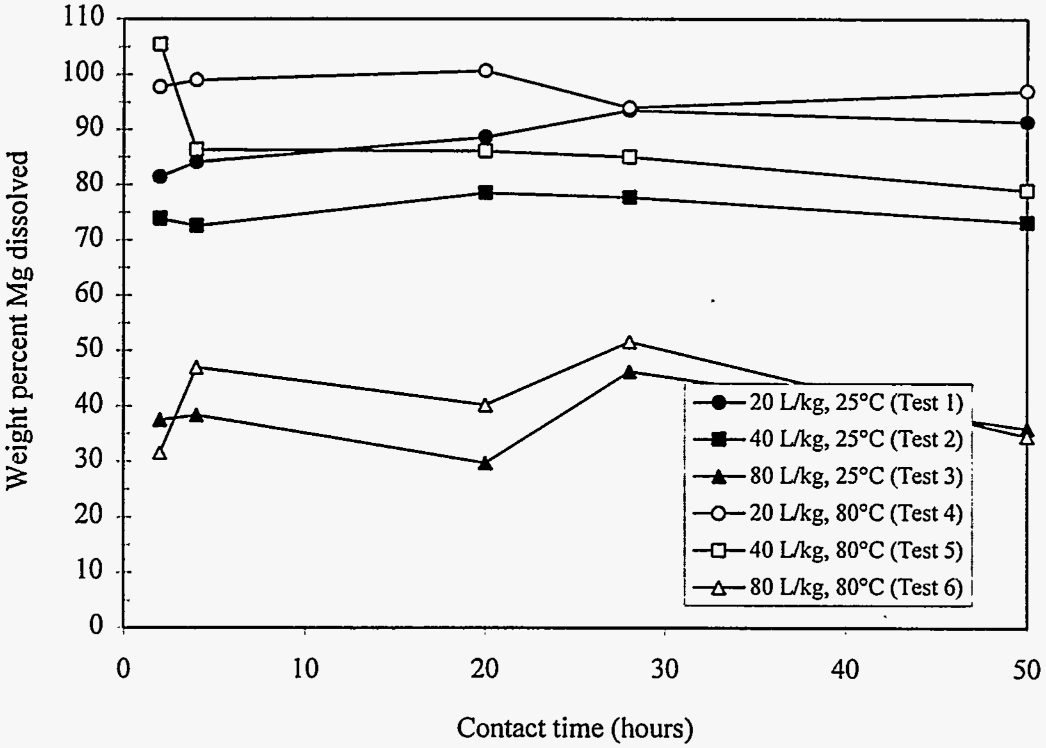

Figure I.7. Weight Percent of Sludge Mg Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.8. Weight Percent of Sludge Mn Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	81	88	95	92	97	87
4	86	91	95	87	95	95
20	78	94	91	95	95	94
28	85	95	96	86	92	96
50	83	90	90	86	92	88

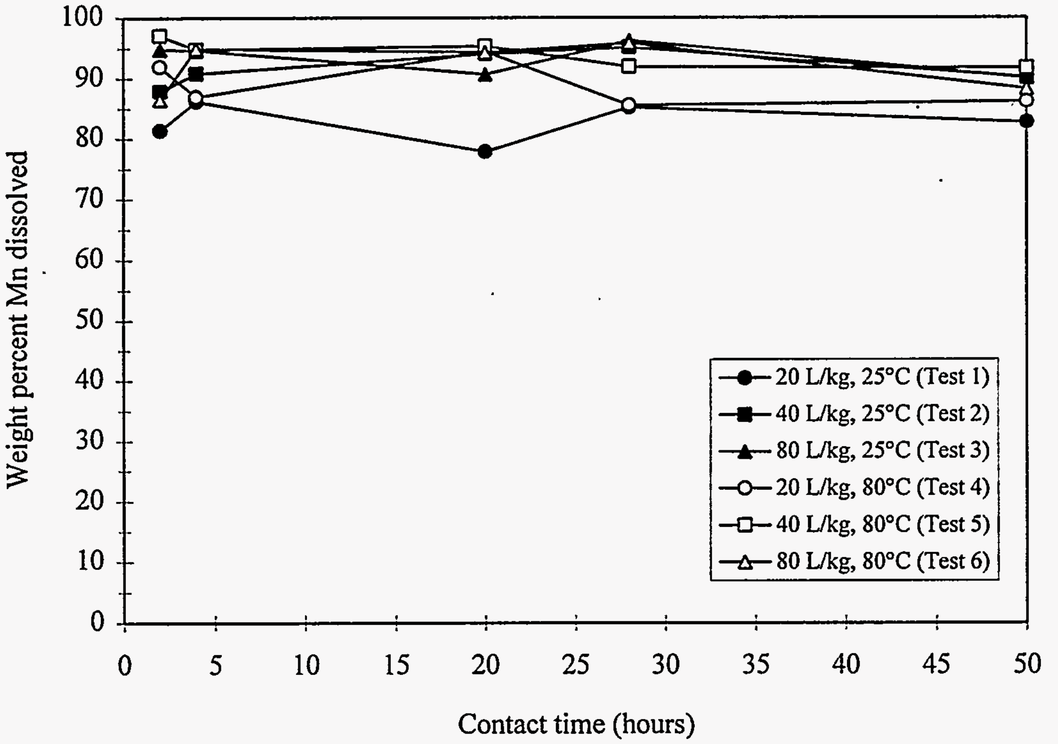

Figure I.8. Weight Percent of Sludge Mn Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.9. Weight Percent of Sludge Na Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	79	84	91	96	91	78
4	82	82	93	88	83	100
20	85	80	86	94	87	94
28	84	82	96	89	85	99
50	83	82	88	88	85	89

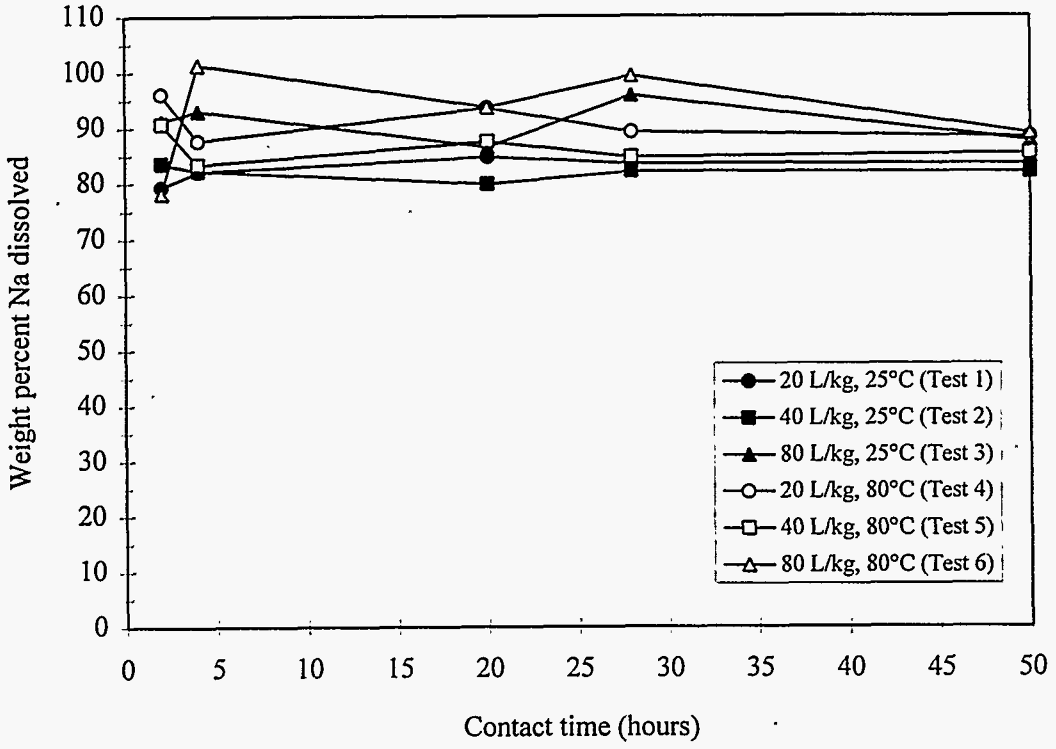

Figure I.9. Weight Percent of Sludge Na Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.10. Weight Percent of Sludge Ni Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	77	70	86	90	87	64
4	81	72	81	78	86	77
20	68	83	99	88	85	83
28	79	81	84	75	79	85
50	66	78	78	69	79	72

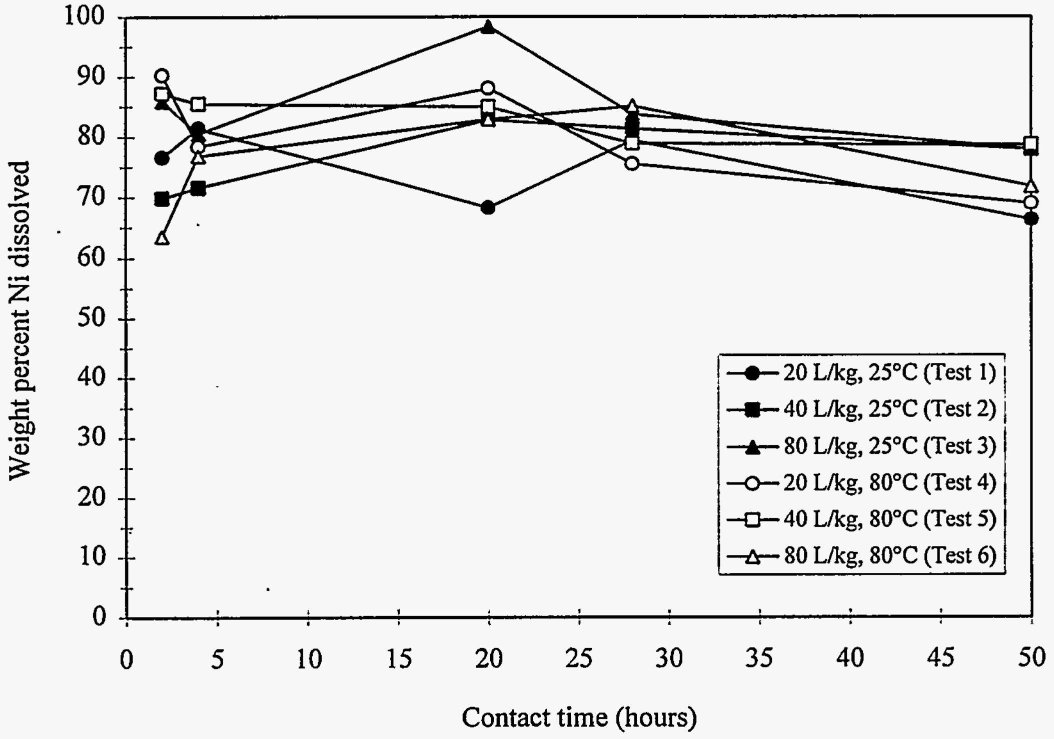

Figure I.10. Weight Percent of Sludge Ni Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.11. Weight Percent of Sludge Sr Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	80	100	100	110	110	100
4	93	95	110	84	100	110
20	78	94	100	110	100	100
28	88	97	100	80	99	110
50	85	100	97	95	100	100

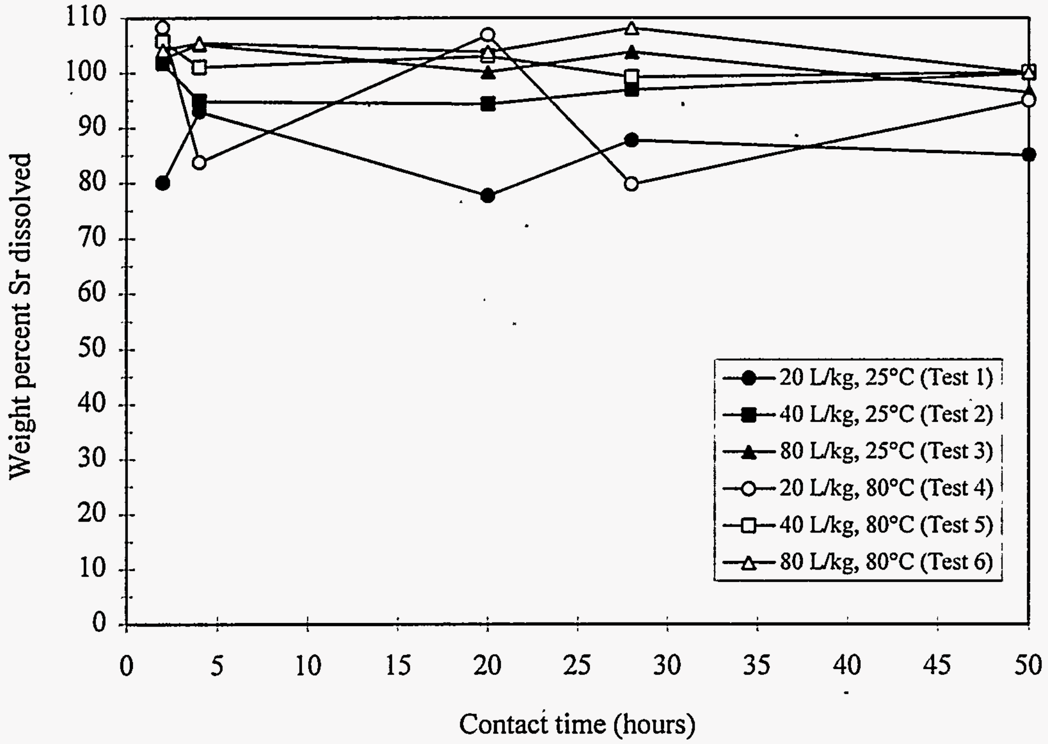

Figure I.11. Weight Percent of Sludge Sr Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.12. Weight Percent of Sludge Ti Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	90	82	90	82	100	83
4	82	82	87	81	93	88
20	81	83	85	89	92	87
28	83	100	77	83	91	92
50	81	84	80	81	89	83

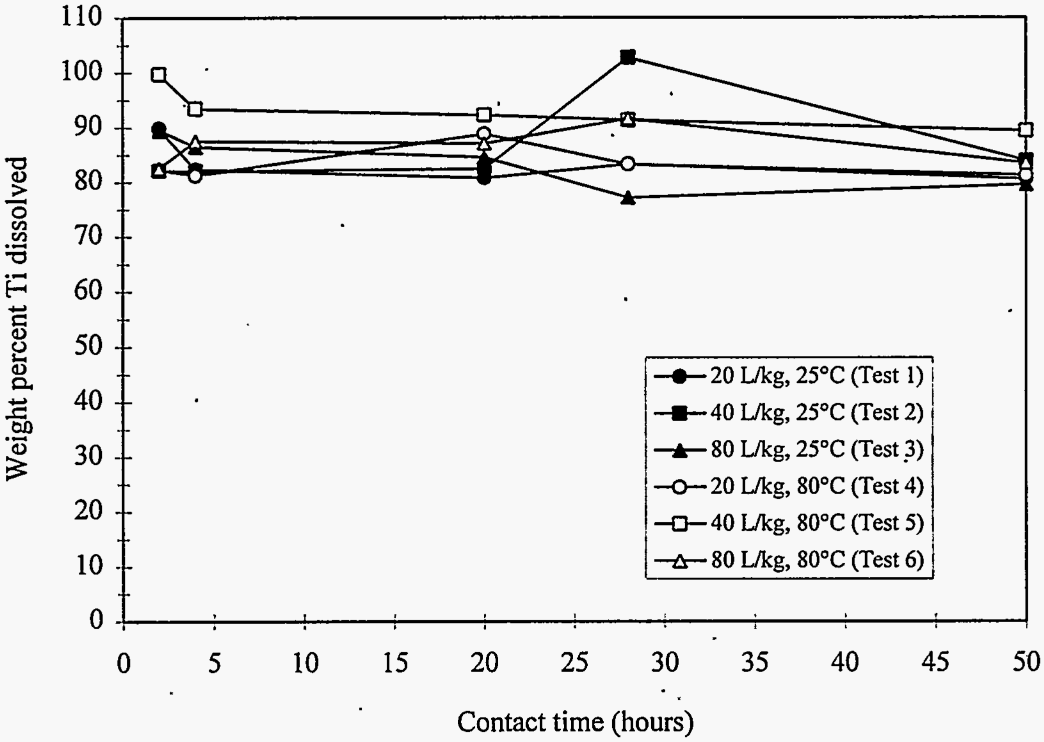

Figure I.12. Weight Percent of Sludge Ti Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Table I.13. Weight Percent of Sludge Zr Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

time (hours)	Test 1 20 L/kg 25°C	Test 2 40 L/kg 25°C	Test 3 80 L/kg 25°C	Test 4 20 L/kg 80°C	Test 5 40 L/kg 80°C	Test 6 80 L/kg 80°C
2	1.2	1.1	1.6	1.1	1.4	1.4
4	1.3	1.1	1.5	1.1	1.2	1.5
20	2.1	1.2	1.8	1.3	1.2	1.5
28	2.6	1.2	1.5	1.3	1.2	1.9
50	2.6	1.2	2.0	1.3	1.2	1.6

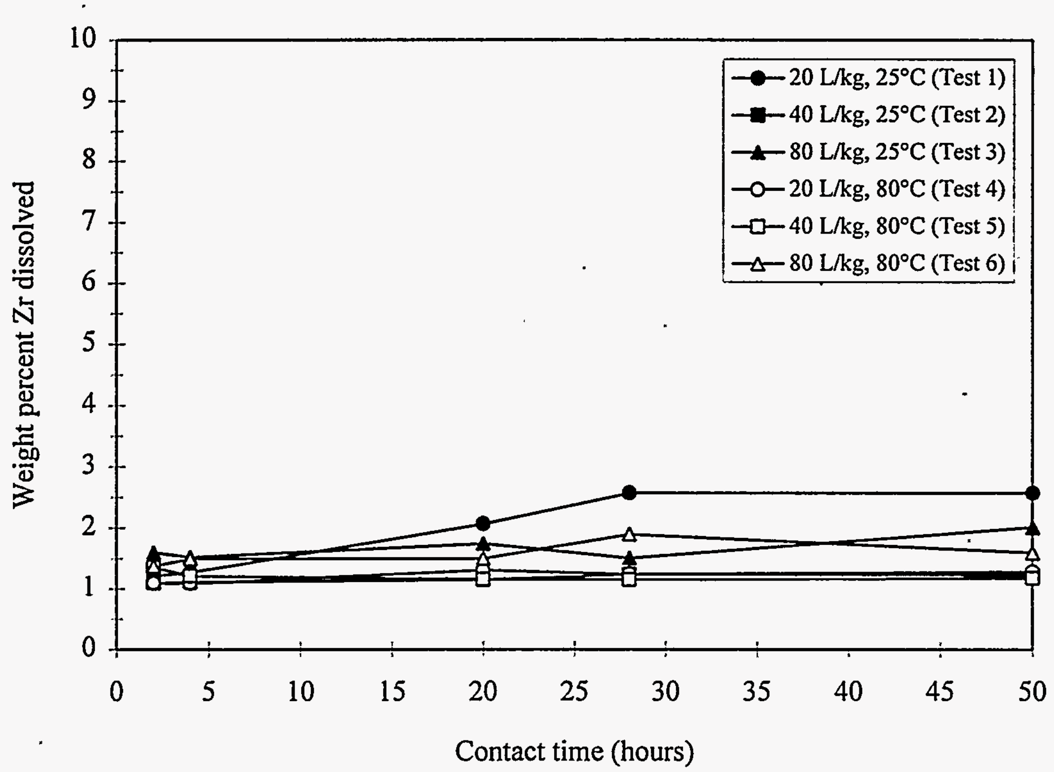

Figure I.13. Weight Percent of Sludge Zr Dissolved During Experiment to Determine Optimum Process Conditions for Sludge Dissolution

Appendix J

Sludge-Dissolution Experiments: Sample Calculations and Formulas Used in Determination of Percent Sludge Elements Dissolved

Table J.1. Sample Calculations and Formulas Used in Determination of Percent Sludge Element Dissolved in Experiment to Determine Optimum Process Conditions for Sludge Dissolution, Repeatability, and Long-Term Experiments

	A	B	C
1			
2	**Dissolution of Aluminum**	**Test 1**	
3	**Time 0 : initial conditions**		
4	initial amount of sludge simulant in reaction vessel, g	5.63	measured in laboratory
5	ug Al per g sludge simulant	6537	ICP-MS measurement
6	total amount Al initially present in vessel, ug	36803	=B4*B5
7	weight of oxalic acid initially present, g	114.64	measured in laboratory
8	total initial material weight, g	120.27	=B4+B7
9	weight loss per hour, g	0.09	measured in laboratory
10	final vessel weight	2116.0	measured in laboratory
11	empty vessel weight	2044.0	measured in laboratory
12	total finish weight	72.0	=B10-B11
13	**Time 1 = 2 hours**	2	measured in laboratory
14	weight of material present at T1, g	120.09	=B8-(B13*B9)
15	weight of sample removed at T1, g	9.77	measured in laboratory
16	ug Al per gram sample -- run 1	176	ICP-MS measurement
17	ug Al per gram sample -- run 2	177	ICP-MS measurement
18	average ug Al per gram sample	176.5	=(B16+B17)/2
19	amount Al in solution at T1, ug	21196	=B14*B18
20	**percentage Al dissolved at T1**	**57.59**	**=(B19/B6)*100**
21	**Time 2 = 4 hours**	4	measured in laboratory
22	weight of material present at T2, g	110.14	=B14-B15-(B21-B13)*B9
23	weight of sample removed at T2, g	8.44	measured in laboratory
24	ug Al per gram sample -- run 1	182	ICP-MS measurement
25	ug Al per gram sample -- run 2	210	ICP-MS measurement
26	average ug Al per gram sample	196	=(B24+B25)/2
27	amount of Al in solution at T2, ug	21587	=B22*B26
28	amount of Al removed during T1 sampling, ug	1724	=B15*B18
29	total amount of Al dissolved, ug	23312	=B27+B28
30	**percentage of Al dissolved at T2**	**63.34**	**=(B29/B6)*100**
31	**Time 3 = 20 hours**	20	measured in laboratory
32	weight of material present at T3, g	100.26	=B22-B23-(B31-B21)*B9
33	weight of sample removed at T3, g	8.60	measured in laboratory
34	ug Al per gram sample -- run 1	192	ICP-MS measurement
35	ug Al per gram sample -- run 2	251	ICP-MS measurement
36	average ug Al per gram sample	221.5	=(B34+B35)/2
37	amount of Al in solution at T3, ug	22208	=B32*B36
38	amount of Al removed during T1 sampling, ug	1724	=B28
39	amount of Al removed during T2 sampling, ug	1654	=B23*B26
40	total amount of Al removed during sampling, ug	3379	=SUM(B38:B39)

Table J.1, continued

	A	B	C
41	total amount of Al dissolved at T3, ug	25586	=B37+B40
42	**percentage of Al dissolved at T3**	**69.52**	**=(B41/B6)*100**
43	**Time 4 = 28 hours**	28	measured in laboratory
44	weight of material present at T4, g	90.94	=B32-B33-(B43-B31)*B9
45	weight of sample removed at T4, g	8.20	measured in laboratory
46	ug Al per gram sample -- run 1	206	ICP-MS measurement
47	ug Al per gram sample -- run 2	284	ICP-MS measurement
48	average ug Al per gram sample	245	=(B46+B47)/2
49	amount of Al in solution at T4, ug	22280	=B44*B48
50	amount of Al removed during T1 sampling, ug	1724	=B28
51	amount of Al removed during T2 sampling, ug	1654	=B39
52	amount of Al removed during T3 sampling, ug	1905	=B33*B36
53	total amount of Al removed during sampling, ug	5284	=SUM(B50:B52)
54	total amount of Al dissolved at T4, ug	27564	=B49+B53
55	**percentage of Al dissolved at T4**	**74.90**	**=(B54/B6)*100**
56	**Time 5 = 50 hours**	50	measured in laboratory
57	weight of material present at T5, g	80.76	=B44-B45-(B56-B43)*B9
58	weight of sample removed at T5, g	8.67	measured in laboratory
59	ug Al per gram sample -- run 1	215	ICP-MS measurment
60	ug Al per gram sample -- run 2	294	ICP-MS measurement
61	average ug Al per gram sample	255	=(B59+B60)/2
62	amount of Al in solution at T5, ug	20553	=B57*B61
63	amount of Al removed during T1 sampling, ug	1724	=B28
64	amount of Al removed during T2 sampling, ug	1654	=B39
65	amount of Al removed during T3 sampling, ug	1905	=B52
66	amount of Al removed during T4 sampling, ug	2009	=B45*B48
67	total amount of Al removed during sampling, ug	7293	=SUM(B63:B66)
68	total amount of Al dissolved at T5, ug	27846	=B62+B67
69	**percentage of Al dissolved at T5**	**75.66**	**=(B68/B6)*100**

Table J.2. Sample Calculations and Formulas Used in Determination of Percent Sludge Element Dissolved in Multiple-Contact Sludge-Dissolution Experiment in which Oxalic Acid was Replaced with Each Contact

	A	B	C
1			
2	Oxalic acid replaced after each contact	**Test 10**	
3	**Time 0 : initial conditions**		
4	initial amount of sludge simulant in reaction vessel, g	5.64	measured in laboratory
5	ug Al per g sludge simulant	6537	ICP-MS measurement
6	total amount Al initially present in vessel, ug	36869	=B4*B5
7	weight of oxalic acid initially present, g	229.3	measured in laboratory
8	weight of Fe_2O_3 present, g	0	Fe_2O_3 not used in this test
9	total initial material weight, g	234.94	=B4+B7
10	weight loss per hour, g	0	measured in laboratory
11	weight of loaded assembly	2236.0	measured in laboratory
12	weight of empty vessel	2001.1	measured in laboratory
13			
14	**Contact 1**		
15	amount of liquid present at T1 sampling	230.9	measured in laboratory
16	ug Al per gram sample -- run 1	111	ICP-MS measurement
17	ug Al per gram sample -- run 2	122	ICP-MS measurement
18	average ug Al per gram sample	116.5	=(B16+B17)/2
19	amount Al in solution at T1, ug	26894	=B15*B18
20	**percentage Al dissolved at T1**	**72.95**	=(B19/B6)*100
21	**Contact 2**		
22	amount of liquid present at T2 sampling, g	229.9	measured in laboratory
23	ug Al per gram sample -- run 1	3.30	ICP-MS measurement
24	ug Al per gram sample -- run 2	3.82	ICP-MS measurement
25	average ug Al per gram sample	3.56	=(B23+B24)/2
26	amount of Al in solution at T2, ug	819	=B22*B25
27	**percentage of Al dissolved at T2**	**2.22**	=(B26/B6)*100
28	**Contact 3**		
29	amount of liquid present at T3 sampling, g	235.2	measured in laboratory
30	ug Al per gram sample -- run 1	1.04	ICP-MS measurement
31	ug Al per gram sample -- run 2	1.03	ICP-MS measurement
32	average ug Al per gram sample	1.04	=(B30+B31)/2
33	amount of Al in solution at T3, ug	243	=B29*B32
34	**percentage of Al dissolved at T3**	**0.66**	=(B33/B6)*100
35	cumulative amounts dissolved:		
36	contact 1	72.95	=B20
37	contact 2	75.17	=B20+B27
38	contact 3	75.83	=B20+B27+B34

Table J.3. Sample Calculations and Formulas Used in Determination of Percent Sludge Element Dissolved in Multiple-Contact Sludge-Dissolution Experiment in which Oxalic Acid Remained After Each Contact

	A	B	C
1			
2	Oxalic acid remains after each contact	Test 11	
3	**Time 0 : initial conditions**		
4	initial amount of sludge simulant in reaction vessel, g	5.64	measured in laboratory
5	ug Al per g sludge simulant	6537	ICP-MS measurement
6	total amount Al initially present in vessel, ug	36869	=B4*B5
7	weight of oxalic acid initially present, g	229.3	measured in laboratory
8	weight of Fe_2O_3 present, g	0	Fe_2O_3 not used in this test
9	total initial material weight, g	234.94	=B4+B7
10	weight loss per hour, g	0	measured in laboratory
11	weight of loaded assembly	3151.9	measured in laboratory
12	weight of empty vessel	2917.0	measured in laboratory
13	**Contact 1**		
14	amount of liquid present at T1 sampling	230.0	measured in laboratory
15	amount of material removed at T1 sampling	10.56	measured in laboratory
16	ug Al per gram sample -- run 1	124	ICP-MS measurement
17	ug Al per gram sample -- run 2	130	ICP-MS measurement
18	average ug Al per gram sample	127	=(B16+B17)/2
19	amount Al in solution at T1, ug	29210	=B14*B18
20	**percentage Al dissolved at T1**	**79.23**	**=(B19/B6)*100**
21	**Contact 2**		
22	amount of liquid present at T2 sampling, g	440.2	measured in laboratory
23	amount of material removed at T2 sampling, g	10.44	measured in laboratory
24	ug Al per gram sample -- run 1	53.3	ICP-MS measurement
25	ug Al per gram sample -- run 2	72.0	ICP-MS measurement
26	average ug Al per gram sample	62.7	=(B24+B25)/2
27	amount of Al in solution at T2, ug	27579	=B22*B26
28	amount of Al removed during T1 sampling, ug	1341	=B15*B18
29	total amount of Al dissolved at T2, ug	28920	=B27+B28
30	**percentage of Al dissolved at T2**	**74.80**	**=(B27/B6)*100**

Table J.3, continued

	A	B	C
31	**Contact 3**		
32	amount of liquid present at T3 sampling, g	653.9	measured in laboratory
33	amount of material removed at T3 sampling, g	9.94	measured in laboratory
34	ug Al per gram sample -- run 1	38.9	ICP-MS measurement
35	ug Al per gram sample -- run 2	43.6	ICP-MS measurement
36	average ug Al per gram sample	41.3	=(B34+B35)/2
37	amount of Al in solution at T3, ug	26973	=B32*B36
38	amount of Al removed during T1 sampling, ug	1341	=B28
39	amount of Al removed during T2 sampling, ug	654	=B23*B26
40	total amount of Al dissolved at T3, ug	28969	=SUM(B37:B39)
42	**percentage of Al dissolved at T3**	73.16	=(B37/B6)*100
43	cumulative percentage dissolved:		
44	contact 1	79.23	=B20
45	contact 2	74.80	=B30
46	contact 3	73.16	=B42

Table J.4. Sample Calculations and Formulas Used in Determination of Percent Sludge Element Dissolved in Experiment to Asses the Effects of Condition Modifications

	A	B	C
1			
2		Test 12	
3	**Time 0 : initial conditions**		
4	initial amount of sludge simulant in reaction vessel, g	5.64	measured in laboratory
5	ug Al per g sludge simulant	6537	ICP-ES measurement
6	total amount Al initially present in vessel, ug	36869	=B4*B5
7	weight of oxalic acid initially present, g	229.3	measured in laboratory
8	weight of Fe_2O_3 present, g	0.096	measured in laboratory
9	total initial material weight, g	235.04	=B4+B7+B8
10	weight loss per hour, g	0.02	measured in laboratory
11	final vessel weight	2182.3	measured in laboratory
12	empty vessel weight	1996.0	measured in laboratory
13	total finish weight	186.3	=B11-B12
14	**Time 1 = 2 hours**	2	measured in laboratory
15	weight of material present at T1, g	235.00	=B9-(B14*B10)
16	weight of sample removed at T1, g	9.34	measured in laboratory
17	ug Al per gram sample -- run 1	103	ICP-MS measurement
18	ug Al per gram sample -- run 2		
19	average ug Al per gram sample	103	=B17
20	amount Al in solution at T1, ug	24205	=B15*B19
21	**percentage Al dissolved at T1**	**65.65**	**=(B20/B6)*100**
22	**Time 2 = 4 hours**	4	measured in laboratory
23	weight of material present at T2, g	225.62	=B15-B16-(B22-B14)*B10
24	weight of sample removed at T2, g	9.28	measured in laboratory
25	ug Al per gram sample -- run 1	110	ICP-MS measurement
26	ug Al per gram sample -- run 2		
27	average ug Al per gram sample	110	=B25
28	amount of Al in solution at T2, ug	24818	=B23*B27
29	amount of Al removed during T1 sampling, ug	962	=B16*B19
30	total amount of Al dissolved, ug	25780	=B28+B29
31	**percentage of Al dissolved at T2**	**69.92**	**=(B30/B6)*100**
32	**Time 3 = 20 hours**	20	measured in laboratory
33	weight of material present at T3, g	216.02	=B23-B24-(B32-B22)*B10
34	weight of sample removed at T3, g	9.27	measured in laboratory
35	ug Al per gram sample -- run 1	181	ICP-MS measurement
36	ug Al per gram sample -- run 2	157	ICP-MS measurement
37	average ug Al per gram sample	169	=(B35+B36)/2
38	amount of Al in solution at T3, ug	36507	=B33*B37
39	amount of Al removed during T1 sampling, ug	962	=B29
40	amount of Al removed during T2 sampling, ug	1021	=B24*B27
41	total amount of Al removed during sampling, ug	1983	=SUM(B39:B40)
42	total amount of Al dissolved at T3, ug	38490	=B38+B41
43	**percentage of Al dissolved at T3**	**104.40**	**=(B42/B6)*100**

Table J.4, continued

	A	B	C
44	**Time 4 = 28 hours**	28	measured in laboratory
45	weight of material present at T4, g	206.59	=B33-B34-(B44-B32)*B10
46	weight of sample removed at T4, g	10.44	measured in laboratory
47	ug Al per gram sample -- run 1	165	ICP-MS measurement
48	ug Al per gram sample -- run 2		
49	average ug Al per gram sample	165	=B47
50	amount of Al in solution at T4, ug	34087	=B45*B49
51	amount of Al removed during T1 sampling, ug	962	=B29
52	amount of Al removed during T2 sampling, ug	1021	=B40
53	amount of Al removed during T3 sampling, ug	1567	=B34*B37
54	total amount of Al removed during sampling, ug	3549	=SUM(B51:B53)
55	total amount of Al dissolved at T4, ug	37636	=B50+B54
56	**percentage of Al dissolved at T4**	**102.08**	**=(B55/B6)*100**
57	**Time 5 = 50 hours**	50	measured in laboratory
58	weight of material present at T5, g	195.71	=B45-B46-(B57-B44)*B10
59	weight of sample removed at T5, g	9.38	measured in laboratory
60	ug Al per gram sample -- run 1	142	ICP-MS measurement
61	ug Al per gram sample -- run 2		
62	average ug Al per gram sample	142	=B60
63	amount of Al in solution at T5, ug	27790	=B58*B62
64	amount of Al removed during T1 sampling, ug	962	=B29
65	amount of Al removed during T2 sampling, ug	1021	=B40
66	amount of Al removed during T3 sampling, ug	1567	=B53
67	amount of Al removed during T4 sampling, ug	1723	=B46*B49
68	total amount of Al removed during sampling, ug	5272	=SUM(B64:B67)
69	total amount of Al dissolved at T5, ug	33062	=B63+B68
70	**percentage of Al dissolved at T5**	**89.68**	**=(B69/B6)*100**

Appendix K

Sludge-Dissolution Tests to Assess the Effects of Condition Modifications: Data and Plots of Percentages of Sludge Elements Dissolved

Table K.1. Weight Percent of Sludge Al Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	66	88	80	70	110
4	70	75	110	92	71
20	100	76	80	73	75
28	100	100	78	72	110
50	90	110	85	70	91

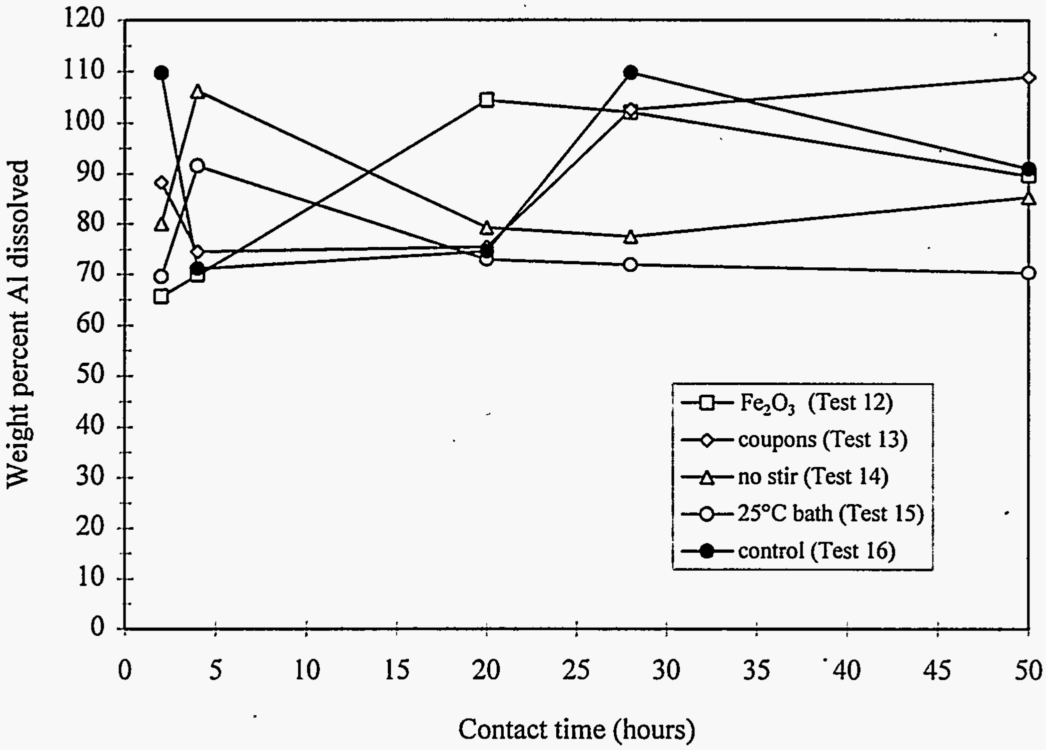

Figure K.1. Weight Percent of Sludge Al Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.2. Weight Percent of Sludge Ca Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	430	410	420	300	440
4	350	430	230	220	400
20	230	300	390	250	470
28	260	260	380	250	320
50	350	490	380	220	360

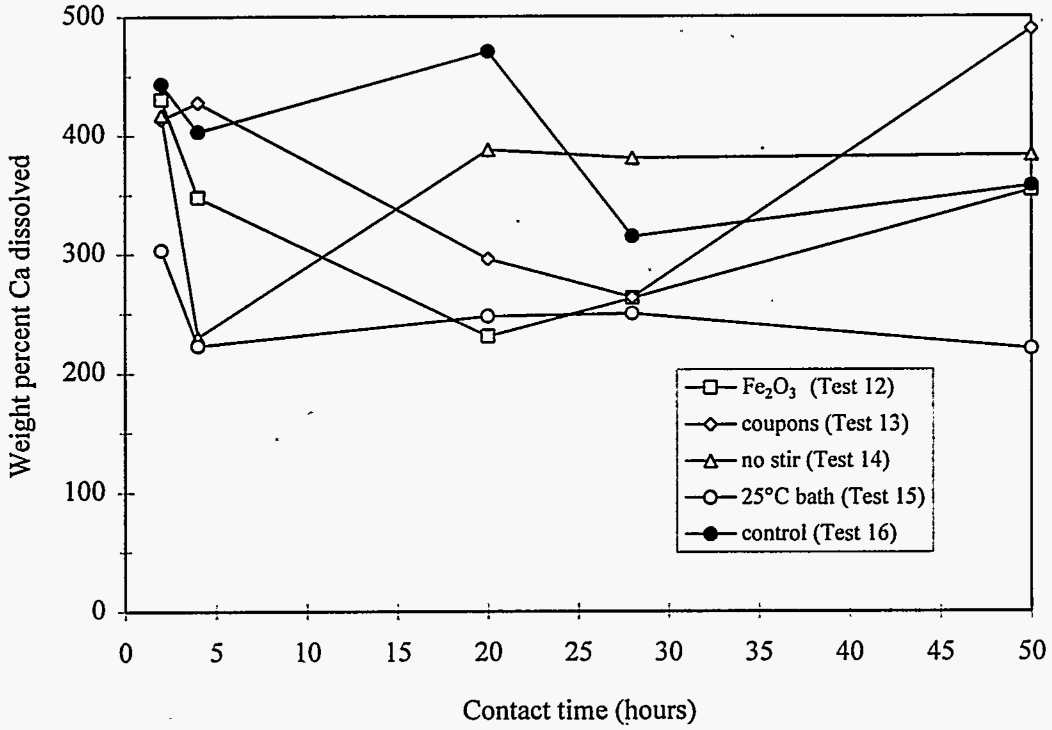

Figure K.2. Weight Percent of Sludge Ca Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.3. Weight Percent of Sludge Ce Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	3.4	2.2	3.0	1.9	1.7
4	1.9	2.2	1.3	2.6	1.5
20	1.6	1.8	1.4	2.1	1.4
28	1.5	1.5	1.5	2.9	1.8
50	3.9	2.0	1.7	2.3	2.0

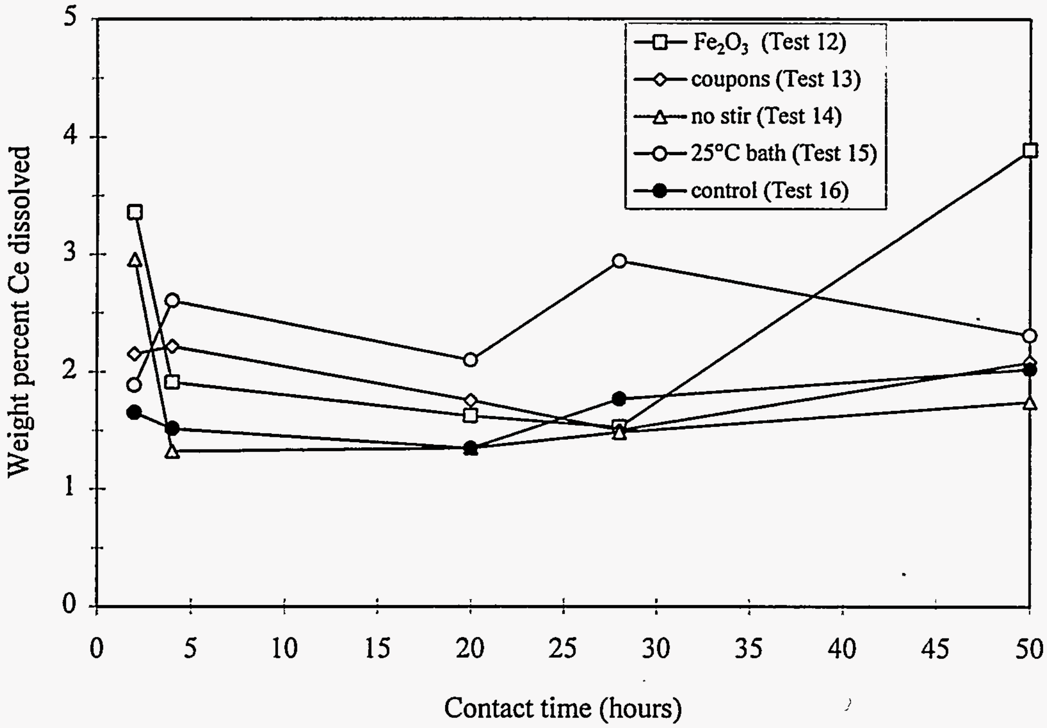

Figure K.3. Weight Percent of Sludge Ce Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.4. Weight Percent of Sludge Cr Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	86	84	88	87	86
4	89	89	86	80	81
20	90	90	84	84	90
28	82	90	91	86	90
50	84	93	88	83	86

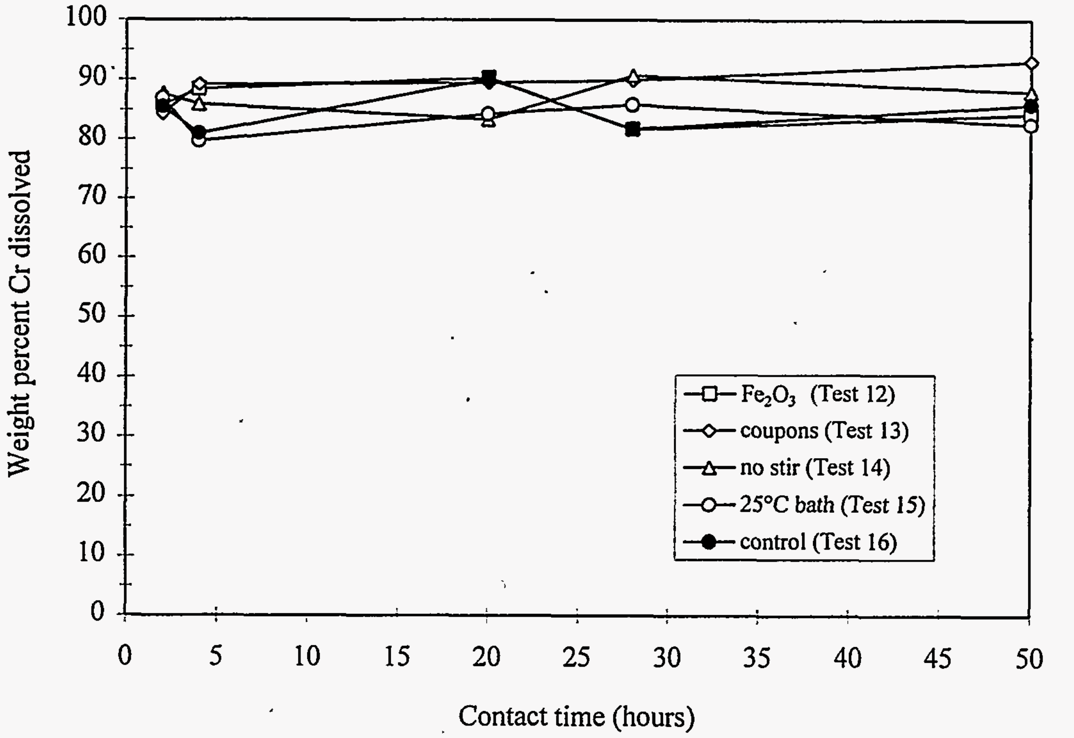

Figure K.4. Weight Percent of Sludge Cr Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.5. Weight Percent of Sludge Fe Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	79	110	86	100	92
4	77	94	86	100	96
20	82	120	100	100	110
28	87	130	93	96	120
50	97	160	98	98	96

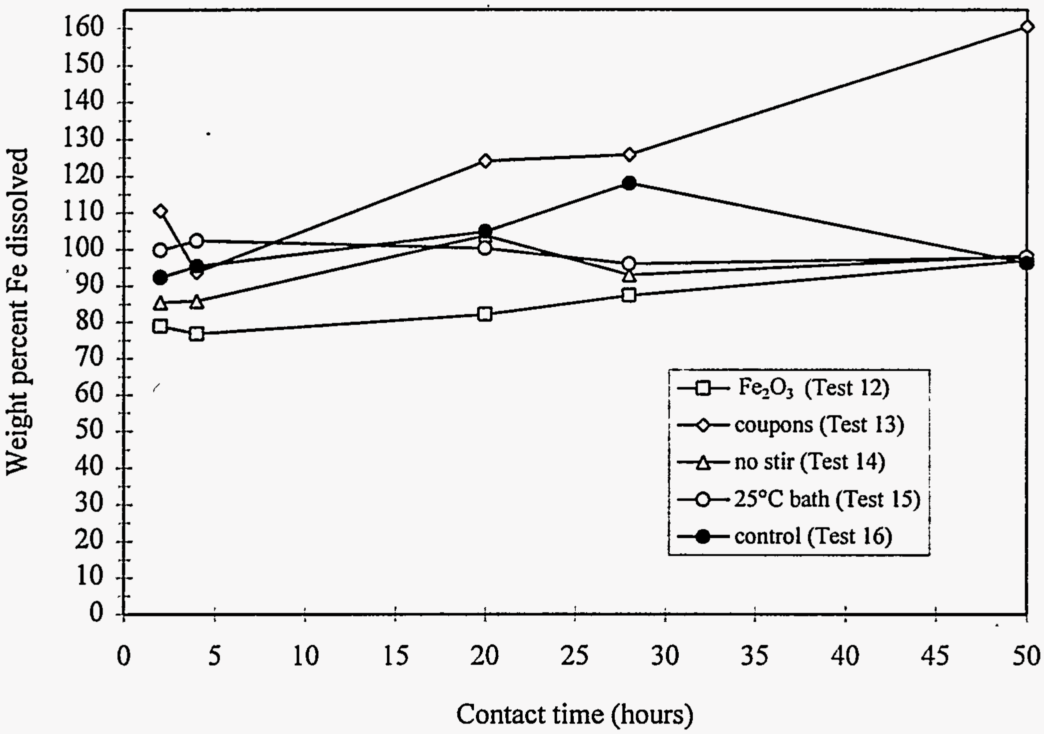

Figure K.5. Weight Percent of Sludge Fe Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.6. Weight Percent of Sludge K Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	70	70	71	73	71
4	71	73	72	69	68
20	72	72	69	70	74
28	69	72	75	72	67
50	69	75	74	71	69

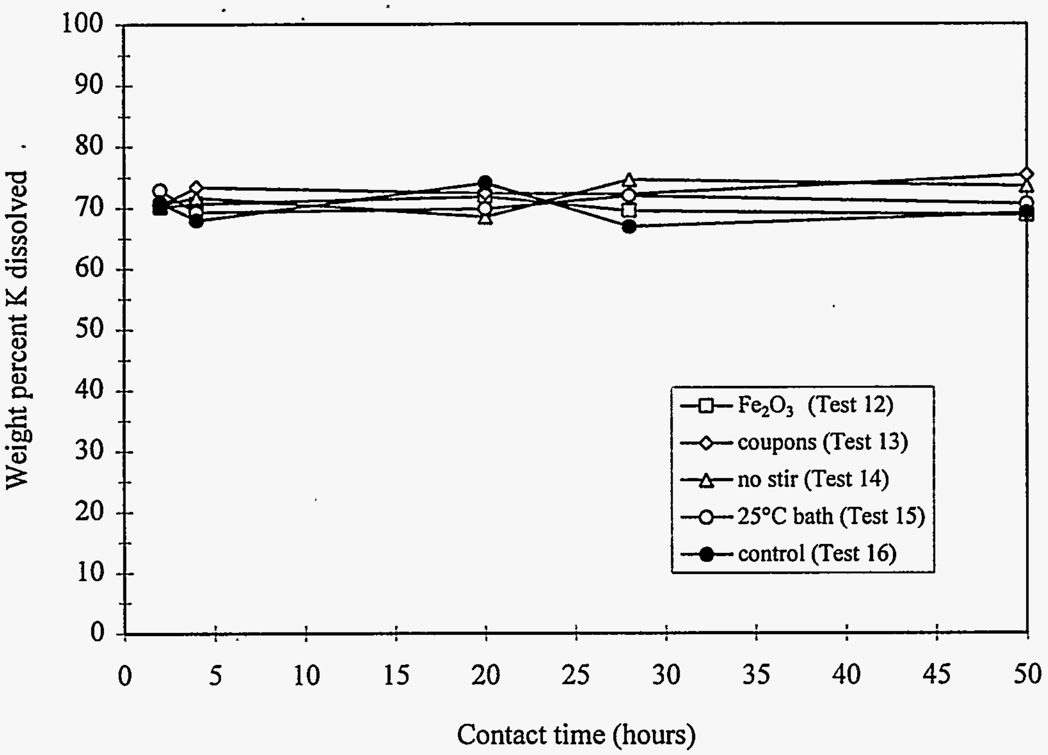

Figure K.6. Weight Percent of Sludge K Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.7. Weight Percent of Sludge Mg Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	86	86	88	83	84
4	86	90	87	77	82
20	86	89	86	83	90
28	84	87	91	85	85
50	84	91	93	82	87

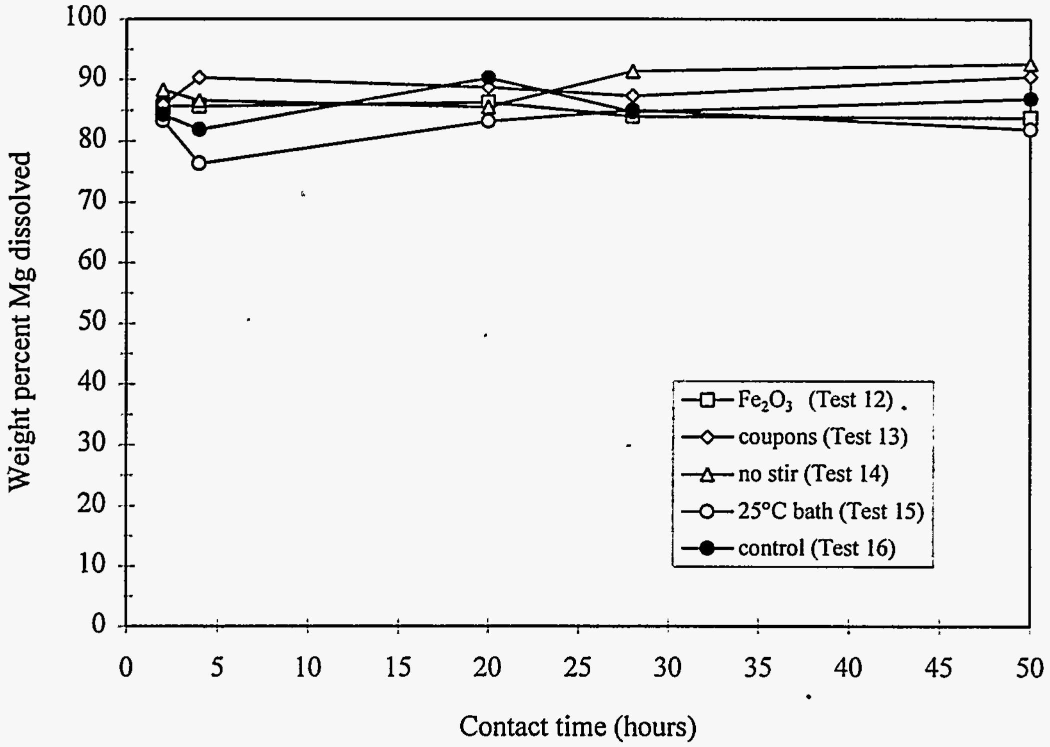

Figure K.7. Weight Percent of Sludge Mg Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.8. Weight Percent of Sludge Mn Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	99	110	94	100	95
4	93	85	99	90	94
20	98	91	110	99	98
28	96	90	95	95	110
50	110	100	100	97	100

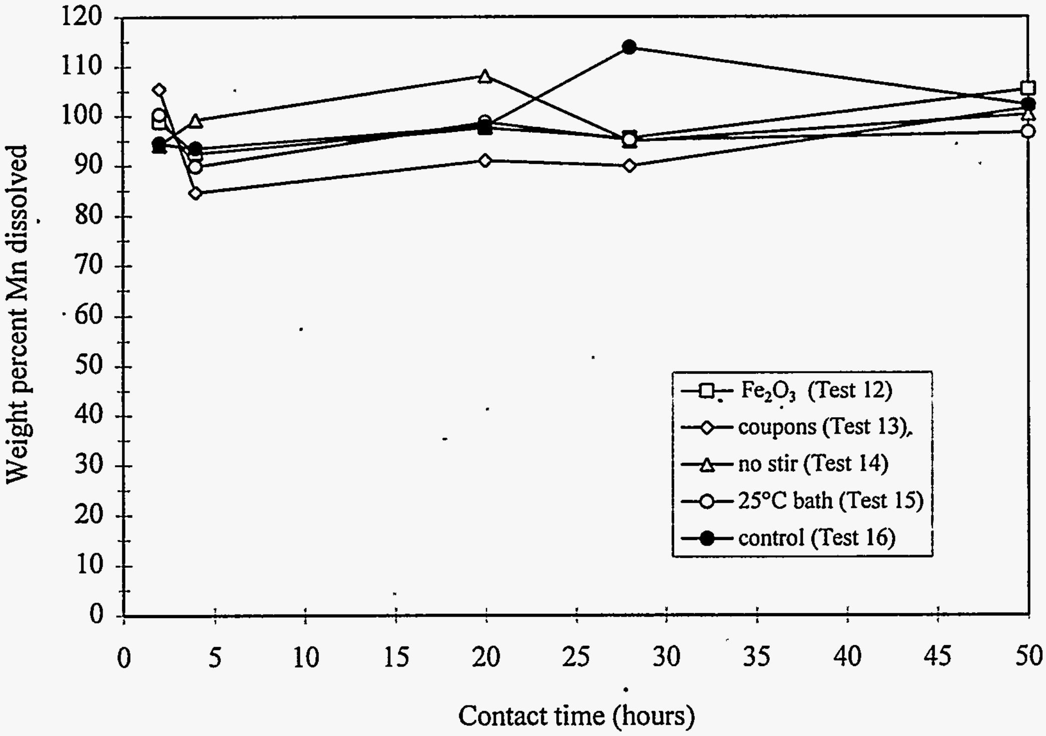

Figure K.8. Weight Percent of Sludge Mn Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.9. Weight Percent of Sludge Na Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	120	110	82	110	95
4	99	93	100	100	93
20	100	98	110	100	91
28	104	90	96	96	120
50	100	110	100	97	110

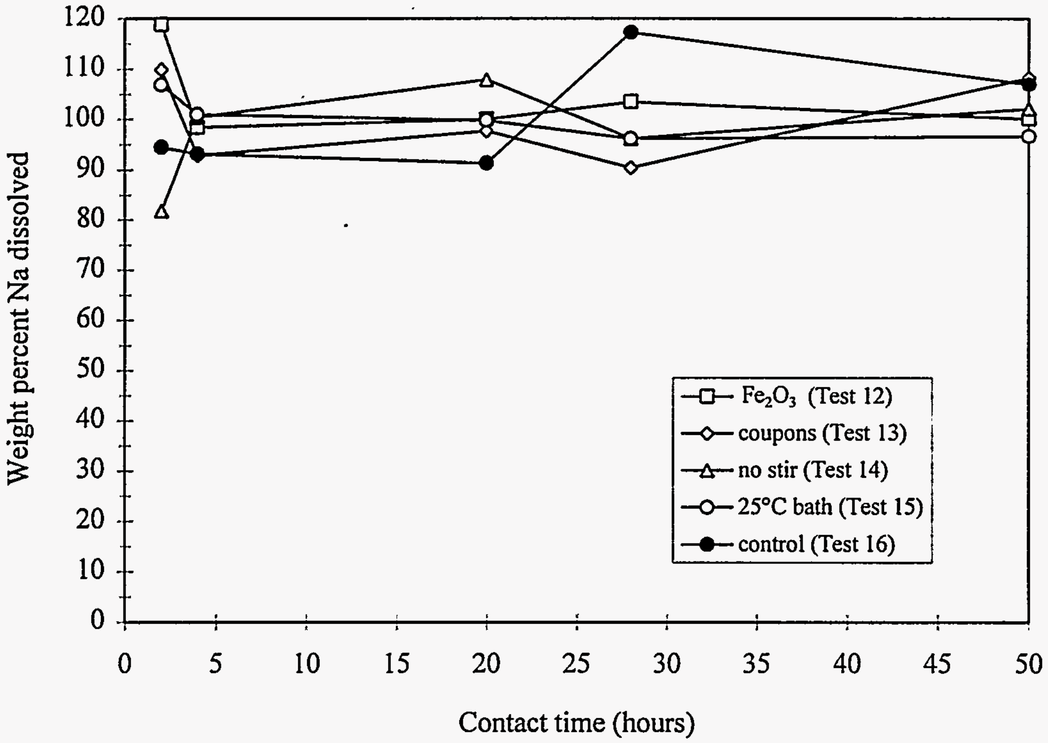

Figure K.9. Weight Percent of Sludge Na Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.10. Weight Percent of Sludge Ni Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	82	66	78	14	37
4	75	70	27	70	64
20	73	35	31	72	50
28	35	30	49	82	79
50	75	27	56	49	75

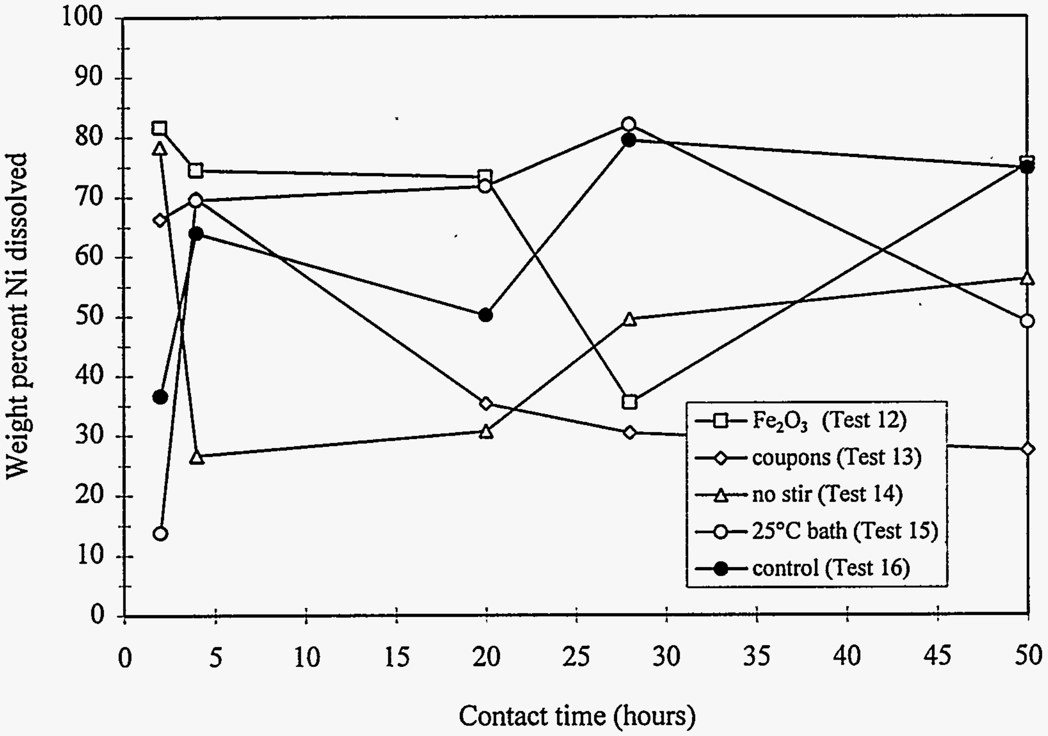

Figure K.10. Weight Percent of Sludge Ni Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.11. Weight Percent of Sludge Sr Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	100	110	98	71	79
4	100	95	97	86	88
20	63	96	100	56	96
28	110	97	97	54	81
50	100	100	100	42	83

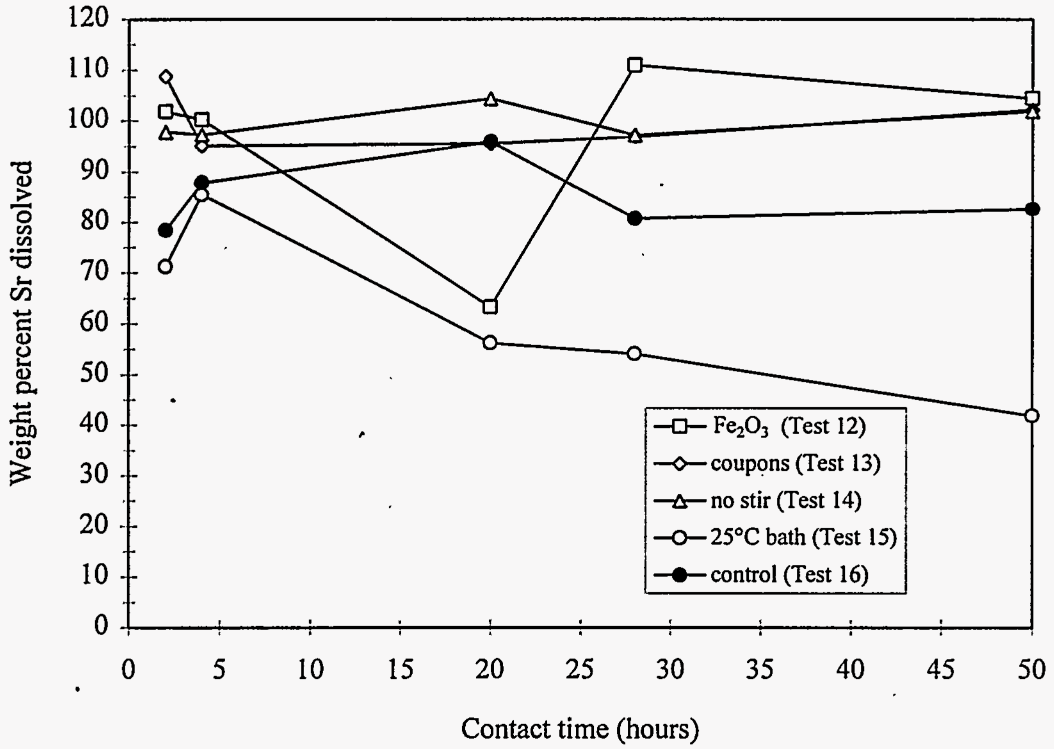

Figure K.11. Weight Percent of Sludge Sr Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.12. Weight Percent of Sludge Ti Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	62	65	63	63	63
4	62	66	68	60	64
20	68	66	64	61	68
28	66	63	72	64	60
50	66	70	69	61	66

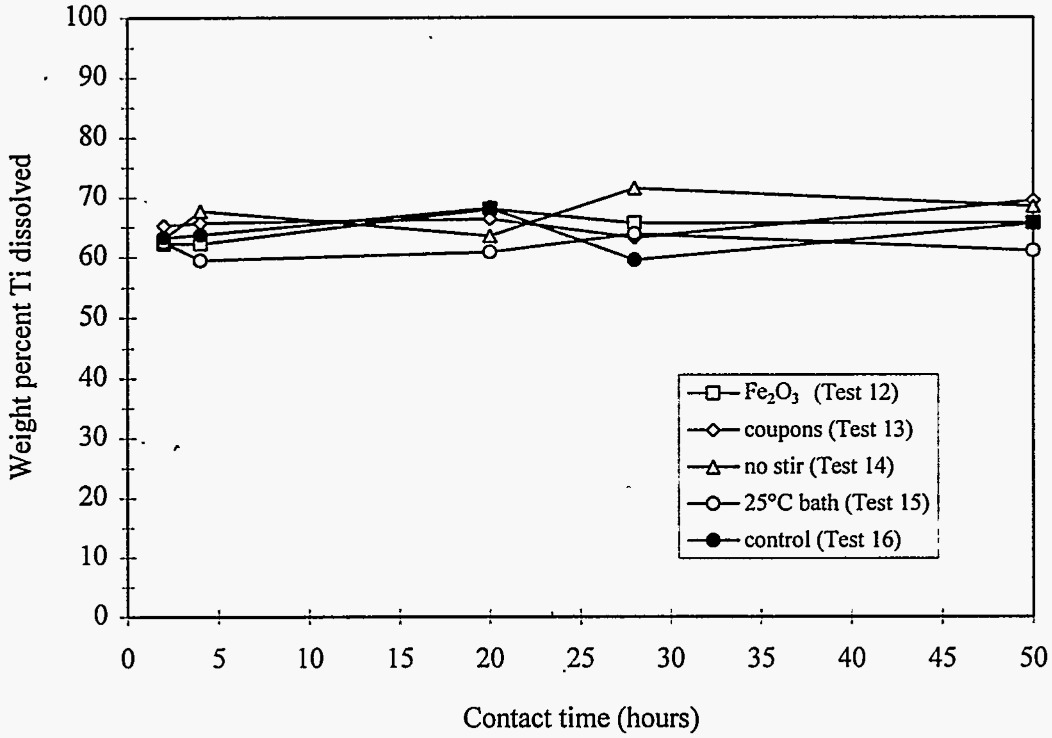

Figure K.12. Weight Percent of Sludge Ti Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Table K.13. Weight Percent of Sludge Zr Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Time (hours)	Test 12 Fe_2O_3	Test 13 Coupons	Test 14 No Stir	Test 15 25°C Bath	Test 16 Control
2	0.96	0.90	0.95	0.86	0.78
4	0.93	0.99	0.82	0.86	0.79
20	1.0	0.88	0.81	1.0	0.85
28	0.90	0.93	0.89	0.83	0.79
50	0.96	1.1	0.95	0.98	0.86

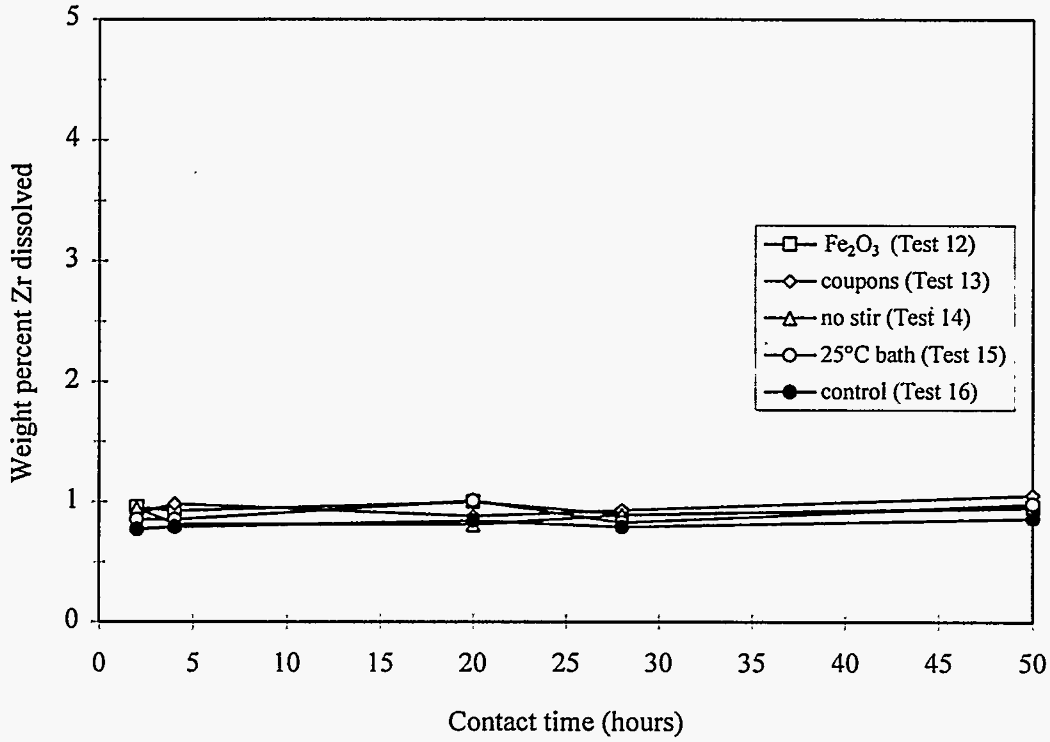

Figure K.13. Weight Percent of Sludge Zr Dissolved During Experiment to Determine Effects of Condition Modifications on Sludge Dissolution

Appendix L

Weight Loss-Derived Corrosion Rate Data and Pit Depths (Measured and Estimated) for Mild-Steel Corrosion Test Specimens

Table L.1. Corrosion Rate Data and Pitting Measurements for Mild-Steel Test Specimens

Vessel Number	Specimen ID Number	Wt% Acid	Exposure (days)	Specimen Position	Corrosion Rate (mpy*)	Corrosion Rate (mm/yr)	Pit Depths (mils)	Pit Depths (mm)	Pit Population
1	466	4	7	vapor space	8	0.20	2 - 4	0.05 - 0.10	very few
1	480	4	7	vapor space	7	0.18	2 - 4	0.05 - 0.10	very few
1	474	4	7	submerged	37	0.94	~ 2	~ 0.05	many
1	458	4	7	submerged	41	1.04	~ 2	~ 0.05	many
1	500	4	14	vapor space	4	0.10	< 1	< 0.02	few
1	419	4	14	vapor space	2	0.05	< 1	< 0.02	few
1	493	4	14	submerged	107	2.72	8 - 10	0.20 - 0.25	many
1	439	4	14	submerged	100	2.54	8 - 10	0.20 - 0.25	many
1	462	4	21	vapor space	2	0.05	< 1	< 0.02	few
1	478	4	21	vapor space	1	0.02	< 1	< 0.02	few
1	468	4	21	interface	120	3.05	8 - 10	0.20 - 0.25	many
1	476	4	21	interface	120	3.05	8 - 10	0.20 - 0.25	many
1	457	4	21	submerged	126	3.20	8 - 10	0.20 - 0.25	many
1	459	4	21	submerged	130	3.30	8 - 10	0.20 - 0.25	many
2	456	4	7	vapor space	0	0.00	--	--	none
2	455	4	7	vapor space	3	0.08	--	--	none
2	484	4	7	submerged	103	2.62	~ 6	~ 0.15	many
2	490	4	7	submerged	118	3.00	~ 6	~ 0.15	many
2	491	4	14	vapor space	1	0.02	~ 1	~ 0.02	few
2	420	4	14	vapor space	5	0.13	~ 1	~ 0.02	few
2	498	4	14	submerged	110	2.79	8 - 10	0.20 - 0.25	many
2	440	4	14	submerged	75	1.91	8 - 10	0.20 - 0.25	many
2	451	4	21	vapor space	1	0.02	< 1	< 0.02	few
2	465	4	21	vapor space	2	0.05	< 1	< 0.02	few
2	461	4	21	interface	147	3.73	10 - 15	0.25 - 0.38	many
2	463	4	21	interface	168	4.27	10 - 15	0.25 - 0.38	many
2	473	4	21	submerged	165	4.19	10 - 15	0.25 - 0.38	many
2	460	4	21	submerged	164	4.17	10 - 15	0.25 - 0.38	many

* mpy = mils per year

Table L.1, cont.

Vessel Number	Specimen ID Number	Wt% Acid	Exposure (days)	Specimen Position	Corrosion Rate (mpy*)	(mm/yr)	Pit Depths (mils)	(mm/yr)	Pit Population
3	464	8	7	vapor space	1	0.02	--	--	none
3	469	8	7	vapor space	1	0.02	--	--	none
3	481	8	7	submerged	30	0.76	1 - 2	0.02 - 0.05	moderate
3	472	8	7	submerged	30	0.76	1 - 2	0.02 - 0.05	moderate
3	494	8	14	vapor space	0	0.00	< 1	< 0.02	few
3	418	8	14	vapor space	0	0.00	< 1	< 0.02	few
3	492	8	14	submerged	17	0.43	< 1	< 0.02	many
3	417	8	14	submerged	16	0.41	< 1	< 0.02	many
3	485	8	21	vapor space	1	0.02	< 1	< 0.02	few
3	489	8	21	vapor space	0	0.00	< 1	< 0.02	few
3	452	8	21	interface	14	0.36	< 1	< 0.02	many
3	470	8	21	interface	15	0.38	< 1	< 0.02	many
3	486	8	21	submerged	16	0.41	< 1	< 0.02	many
3	454	8	21	submerged	16	0.41	< 1	< 0.02	many
4	482	8	7	vapor space	8	0.20	1 - 2	0.02 - 0.05	very few
4	453	8	7	vapor space	4	0.10	1 - 2	0.02 - 0.05	very few
4	483	8	7	submerged	47	1.19	2 - 4	0.05 - 0.10	many
4	487	8	7	submerged	47	1.19	2 - 4	0.05 - 0.10	many
4	497	8	14	vapor space	0	0.00	--	--	none
4	438	8	14	vapor space	0	0.00	--	--	none
4	495	8	14	submerged	19	0.48	< 1	< 0.02	moderate
4	437	8	14	submerged	20	0.51	< 1	< 0.02	moderate
4	475	8	21	vapor space	1	0.02	--	--	none
4	488	8	21	vapor space	0	0.00	--	--	none
4	477	8	21	interface	17	0.43	~ 1	~ 0.02	few
4	467	8	21	interface	18	0.46	~ 1	~ 0.02	few
4	479	8	21	submerged	20	0.51	~ 1	~ 0.02	few
4	471	8	21	submerged	20	0.51	~ 1	~ 0.02	few

* mpy = mils per year

Appendix M

Photographs of (Post-Test) Mild-Steel Corrosion Specimens

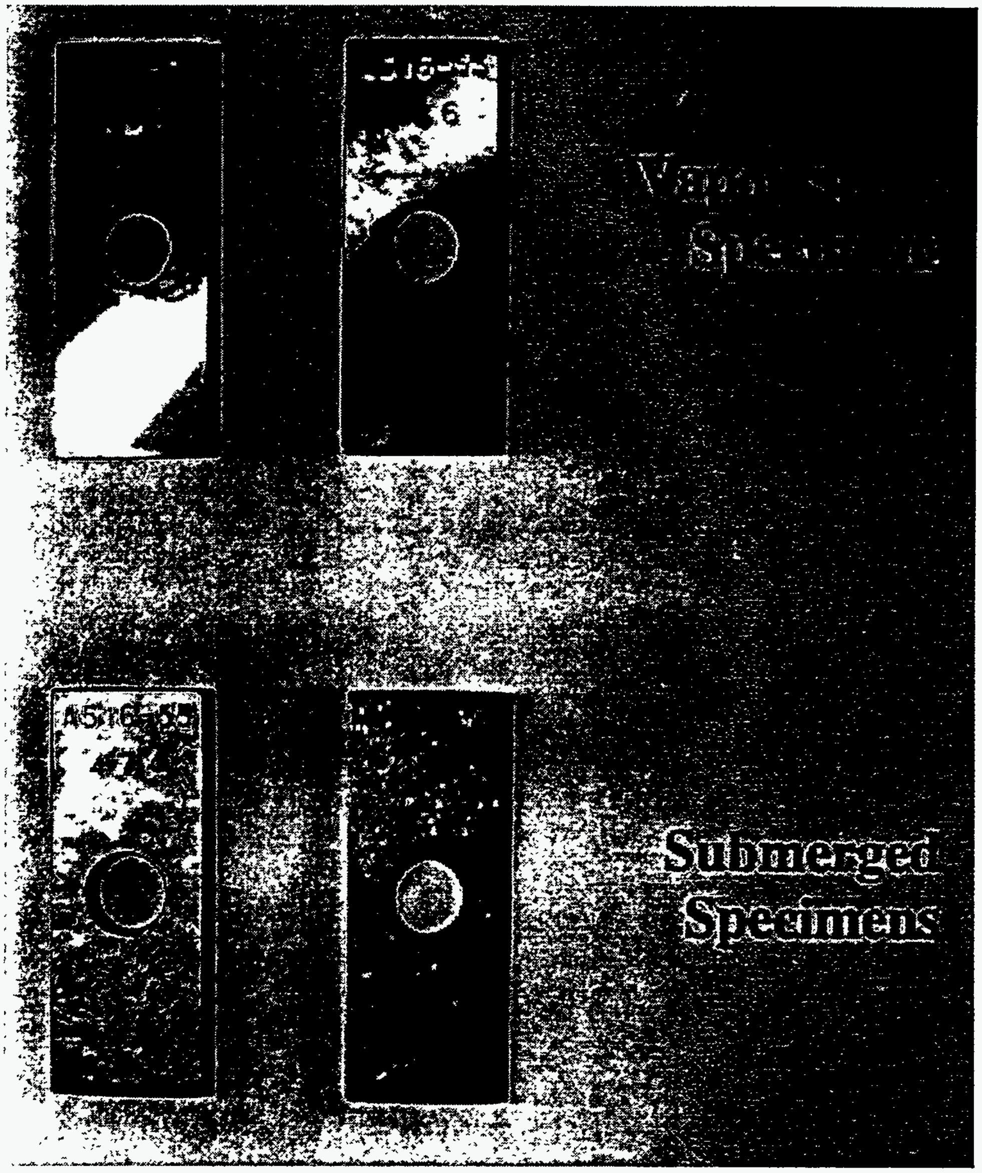

Figure M.1. Mild-Steel Specimens Removed From Vessel 1 (4 wt% Oxalic Acid) After One Week Exposure

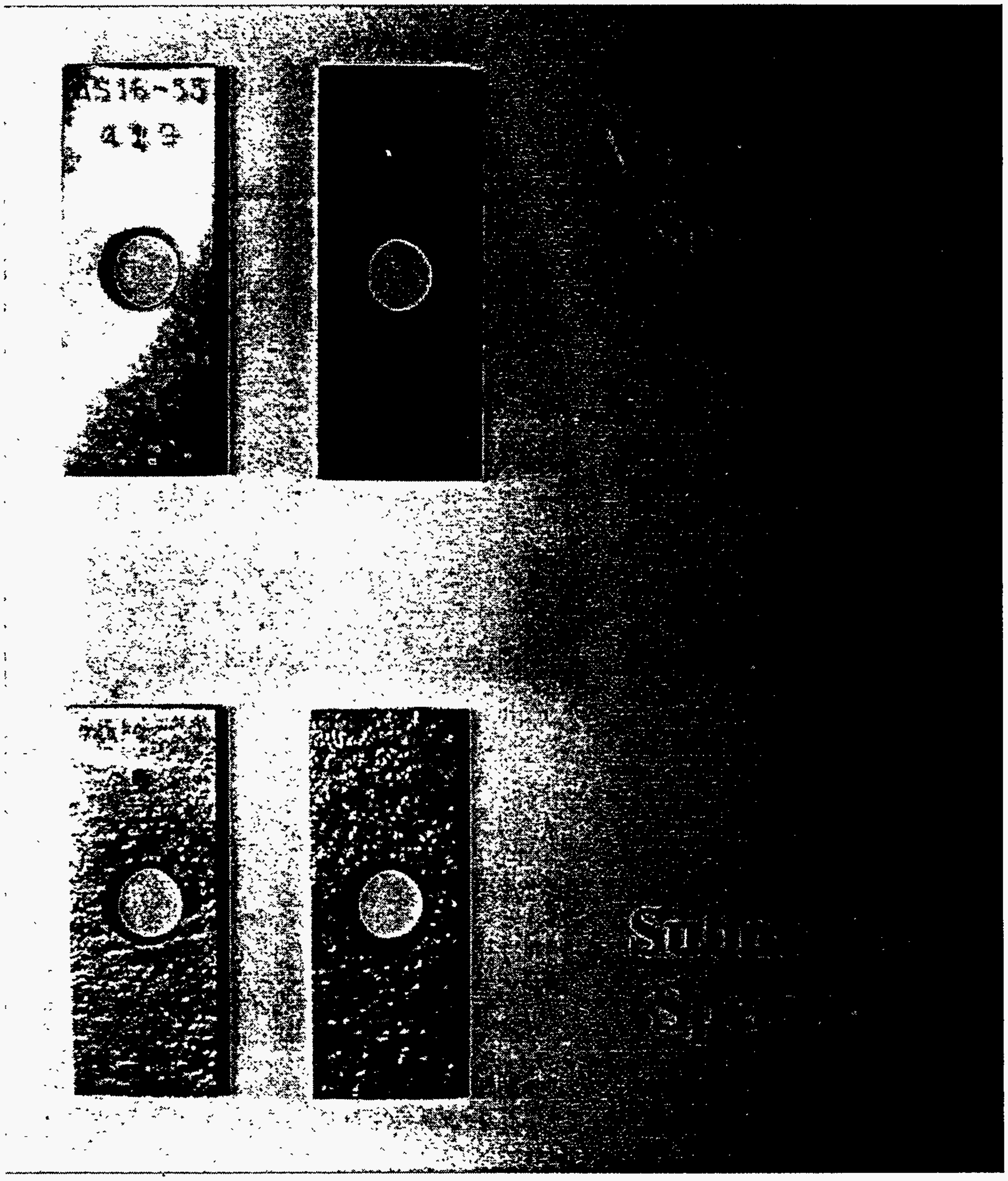

Figure M.2. Mild-Steel Specimens Removed from Vessel 1 (4 wt% Oxalic Acid) After Two Weeks Exposure

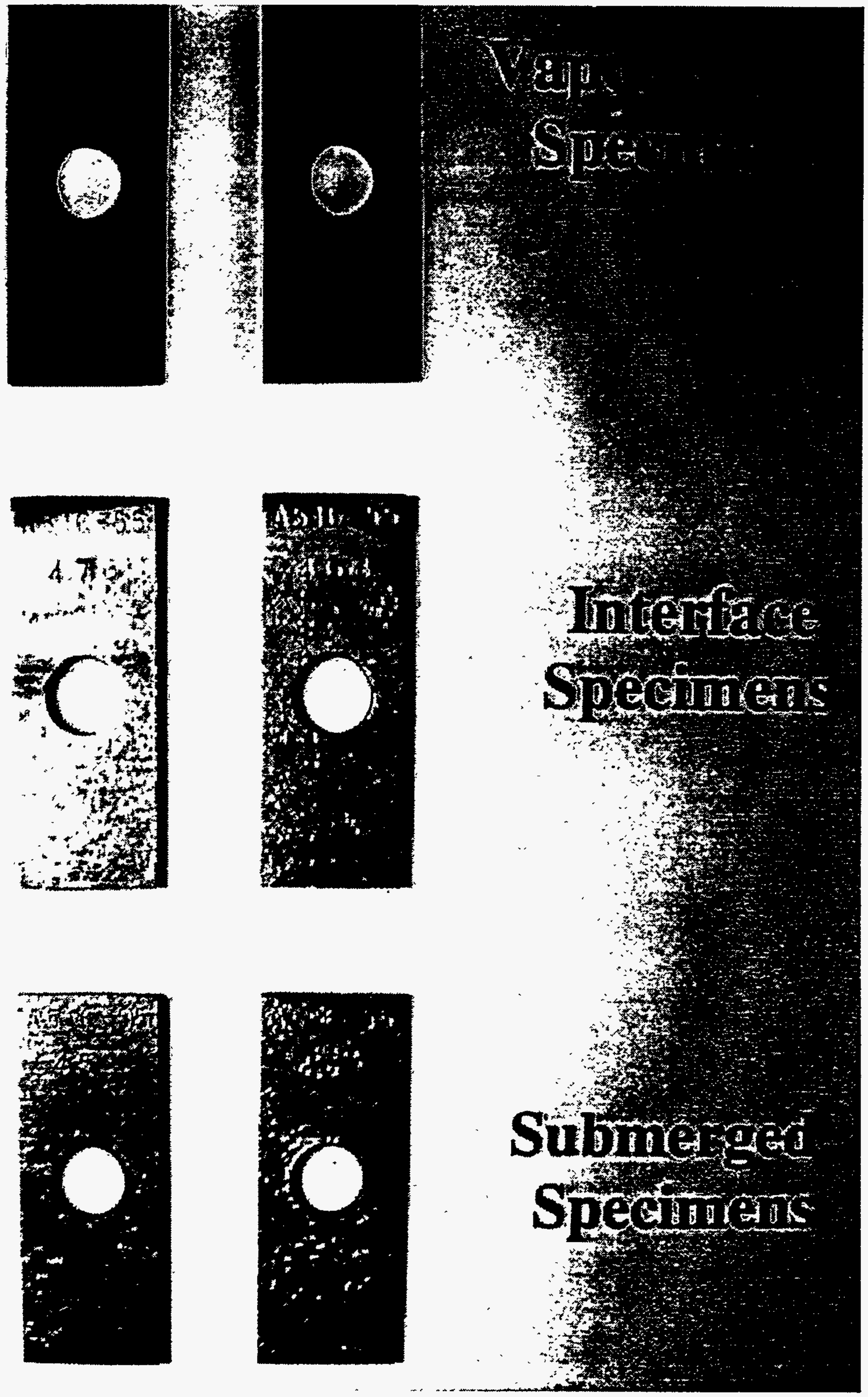

Figure M.3. Mild-Steel Specimens Removed from Vessel 1 (4 wt% Oxalic Acid) After Three Weeks Exposure

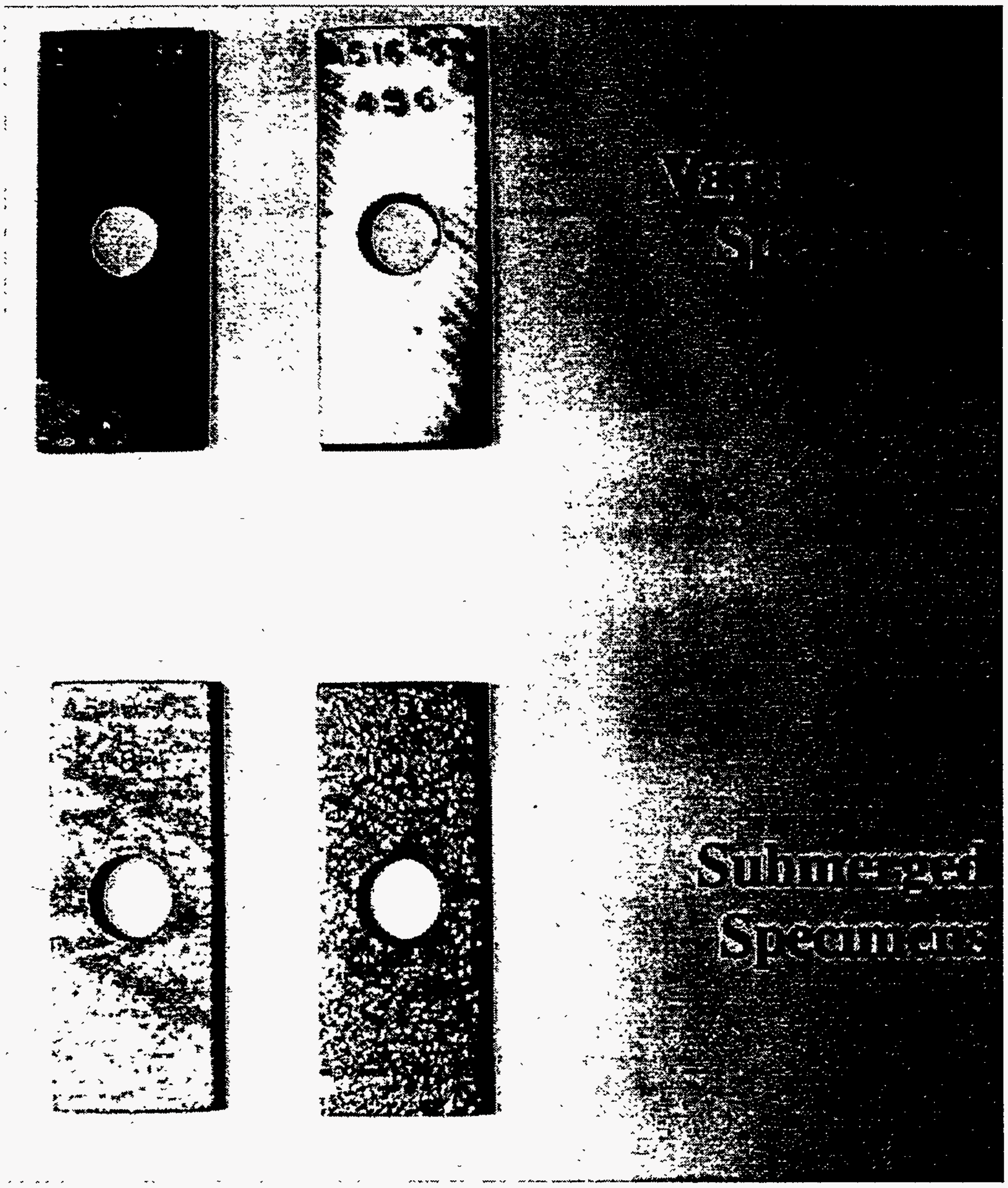

Figure M.4. Mild-Steel Specimens Removed from Vessel 2 (4 wt% Oxalic Acid) After One Week Exposure

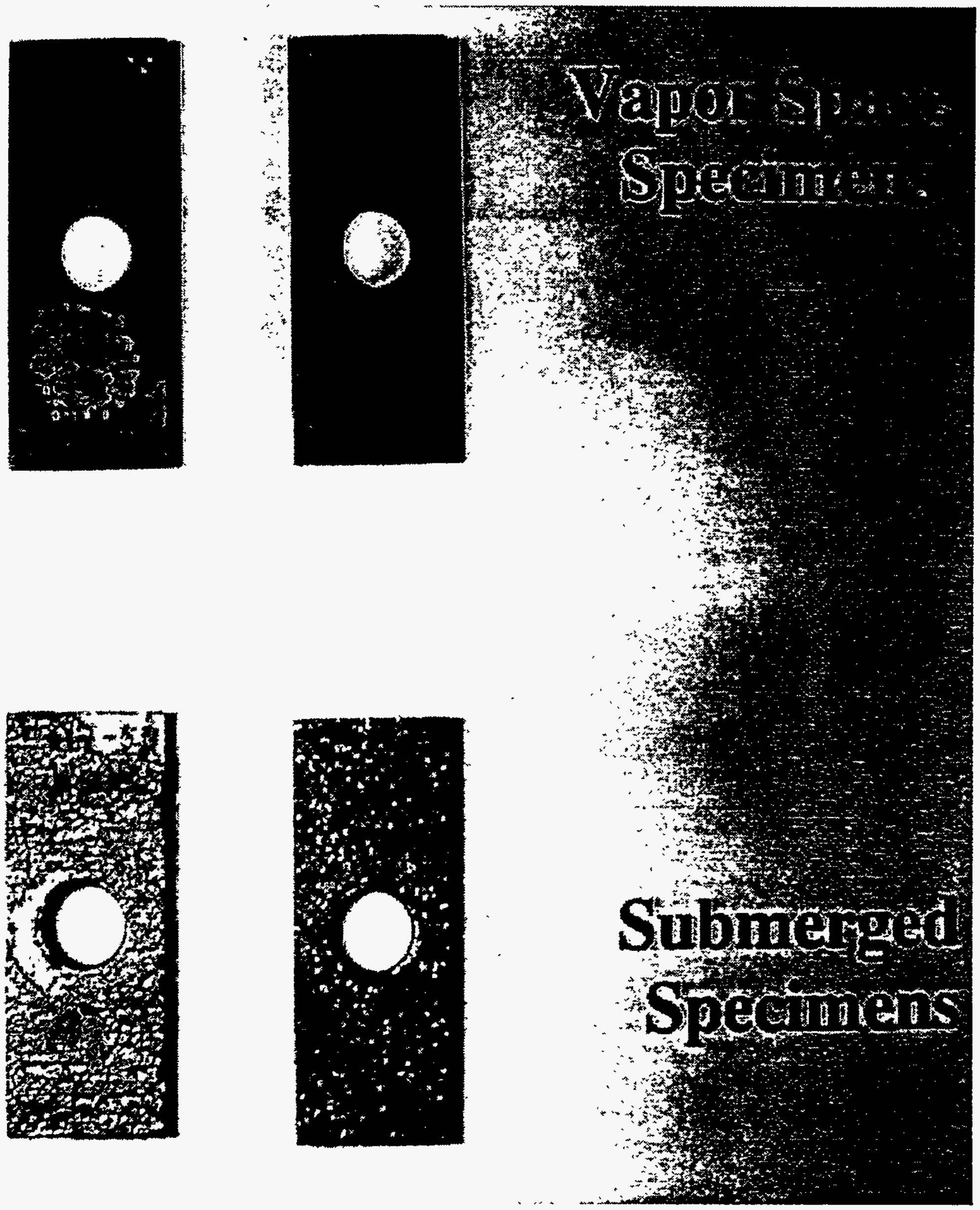

Figure M.5. Mild-Steel Specimens Removed from Vessel 2 (4 wt% Oxalic Acid) After Two Weeks Exposure

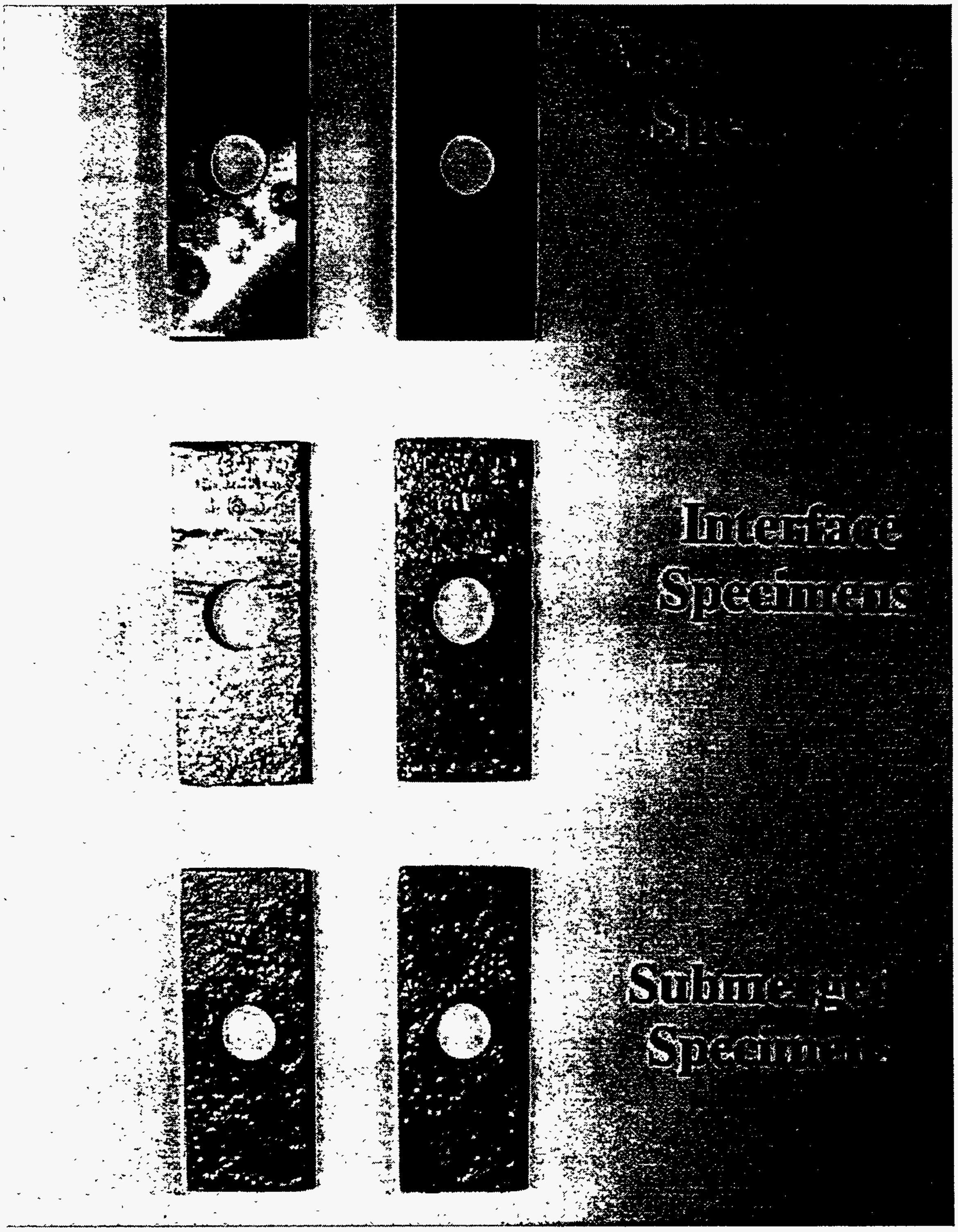

Figure M.6. Mild-Steel Specimens Removed from Vessel 2 (4 wt% Oxalic Acid) After Three Weeks Exposure

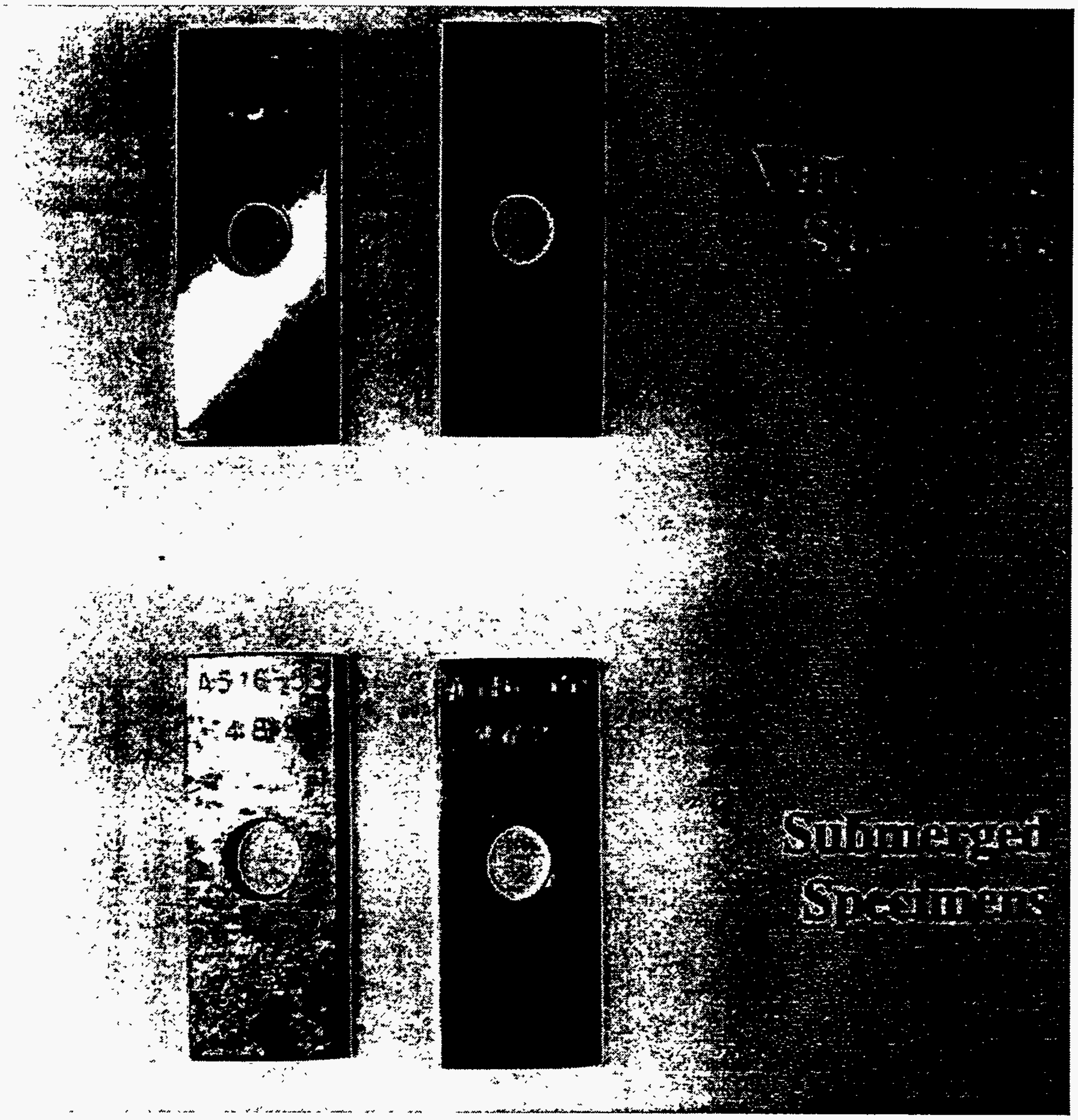

Figure M.7. Mild-Steel Specimens Removed from Vessel 3
(8 wt% Oxalic Acid) After One Week Exposure

Figure M.8. Mild-Steel Specimens Removed from Vessel 3 (8 wt% Oxalic Acid) After Two Weeks Exposure

Figure M.9. Mild-Steel Specimens Removed from Vessel 3 (8 wt% Oxalic Acid) After Three Weeks Exposure

Figure M.10. Mild-Steel Specimens Removed from Vessel 4 (8 wt% Oxalic Acid) After One Week Exposure

Figure M.11. Mild-Steel Specimens Removed from Vessel 4 (8 wt% Oxalic Acid) After Two Weeks Exposure

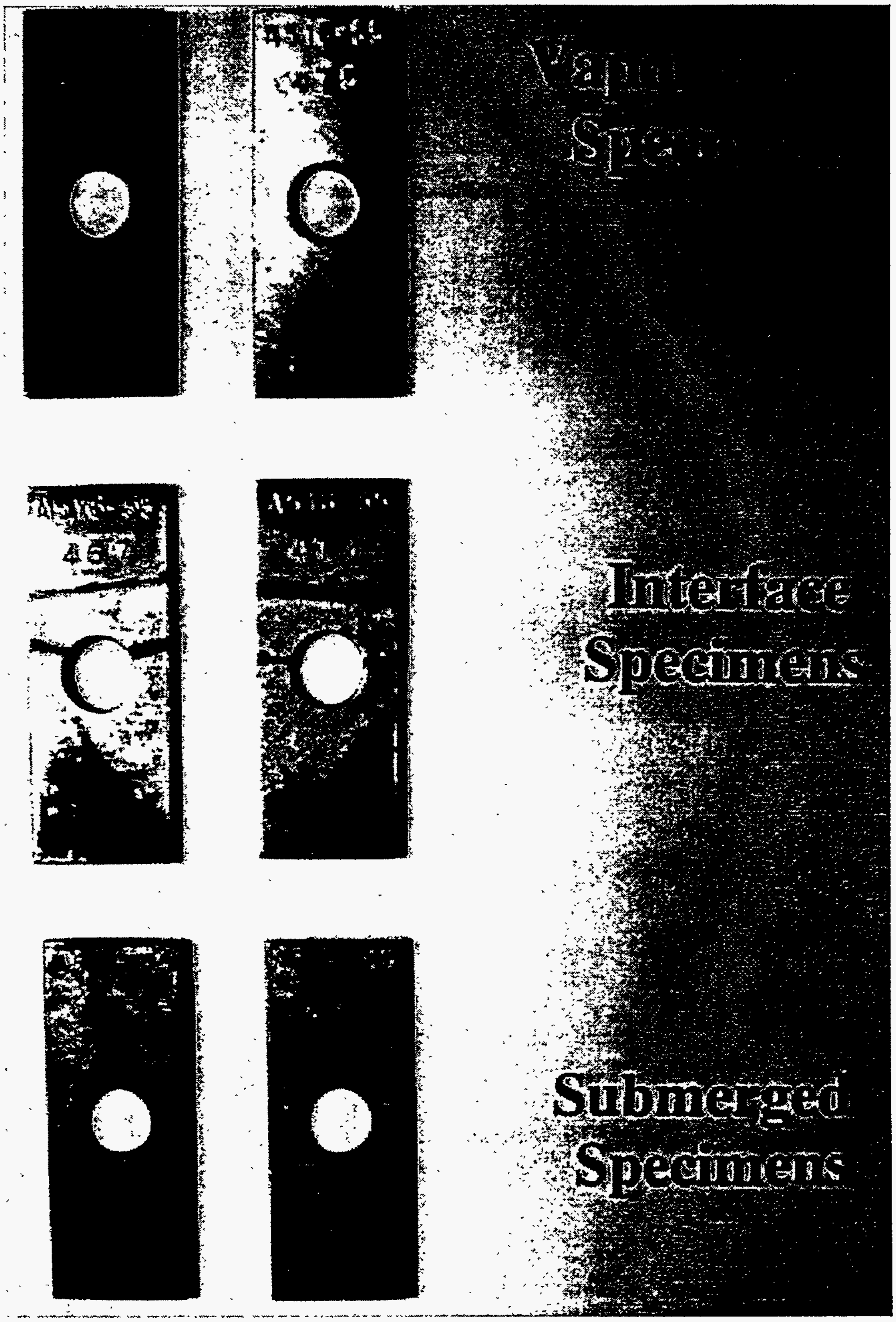

Figure M.12. Mild-Steel Specimens Removed from Vessel 4 (8 wt% Oxalic Acid) After Three Weeks Exposure

Appendix N

Supplemental Modeling Data

Table N.1. Species Considered in Modeling of WVNS Sludge Oxalic Acid System

$Al(NO_3)_3.6H_2O$
$Al(NO_3)_3.8H_2O$
$Al(NO_3)_3.9H_2O$
$Al(OH)_2ION$
$Al(OH)_3AQ$
$Al(OH)_3PPT$
$Al(OH)_4ION$
Al_2O_3PPT
ALBITEPPT
AlION
AlOHION
AlOOHPPT
$AlPO_4PPT$
$AlSO_4.16H_2O$
$AlSO_4PPT$
$Ca(NO_2)_2.1H_2O$
$Ca(NO_2)_2.4H_2O$
$Ca(NO_2)_2PPT$
$Ca(NO_3)_2.3H_2O$
$Ca(NO_3)_2.4H_2O$
$Ca(NO_3)_2PPT$
$Ca(OH)_2PPT$
$Ca_3(PO_4)_2PPT$
$CaC_2O_4.1H_2O$
CaC_2O_4AQ
CaC_2O_4PPT
$CaCO_3AQ$
$CaCO_3PPT$
$CaH(C_2O_4)_2AQ$
$CaH(C_2O_4)ION$
$CaH_2(PO_4)_2.1H_2O$
$CaH_2(PO_4)_2PPT$
CaH_2PO_4ION
$CaHCO_3ION$
$CaHPO_4.2H_2O$
$CaHPO_4PPT$
CaION
CaOHION
$CaPO_4ION$
$CaSO_4.2H_2O$
$CaSO_4AQ$
$CaSO_4PPT$
$Ce(C_2O_4)_2ION$
$Ce(C_2O_4)_3ION$
$Ce(C_2O_4)ION$
$Ce(NO_3)_3.6H_2O$
$Ce(NO_3)_3PPT$
$Ce(SO_4)_2ION$
$Ce_2(SO_4)_3PPT$
Ce_2O_3PPT
$Ce_2OX_3.10H_2O$
$Ce_2OX_3.9H_2O$
CeH_2PO_4ION
CeION
$CeNO_3ION$
CeOHION
$CePO_4.2H_2O$
$CePO_4PPT$
$CeSO_4ION$
CO_2AQ
CO_3ION
$Cr(NO_3)_3.9H_2O$
$Cr(OH)_2ION$
$Cr(OH)_3AQ$
$Cr(OH)_3PPT$
$Cr(OH)_4ION$
$Cr_2(SO_4)_3PPT$
CrH_2PO_4ION
$CrHPO_4ION$
CrION
$CrNO_3ION$
CrOHION
$Fe(C_2O_4)_2ION$
$Fe(C_2O_4)_3ION$
$Fe(NO_3)_3.9H_2O$
$Fe(OH)_2ION$
$Fe(OH)_3AQ$
$Fe(OH)_3PPT$
$Fe(OH)_4ION$
$Fe_2(OH)_2ION$
Fe_2O_3PPT
FeC_2O_4ION
FeH_2PO_4ION
$FeHC_2O_4ION$
$FeHPO_4ION$
FeION
$FeNO_3ION$
FeOHION
$FePO_4.2H_2O$
$FePO_4PPT$
H_2O
H_2OPPT
$H_2P_2O_7ION$
H_2PO_4ION
H_2SO_4AQ
$H_3P_2O_7ION$
H_3SIO_4ION
HCO_3ION
HION
HNO_2AQ
HNO_3AQ
HOXALATION
HP_2O_7ION
HPO_4ION
HSO_4ION
$K_2C_2O_4.1H_2O$
$K_2C_2O_4PPT$
$K_2CO_3.1.5H_2O$
K_2CO_3PPT
$K_2HPO_4.3H_2O$
$K_2HPO_4.6H_2O$
K_2HPO_4PPT
$K_2SO_4.1H_2O$
K_2SO_4PPT
$K_3PO_4.3H_2O$
$K_3PO_4.7H_2O$
K_3PO_4PPT
KH_2PO_4PPT
$KHCO_3PPT$
$KHSO_4AQ$
$KHSO_4PPT$
KION
KNO_2PPT
KnO_3PPT
$KOH.1H_2O$
$KOH.2H_2O$
KOHPPT
KSO_4ION
$Mn(C_2O_4)_2ION$
$Mn(C_2O_4)_3ION$
$Mn(NO_3)_2.1H_2O$
$Mn(NO_3)_2.4H_2O$
$Mn(NO_3)_2.6H_2O$
$Mn(NO_3)_2AQ$
$Mn(OH)_2AQ$
$Mn(OH)_2PPT$
$Mn(OH)_3ION$
$Mn(OH)_4ION$
$Mn_3(PO_4)_2.3H_2O$
$Mn_3(PO_4)_2.6H_2O$
$Mn_3(PO_4)_2PPT$
$MNC_2O_4.2H_2O$
MnC_2O_4AQ
MnC_2O_4PPT
$MnCO_3PPT$
MnION
$MnNO_3ION$
MnOHION
$MnSO_4.1H_2O$
$MnSO_4.5H_2O$
$MnSO_4.7H_2O$
$MnSO_4AQ$
$MnSO_4PPT$
$Na_2C_2O_4PPT$
$Na_2CO_3.10H_2O$
$Na_2CO_3.1H_2O$
$Na_2CO_3.7H_2O$
Na_2CO_3PPT
$Na_2HPO_4.12H_2O$
$Na_2HPO_4.2H_2O$
$Na_2HPO_4.7H_2O$
Na_2HPO_4PPT
Na_2OPPT
$Na_2SO_4.10H_2O$
Na_2SO_4PPT
$Na_3PO_4.1H_2O$
$Na_3PO_4.6H_2O$
$Na_3PO_4.8H_2O$
Na_3PO_4PPT
$NaAlO_2PPT$
$NaCO_3ION$
$NaH_2PO_4.1H_2O$
$NaH_2PO_4.2H_2O$
NaH_2PO_4PPT
$NaHCO_3AQ$
$NaHCO_3PPT$
$NaHSO_4PPT$
NaION
$NaNO_2PPT$
$NaNO_3AQ$
$NaNO_3PPT$
$NaOH.1H_2O$
NaOHPPT
$NaPHOH.12H_2O$

Table N.1, cont.

$NaSO_4ION$
$Ni(C_2O_4)_2ION$
$Ni(NO_3)_2.2H_2O$
$Ni(NO_3)_2.4H_2O$
$Ni(NO_3)_2.6H_2O$
$Ni(NO_3)_2PPT$
$Ni(OH)_2AQ$
$Ni(OH)_2PPT$
$Ni_3(PO_4)_2PPT$
$NiC_2O_4.2H_2O$
NiC_2O_4AQ
NiC_2O_4PPT
$NiCO_3PPT$
$NiION$
$NiNO_3ION$
$NiOH_3ION$
$NiOHION$
$NiSO_4.6H_2O$
$NiSO_4.7H_2O$
$NiSO_4AQ$
$NiSO_4PPT$
NO_2ION
NO_3ION
OHION
$OXALAC.2H_2O$
OXALACAQ
OXALACPPT
OXALATION
P_2O_7ION
PO_4ION
SIO_2AQ
SiO_2PPT
SO_2AQ
SO_4ION
$Sr(NO_2)_2.1H_2O$
$Sr(NO_2)_2PPT$
$Sr(NO_3)_2.4H_2O$
$Sr(NO_3)_2AQ$
$Sr(NO_3)_2PPT$
$Sr(OH)_2.1H_2O$
$Sr(OH)_2.8H_2O$
$Sr(OH)_2PPT$
$Sr_3(PO_4)_2PPT$
$SrC_2O_4.1H_2O$
SrC_2O_4AQ
SrC_2O_4PPT
$SrCO_3PPT$
$SrH(C_2O_4)_2AQ$
$SrH(CO_3)_2PPT$
$SrH_2(PO_4)_2PPT$
$SrHC_2O_4ION$
$SrHPO_4AQ$
$SrHPO_4PPT$
SrION
$SrNO_3ION$
SrOHION
$SrPO_4ION$
$SrSO_4PPT$
$Th(C_2O_4)_3ION$
$Th(C_2O_4)_4ION$
$Th(NO_3)_2ION$
$Th(NO_3)_3ION$
$Th(NO_3)_4.4H_2O$
$Th(NO_3)_4.5H_2O$
$Th(NO_3)_4.6H_2O$
$Th(NO_3)_4AQ$
$Th(NO_3)_4PPT$
$Th(OH)_2ION$
$Th(OH)_3ION$
$Th(OH)_4AQ$
$Th(SO_4)_2AQ$
$Th(SO_4)_2PPT$
$Th(SO_4)_3ION$
$Th(SO_4)_4ION$
Th_2OH_2ION
ThC_2O_4ION
$ThH(PO_4)_2AQ$
$ThH(PO_4)_3ION$
$ThH_2(PO_4)_2ION$
ThH_2PO_4ION
$ThHC_2O_4ION$
$ThHPO_4ION$
ThION
$ThNO_3ION$
ThO_2PPT
ThOHION
$ThSO_4ION$
$(UO_2)_2(OH)_2ION$
$(UO_2)_2P_2O_7PPT$
$(UO_2)_3(OH)_5ION$
$(UO_2)_3(PO_4)_2.4H_2O$
$(UO_2)_3(PO_4)_2PPT$
$UO_2(CO_3)_3ION$
$UO_2(NO_3)_2.2H_2O$
$UO_2(NO_3)_2.3H_2O$
$UO_2(NO_3)_2.6H_2O$
$UO_2(NO_3)_2PPT$
$UO_2(OH)_2AQ$
$UO_2(OH)_2PPT$
$UO_2(SO_4)_2ION$
$UO_2C_2O_4.3H_2O$
UO_2CO_3AQ
UO_2ION
UO_2OHION
$UO_2SO_4.3H_2O$
UO_2SO_4AQ
UO_2SO_4PPT
$Zr(C_2O_4)_2AQ$
$Zr(NO_3)_2ION$
$Zr(OH)_2ION$
$Zr(OH)_3ION$
$Zr(OH)_4AQ$
$Zr(OH)_5ION$
$Zr(SO_4)_2.1H_2O$
$Zr(SO_4)_2.4H_2O$
$Zr(SO_4)_2PPT$
ZrC_2O_4ION
ZrION
$ZrNO_3ION$
ZrO_2PPT
ZrOHION
$ZrSO_4ION$

Table N.2. Equations Considered in Modeling

$Al(NO_3)_3.6H_2O=AlION+3NO_3ION+6H_2O$
$Al(NO_3)_3.8H_2O=AlION+3NO_3ION+8H_2O$
$Al(NO_3)_3.9H_2O=AlION+3NO_3ION+9H_2O$
$AL_2O_3PPT+6HION=2ALION+3H_2O$
$ALBITEPPT+4HION=NaION+AlION+3SIO_2AQ+2H_2O$
$AlOH_2ION=AlOHION+OHION$
$AlOH_3AQ=AlOH_2ION+OHION$
$AlOH_3PPT+OHION=AlOH_4ION$
$AlOH_4ION=AlOH_3AQ+OHION$
$AlOHION=AlION+OHION$
$AlOOHPPT+OHION+H_2O=Al(OH)_4ION$
$AlPO_4PPT=AlION+PO_4ION$
$AlSO_4.16H_2O=2AlION+3SO_4ION+16H_2O$
$AlSO_4PPT=2AlION+3SO_4ION$
$Ca(NO_2)_2.1H_2O=CaION+2NO_2ION+1H_2O$
$Ca(NO_2)_2.4H_2O=CaION+2NO_2ION+4H_2O$
$Ca(NO_2)_2PPT=CaION+2NO_2ION$
$Ca(NO_3)_2.3H_2O=CaION+2NO_3ION+3H_2O$
$Ca(NO_3)_2.4H_2O=CaION+2NO_3ION+4H_2O$
$Ca(NO_3)_2PPT=CaION+2NO_3ION$
$Ca(OH)_2PPT=CaION+2OHION$
$Ca_3(PO_4)_2PPT=3CaION+2PO_4ION$
$CaC_2O_4.1H_2O=CaION+OXALATION+H_2O$
$CaC_2O_4AQ=CaION+OXALATION$
$CaC_2O_4PPT=CaION+OXALATION$
$CaCO_3AQ=CaION+CO_3ION$
$CaCO_3PPT=CaION+CO_3ION$
$CaH(C_2O_4)_2AQ=CaION+2HOXALATION$
$CaH_2(PO_4)_2.1H_2O=CaION+2H_2PO_4ION+H_2O$
$CaH_2(PO_4)_2PPT=CaION+2H_2PO_4ION$
$CaH_2PO_4ION=CaION+H_2PO_4ION$
$CaHC_2O_4ION=CaION+HOXALATION$
$CaHCO_3ION=CaION+HCO_3ION$
$CaHPO_4.2H_2O=CaION+HPO_4ION+2H_2O$
$CaHPO_4PPT=CaION+HPO_4ION$
$CaOHION=CaION+OHION$
$CaPO_4ION=CaION+PO_4ION$
$CaSO_4.2H_2O=CaION+SO_4ION+2H_2O$
$CaSO_4AQ=CaION+SO_4ION$
$CaSO_4PPT=CaION+SO_4ION$
$.1Ce(NO_3)_3PPT=.1CeION+.3NO_3ION$
$.1Ce_2O_3PPT+.3HION=.2CeION+.3OHION$
$Ce(NO_3)_3.6H_2O=CeION+3NO_3ION+6H_2O$
$Ce(SO_4)_2ION=CeION+2SO_4ION$
$Ce_2(SO_4)_3PPT=2CeION+3SO_4ION$
$Ce_2OX3.10H_2O=2CeION+3OXALATION+10H_2O$

Table N.2, cont.

$Ce_2OX3.9H_2O=2CeION+3OXALATION+9H_2O$

$CeC_2O_4ION=CeION+OXALATION$

$CeH_2PO_4ION=CeION+H_2PO_4ION$

$CeNO_3ION=CeION+NO_3ION$

$CeOHION=CeION+OHION$

$CePO_4.2H_2O=CeION+PO_4ION+2H_2O$

$CePO_4PPT=CeION+PO_4ION$

$CeSO_4ION=CeION+SO_4ION$

$CO_2AQ+H_2O=HION+HCO_3ION$

$Cr(NO_3)_3.9H_2O=CrION+3NO_3ION+9H_2O$

$Cr_2(SO_4)3PPT=2CrION+3SO_4ION$

$CrH_2PO_4ION=CrION+H_2PO_4ION$

$CrHPO_4ION=CrION+H_PO_4ION$

$CrNO_3ION=CrION+NO_3ION$

$CrOH_2ION=CrION+2OHION$

$CrOH_3AQ=CrION+3OHION$

$CrOH_3PPT=CrION+3OHION$

$CrOH_4ION=CrION+4OHION$

$CrOHION=CrION+OHION$

$Fe(C_2O_4)_2ION=FeION+2OXALATION$

$Fe(C_2O_4)_3ION=FeION+3OXALATION$

$Fe(NO_3)_3.9H_2O=FeION+3NO_3ION+9H_2O$

$Fe(OH)_2ION=FeOHION+OHION$

$Fe(OH)_3AQ=Fe(OH)_2ION+OHION$

$Fe(OH)_3PPT=FeION+3OHION$

$Fe(OH)_4ION=Fe(OH)_3AQ+OHION$

$FE_2O_3PPT+6HION=2FEION+3H_2O$

$Fe_2OH_2ION=2FeION+2OHION$

$FeC_2O_4ION=FeION+OXALATION$

$FeH_2PO_4ION=FeION+H_2PO_4ION$

$FeHC_2O_4ION=FeION+HOXALATION$

$FeHPO_4ION=FeION+HPO_4ION$

$FeNO_3ION=FeION+NO_3ION$

$FeOHION=FeION+OHION$

$FePO_4.2H_2O=FeION+PO_4ION+2H_2O$

$FePO_4PPT=FeION+PO_4ION$

$H_2O=HION+OHION$

$H_2OPPT=H_2O$

$H_2P_2O_7ION=HION+HP_2O_7ION$

$H_2PO_4ION=HION+H_PO_4ION$

$H_2SO_4AQ=HION+HSO_4ION$

$H_3P_2O_7ION=HION+H_2P_2O_7ION$

$HCO_3ION=HION+CO_3ION$

$HNO_2AQ=HION+NO_2ION$

$HNO_3AQ=HION+NO_3ION$

$HOXALATION=HION+OXALATION$

Table N.2, cont.

$HP_2O_7ION=HION+P_2O_7ION$
$HPO_4ION=HION+PO_4ION$
$HSO_4ION=HION+SO_4ION$
$K_2C_2O_4.1H_2O=2KION+OXALATION+H_2O$
$K_2C_2O_4PPT=2KION+OXALATION$
$K_2CO_3.1.5H_2O=2KION+CO_3ION+1.5H_2O$
$K_2CO_3PPT=2KION+CO_3ION$
$K_2HPO_4.3H_2O=2KION+HPO_4ION+3H_2O$
$K_2HPO_4.6H_2O=2KION+HPO_4ION+6H_2O$
$K_2HPO_4PPT=2KION+HPO_4ION$
$K_2SO_4.1H_2O=2KION+SO_4ION+1H_2O$
$K_2SO_4PPT=2KION+SO_4ION$
$K_3PO_4.3H_2O=3KION+PO_4ION+3H_2O$
$K_3PO_4.7H_2O=3KION+PO_4ION+7H_2O$
$K_3PO_4PPT=3KION+PO_4ION$
$KH_2PO_4PPT=KION+H_2PO_4ION$
$KHCO_3PPT=KION+HCO_3ION$
$KHSO_4AQ=KION+HSO_4ION$
$KHSO_4PPT=KION+HSO_4ION$
$KNO_2PPT=KION+NO_2ION$
$KNO_3PPT=KION+NO_3ION$
$KOH.1H_2O=KION+OHION+1H_2O$
$KOH.2H_2O=KION+OHION+2H_2O$
KOHPPT=KION+OHION
$KSO_4ION=KION+SO_4ION$
$Mn(C_2O_4)_2ION=MnION+2OXALATION$
$Mn(C_2O_4)_3ION=MnION+3OXALATION$
$Mn(NO_3)_2.1H_2O=MnION+2NO_3ION+1H_2O$
$Mn(NO_3)_2.4H_2O=MnION+2NO_3ION+4H_2O$
$Mn(NO_3)_2.6H_2O=MnION+2NO_3ION+6H_2O$
$Mn(NO_3)_2AQ=MnION+2NO_3ION$
$Mn(OH)_2AQ=MnION+2OHION$
$Mn(OH)_2PPT=MnION+2OHION$
$Mn(OH)_3ION=MnION+3OHION$
$Mn(OH)_4ION=MnION+4OHION$
$Mn_3(PO_4)_2.3H_2O=3MnION+2PO_4ION+3H_2O$
$Mn_3(PO_4)_2.6H_2O=3MnION+2PO_4ION+6H_2O$
$Mn_3(PO_4)_2PPT=3MnION+2PO_4ION$
$MnC_2O_4.2H_2O=MnION+OXALATION+2H_2O$
$MnC_2O_4AQ=MnION+OXALATION$
$MnC_2O_4PPT=MnION+OXALATION$
$MnCO_3PPT=MnION+CO_3ION$
$MnNO_3ION=MnION+NO_3ION$
MnOHION=MnION+OHION
$MnSO_4.1H_2O=MnION+SO_4ION+1H_2O$
$MnSO_4.5H_2O=MnION+SO_4ION+5H_2O$

Table N.2, cont.

$MnSO_4.7H_2O=MnION+SO_4ION+7H_2O$
$MnSO_4AQ=MnION+SO_4ION$
$MnSO_4PPT=MnION+SO_4ION$
$Na_2C_2O_4PPT=2NaION+OXALATION$
$Na_2CO_3.10H_2O=2NaION+CO_3ION+10H_2O$
$Na_2CO_3.1H_2O=2NaION+CO_3ION+1H_2O$
$Na_2CO_3.7H_2O=2NaION+CO_3ION+7H_2O$
$Na_2CO_3PPT=2NaION+CO_3ION$
$Na_2HPO_4.12H_2O=2NaION+HPO_4ION+12H_2O$
$Na_2HPO_4.2H_2O=2NaION+HPO_4ION+2H_2O$
$Na_2HPO_4.7H_2O=2NaION+HPO_4ION+7H_2O$
$Na_2HPO_4PPT=2NaION+HPO_4ION$
$Na_2SO_4.10H_2O=2NaION+SO_4ION+10H_2O$
$Na_2SO_4PPT=2NaION+SO_4ION$
$0.1NA_2OPPT+0.2HION=0.2NAION+0.1H_2O$
$Na_3PO_4.1H_2O=3NaION+PO_4ION+H_2O$
$Na_3PO_4.6H_2O=3NaION+PO_4ION+6H_2O$
$Na_3PO_4.8H_2O=3NaION+PO_4ION+8H_2O$
$Na_3PO_4PPT=3NaION+PO_4ION$
$NaAlO_2PPT+2H_2O=NaION+Al(OH)_4ION$
$NaCO_3ION=NaION+CO_3ION$
$NaH_2PO_4.1H_2O=NaION+H_2PO_4ION+1H_2O$
$NaH_2PO_4.2H_2O=NaION+H_2PO_4ION+2H_2O$
$NaH_2PO_4PPT=NaION+H_2PO_4ION$
$NaHCO_3AQ=NaION+HCO_3ION$
$NaHCO_3PPT=NaION+HCO_3ION$
$NaHSO_4PPT=NaION+HSO_4ION$
$NaNO_2PPT=NaION+NO_2ION$
$NaNO_3AQ=NaION+NO_3ION$
$NaNO_3PPT=NaION+NO_3ION$
$NaOH.1H_2O=NaION+OHION+H_2O$
$NaOHPPT=NaION+OHION$
$NaPHOH.12H_2O=3.25NaION+PO_4ION+.25OHION+12H_2O$
$NaSO_4ION=NaION+SO_4ION$
$Ni(C_2O_4)_2ION=NiION+2OXALATION$
$Ni(NO_3)_2.2H_2O=NiION+2NO_3ION+2H2O$
$Ni(NO_3)_2.4H_2O=NiION+2NO_3ION+4H_2O$
$Ni(NO_3)_2.6H_2O=NiION+2NO_3ION+6H_2O$
$Ni(NO_3)_2PPT=NiION+2NO_3ION$
$Ni(OH)_2AQ=NiOHION+OHION$
$Ni(OH)_2PPT=NiION+2OHION$
$NI(OH)_3ION=NI(OH)_2AQ+OHION$
$Ni_3(PO_4)_2PPT=3NiION+2PO_4ION$
$NiC_2O_4.2H_2O=NiION+OXALATION+2H_2O$
$NiC_2O_4AQ=NiION+OXALATION$
$NiC_2O_4PPT=NiION+OXALATION$

Table N.2, cont.

$NiCO_3PPT=NiION+CO_3ION$
$NiNO_3ION=NiION+NO_3ION$
$NiOHION=NiION+OHION$
$NiSO_4.6H_2O=NiION+SO_4ION+6H_2O$
$NiSO_4.7H_2O=NiION+SO_4ION+7H_2O$
$NiSO_4AQ=NiION+SO_4ION$
$NiSO_4PPT=NiION+SO_4ION$
$OXALAC.2H_2O=2HION+OXALATION+2H_2O$
$OXALACAQ=2HION+OXALATION$
$OXALACPPT=2HION+OXALATION$
$P_2O_7ION+H_2O=2PO_4ION+2HION$
$SIO_2AQ+2H_2O=H_3SIO_4ION+HION$
$SIO_2PPT+2H_2O=H_3SIO_4ION+HION$
$SO_3AQ+H_2O=H_2SO_4AQ$
$Sr(NO_2)_2PPT=SrION+2NO_2ION$
$Sr(NO_3)_2.4H_2O=SrION+2NO_3ION+4H_2O$
$Sr(NO_3)_2AQ=SrNO_3ION+NO_3ION$
$Sr(NO_3)_2PPT=SrION+2NO_3ION$
$Sr(OH)_2.1H_2O=SrION+2OHION+H_2O$
$Sr(OH)_2PPT=SrION+2OHION$
$Sr_3(PO_4)_2PPT=3SrION+2PO_4ION$
$SrC_2O_4.1H_2O=SrION+OXALATION+H_2O$
$SrC_2O_4AQ=SrION+OXALATION$
$SrC_2O_4PPT=SrION+OXALATION$
$SrCO_3PPT=SrION+CO_3ION$
$SrH(C_2O_4)_2AQ=SrION+2HOXALATION$
$SrH(CO_3)_2PPT=SrION+2HCO_3ION$
$SrH(CO_3)_2PPT=SrION+2HCO_3ION$
$SrH(CO_3)_2PPT=SrION+2HCO_3ION$
$SrH_2(PO_4)_2PPT=SrION+2H_2PO_4ION$
$SrHC_2O_4ION=SrION+HOXALATION$
$SrHPO_4AQ=SrION+HPO_4ION$
$SrNO_3ION=SrION+NO_3ION$
$SrOH_2.8H_2O=SrION+2OHION+8H_2O$
$SrOHION=SrION+OHION$
$SrPO_4ION=SrION+PO_4ION$
$SrSO_4PPT=SrION+SO_4ION$
$Th(C_2O_4)_3ION=ThION+3OXALATION$
$Th(C_2O_4)_4ION=ThION+4OXALATION$
$Th(NO_3)_2ION=ThNO_3ION+NO_3ION$
$Th(NO_3)_3ION=Th(NO_3)_2ION+NO_3ION$
$Th(NO_3)_4.4H_2O=ThION+4NO_3ION+4H_2O$
$Th(NO_3)_4.5H_2O=ThION+4NO_3ION+5H_2O$
$Th(NO_3)_4.6H_2O=ThION+4NO_3ION+6H_2O$
$Th(NO_3)_4AQ=Th(NO_3)_3ION+NO_3ION$
$Th(NO_3)_4PPT=ThION+4NO_3ION$

Table N.2, cont.

$Th(OH)_2ION=ThOHION+OHION$
$Th(OH)_3ION=Th(OH)_2ION+OHION$
$Th(OH)_4AQ=Th(OH)_3ION+OHION$
$Th(SO_4)_2AQ=ThION+2SO_4ION$
$Th(SO_4)_2PPT=ThION+2SO_4ION$
$Th(SO_4)_3ION=ThION+3SO_4ION$
$Th(SO_4)_4ION=ThION+4SO_4ION$
$Th_2(OH)_2ION=2ThION+2OHION$
$ThC_2O_4ION=ThION+OXALATION$
$ThH(PO_4)_2AQ=ThION+2HPO_4ION$
$ThH(PO_4)_3ION=ThION+3HPO_4ION$
$ThH_2(PO_4)_2ION=ThION+2H_2PO_4ION$
$ThH_2PO_4ION=ThION+H_2PO_4ION$
$ThHC_2O_4ION=ThION+HOXALATION$
$ThHPO_4ION=ThION+HPO_4ION$
$ThNO_3ION=ThION+NO_3ION$
$ThO_2PPT+2H_2O=ThION+4OHION$
$ThOHION=ThION+OHION$
$ThSO_4ION=ThION+SO_4ION$
$(UO_2)_2(OH)_2ION=2UO_2ION+2OHION$
$(UO_2)_2P_2O_7PPT=2UO_2ION+P_2O_7ION$
$(UO_2)_3(PO_4)_2.4H_2O=3UO_2ION+2PO_4ION+4H_2O$
$(UO_2)_3(PO_4)_2PPT=3UO_2ION+2PO_4ION$
$.1(UO_2)_3(OH)_5ION=.3UO_2ION+.5OHION$
$UO_2(CO_3)_3ION=UO_2ION+3CO_3ION$
$UO_2(NO_3)_2.2H_2O=UO_2ION+2NO_3ION+2H_2O$
$UO_2(NO_3)_2.3H_2O=UO_2ION+2NO_3ION+3H_2O$
$UO_2(NO_3)_2.6H_2O=UO_2ION+2NO_3ION+6H_2O$
$UO_2(NO_3)_2PPT=UO_2ION+2NO_3ION$
$UO_2(SO_4)_2ION=UO_2ION+2SO_4ION$
$UO_2C_2O_4.3H_2O=UO_2ION+OXALATION+3H_2O$
$UO_2C_2O_4AQ=UO_2ION+OXALATION$
$UO_2CO_3AQ=UO_2ION+CO_3ION$
$UO_2OH_2AQ=UO_2ION+2OHION$
$UO_2OH_2PPT=UO_2ION+2OHION$
$UO_2OHION+HION=UO_2ION+H_2O$
$UO_2SO_4.3H_2O=UO_2ION+SO_4ION+3H_2O$
$UO_2SO_4AQ=UO_2ION+SO_4ION$
$UO_2SO_4PPT=UO_2ION+SO_4ION$
$Zr(C_2O_4)_2AQ=ZrION+2OXALATION$
$Zr(NO_3)_2ION=ZrION+2NO_3ION$
$Zr(OH)_5ION=Zr(OH)_4AQ+OHION$
$Zr(SO_4)_2.1H_2O=ZrION+2SO_4ION+H_2O$
$Zr(SO_4)_2.4H_2O=ZrION+2SO_4ION+4H_2O$
$Zr(SO_4)_2PPT=ZrION+2SO_4ION$
$ZrC_2O_4ION=ZrION+OXALATION$

Table N.2, cont.

$ZrNO_3ION=ZrION+NO_3ION$
$ZrO_2PPT+2H_2O=ZrION+4OHION$
$ZrOH_2ION=ZrOHION+OHION$
$ZrOH_3ION=ZrOH_2ION+OHION$
$ZrOH_4AQ=ZrOH_3ION+OHION$
$ZrOHION=ZrION+OHION$
$ZrSO_4ION=ZrION+SO_4ION$

Table N.3. Speciation of Modeled Sludge Before Oxalic Acid Addition

Constituent	Liquid Composition (gram-mols)	Solid Composition (gram-mols)
H_2O	3.65E+01	
$Al(OH)_3$	5.87E-11	
CaC_2O_4		
$CaCO_3$	3.95E-17	
$CaH(C_2O_4)_2$		
$CaSO_4$	1.45E-11	
CO_2	6.35E-20	
$CrOH_3$	5.29E-08	
$FeOH_3$	2.29E-13	
H_2SO_4		
HNO_2	1.02E-14	
HNO_3	2.09E-18	
$KHSO_4$	2.64E-22	
MnC_2O_4		
$Mn(NO_3)_2$	1.45E-15	
$Mn(OH)_2$	1.30E-08	1.52E-02
$MnSO_4$	9.48E-19	
$NaHCO_3$	2.87E-10	
$NaNO_3$	4.26E-01	
NiC_2O_4		
$Ni(OH)_2$	9.21E-10	1.62E-03
$NiSO_4$	3.90E-23	
OXAlAC		
SiO_2	5.28E-09	
SO_3		
SrC_2O_4		
$SrH(C_2O_4)_2$		
$SrHPO_4$	1.29E-14	
$Sr(NO_3)_2$	3.47E-05	
$ThH(PO_2)_4$		
$Th(NO_3)_4$		
$Th(OH)_4$	1.60E-14	
$Th(SO_4)_2$		
$UO_2C_2O_4$		
UO_2CO_3	8.03E-22	
$UO_2(OH)_2$	4.75E-09	2.01E-04
UO_2SO_4	3.06E-28	
$Zr(C_2O_4)_2$		
$Zr(OH)_4$	1.91E-13	

Table N.3, cont.

Constituent	Liquid Composition (gram-mols)	Solid Composition (gram-mols)
AlBITE		3.85E-03
$Al(NO_3)_3.9H_2O$		
$Ca_3(PO_4)_2$		
$CaC_2O_4.1H_2O$		
$Ca(OH)_2$		7.03E-03
$Ce(NO_3)_3$		8.79E-03
$Ce(OH)_3$		
$CePO_4$		
$Cr(NO_3)_3$		
Fe_2O_3		5.81E-02
$Fe(NO_3)_3$		
$FePO_4.2H_2O$		
KNO_3		
$Mn_3(PO_4)_2.3H_2O$		
$Mn_3(PO_4)_2.6H_2O$		
$Mn_3(PO_4)_2$		
$MnCO_3$		
Na_2SO_4		
NaH_2PO_4		
$NaNO_2$		
NaOH		
$NiC_2O_4.2H_2O$		
$Ni(NO_3)_2$		
$Sr_3(PO_4)_2$		3.22E-03
$SrCO_3$		6.94E-03
$Sr(OH)_2.8H_2O$		6.45E-03
$Sr(OH)_2$		
$SrSO_4$		
ThO_2		4.54E-03
$(UO_2)_3(PO_4)_2.4H_2O$		
$(UO_2)_3(PO_4)_2$		
$UO_2(NO_3)_2$		
ZrO_2		6.07E-03
$Zr(SO_4)_2.1H_2O$		
$Zr(SO_4)_2.4H_2O$		
$Zr(SO_4)_2$		
AlION		
$Al(OH)_2ION$	4.97E-20	
$Al(OH)_4ION$	1.04E-01	
AlOHION	4.34E-30	
CaH_2PO_4ION	3.43E-24	
$CaHC_2O_4ION$		

Table N.3, cont.

Constituent	Liquid Composition (gram-mols)	Solid Composition (gram-mols)
CaHCO$_3$ION	6.16E-19	
CaION	6.15E-07	
CaOHION	4.36E-06	
CaPO$_4$ION	3.04E-08	
Ce(C$_2$O$_4$)$_2$ION		
Ce(C$_2$O$_4$)$_3$ION		
CeC$_2$O$_4$ION		
CeH$_2$PO$_4$ION		
CeION		
CeNO$_3$ION		
CeOHION	6.89E-34	
Ce(SO$_4$)$_2$ION		
CeSO$_4$ION		
CO$_3$ION	2.29E-04	
CrH$_2$PO$_4$ION		
CrH$_2$PO$_4$ION	2.45E-32	
CrION	3.79E-34	
CrNO$_3$ION	3.18E-32	
Cr(OH)$_2$ION	7.35E-17	
Cr(OH)$_4$ION	5.73E-03	
CrOHION	1.17E-23	
Fe$_2$(OH)$_2$ION		
Fe(C$_2$O$_4$)$_2$ION		
Fe(C$_2$O$_4$)$_3$ION		
FeC$_2$O$_4$ION		
FeH$_2$PO$_4$ION		
FeHC$_2$O$_4$ION		
FeHPO$_4$ION		
FeION		
FeNO$_3$ION		
FeOH$_2$ION	1.40E-22	
FeOH$_4$ION	5.37E-07	
FeOHION	3.05E-33	
H$_2$P$_2$O$_7$ION		
H$_2$PO$_4$ION	6.48E-18	
H$_3$P$_2$O$_7$ION		
H$_3$SiO$_4$ION	5.02E-03	
HCO$_3$ION	2.14E-10	
HION	7.01E-16	
HOXAIATION		
HP$_2$O$_7$ION	4.19E-28	
HPO$_4$ION	9.39E-09	

Table N.3, cont.

Constituent	Liquid Composition (gram-mols)	Solid Composition (gram-mols)
HSO_4ION	7.01E-19	
KION	7.54E-02	
KSO_4ION	1.44E-05	
$Mn(C_2O_4)_2$ION		
$Mn(C_2O_4)_3$ION		
MNiON	3.00E-13	
$MnNO_3$ION	2.04E-15	
$MnOH_3$ION	3.14E-05	
$MnOH_4$ION	1.15E-02	
MnOHION	5.48E-11	
$NaCO_3$ION	1.66E-04	
NaION	4.82E+00	
$NaSO_4$ION	1.21E-04	
$Ni(C_2O_4)_2$ION		
NiION	7.79E-18	
$NiNO_3$ION	1.43E-19	
$NiOH_3$ION	8.85E-05	
NiOHION	5.94E-15	
NO_2ION	6.46E-02	
NO_3ION	1.67E+00	
OHION	3.02E+00	
OXAIATION		
P_2O_7ION	1.18E-19	
PO_4ION	3.38E-05	
SO_4ION	1.25E-03	
$SrHC_2O_4$ION		
SrION	6.58E-04	
$SrNO_3$ION	2.46E-04	
SrOHION	1.31E-03	
$SrPO_4$ION	2.37E-07	
Th_2OH_2ION		
$Th(C_2O_4)_3$ION		
$Th(C_2O_4)_4$ION		
ThC_2O_4ION		
$ThH_2(PO_4)_2$ION		
ThH_2PO_4ION		
$ThHC_2O_4$ION		
$ThH(PO_4)_3$ION		
$ThHPO_4$ION		
ThION		
$Th(NO_3)_2$ION		
$Th(NO_3)_3$ION		

Table N.3, cont.

Constituent	Liquid Composition (gram-mols)	Solid Composition (gram-mols)
$ThNO_3ION$		
$Th(OH)_2ION$		
$Th(OH)_3ION$	3.19E-26	
ThOHION		
$Th(SO_4)_3ION$		
$Th(SO_4)_4ION$		
$ThSO_4ION$		
$(UO_2)_2(OH)_2ION$	4.99E-27	
$(UO_2)_3(OH)_5ION$	1.09E-16	
$UO_2(C_2O_4)_2ION$		
$UO_2(C_2O_4)_3ION$		
$UO_2(CO_3)_2ION$	2.25E-17	
$UO_2(CO_3)_3ION$	2.93E-13	
UO_2ION	2.51E-26	
UO_2OHION	3.67E-16	
$UO_2(SO_4)_2ION$	5.73E-30	
ZrC_2O_4ION		
ZrION		
$Zr(NO_3)_2ION$		
$ZrNO_3ION$		
$Zr(OH)_2ION$	3.91E-35	
$Zr(OH)_3ION$	3.62E-23	
$Zr(OH)_5ION$	9.31E-04	
ZrOHION		
$ZrSO_4ION$		

Table N.4. Speciation of Modeled Sludge After Oxalic Acid Addition at 20, 40, and 80 L/kg

Constituent	20 L/kg liquid (gm-mols)	20 L/kg solid (gm-mols)	40 L/kg liquid (gm-mols)	40 L/kg solid (gm-mols)	80 L/kg liquid (gm-mols)	80 L/kg solid (gm-mols)
H_2O	1.06E+03		2.08E+03		4.13E+03	
$Al(OH)_3$	2.18E-16		1.40E-16		1.16E-16	
CaC_2O_4	2.64E-05		2.28E-05		2.14E-05	
$CaCO_3$	4.64E-26		1.81E-26		8.11E-27	
$CaH(C_2O_4)_2$	2.62E-03		2.52E-03		2.48E-03	
$CaSO_4$	6.46E-11		7.86E-11		1.04E-10	
CO_2	7.34E-03		7.34E-03		7.34E-03	
$CrOH_3$	1.00E-20		9.61E-21		1.13E-20	
$FeOH_3$	3.68E-22		7.75E-22		1.17E-21	
H_2SO_4	3.35E-18		1.05E-17		3.10E-17	
HNO_2	6.42E-02		6.43E-02		6.43E-02	
HNO_3	7.94E-03		1.08E-02		1.27E-02	
$KHSO_4$	3.36E-11		4.52E-11		6.25E-11	
MnC_2O_4	4.66E-03		4.09E-03		3.88E-03	
$Mn(NO_3)_2$	1.22E-04		4.31E-05		1.32E-05	
$Mn(OH)_2$	1.84E-23		1.45E-23		1.32E-23	
$MnSO_4$	3.22E-09		3.98E-09		5.33E-09	
$NaHCO_3$	6.64E-09		2.85E-09		1.33E-09	
$NaNO_3$	8.57E-02		4.91E-02		2.66E-02	
NiC_2O_4	1.06E-03		1.11E-03		1.13E-03	
$Ni(OH)_2$	4.20E-22		3.97E-22		3.85E-22	
$NiSO_4$	4.27E-11		6.30E-11		9.04E-11	
OXAlAC	1.06E+01		2.35E+01		4.92E+01	
SiO_2	1.66E-02		1.66E-02		1.66E-02	
SO_3	4.45E-22		1.39E-21		4.10E-21	
SrC_2O_4	3.85E-05		3.38E-05		3.19E-05	
$SrH(C_2O_4)_2$	1.10E-02		1.07E-02		1.06E-02	
$SrHPO_4$	1.07E-12		6.00E-13		3.34E-13	
$Sr(NO_3)_2$	3.15E-05		1.11E-05		3.39E-06	
$ThH(PO_2)_4$	9.99E-12		1.63E-11		1.22E-11	
$Th(NO_3)_4$	1.64E-14		1.07E-14		2.37E-15	
$Th(OH)_4$	4.06E-25		1.32E-24		2.57E-24	
$Th(SO_4)_2$	1.35E-21		1.07E-20		4.57E-20	
$UO_2C_2O_4$	2.37E-05		4.23E-05		5.44E-05	
UO_2CO_3	2.82E-17		2.27E-17		1.40E-17	
$UO_2(OH)_2$	2.63E-19		4.23E-19		5.20E-19	
UO_2SO_4	4.07E-14		1.02E-13		1.86E-13	
$Zr(C_2O_4)_2$	6.30E-03		6.31E-03		6.31E-03	
$Zr(OH)_4$	3.50E-24		2.84E-24		2.59E-24	

Table N.4, cont.

Constituent	20 L/kg liquid (gm-mols)	20 L/kg solid (gm-mols)	40 L/kg liquid (gm-mols)	40 L/kg solid (gm-mols)	80 L/kg liquid (gm-mols)	80 L/kg solid (gm-mols)
AIBITE						
$Al(NO_3)_3.9H_2O$						
$Ca_3(PO_4)_2$						
$CaC_2O_4.1H_2O$						
$Ca(OH)_2$						
$Ce(NO_3)_3$		8.79E-03		8.79E-03		8.79E-03
$Ce(OH)_3$						
$CePO_4$						
$Cr(NO_3)_3$						
Fe_2O_3						
$Fe(NO_3)_3$						
$FePO_4.2H_2O$						
KNO_3						
$Mn_3(PO_4)_2.3H_2O$						
$Mn_3(PO_4)_2.6H_2O$						
$Mn_3(PO_4)_2$						
$MnCO_3$						
Na_2SO_4						
NaH_2PO_4						
$NaNO_2$						
NaOH						
$NiC_2O_4.2H_2O$						
$Ni(NO_3)_2$						
$Sr_3(PO_4)_2$						
$SrCO_3$						
$Sr(OH)_2.8H_2O$						
$Sr(OH)_2$						
$SrSO_4$						
ThO_2						
$(UO_2)_3(PO_4)_2.4H_2O$						
$(UO_2)_3(PO_4)_2$						
$UO_2(NO_3)_2$						
ZrO_2						
$Zr(SO_4)_2.1H_2O$						
$Zr(SO_4)_2.4H_2O$		6.92E-04		6.91E-04		6.89E-04
$Zr(SO_4)_2$						
AlION	1.08E-01		1.08E-01		1.08E-01	
$Al(OH)_2ION$	8.34E-12		6.33E-12		5.67E-12	
$Al(OH)_4ION$	6.42E-22		3.18E-22		2.32E-22	
AlOHION	9.08E-07		8.01E-07		7.65E-07	
CaH_2PO_4ION	1.55E-06		1.00E-06		5.95E-07	
$CaHC_2O_4ION$	3.69E-03		3.75E-03		3.78E-03	

Table N.4, cont.

Constituent	20 L/kg liquid (gm-mols)	20 L/kg solid (gm-mols)	40 L/kg liquid (gm-mols)	40 L/kg solid (gm-mols)	80 L/kg liquid (gm-mols)	80 L/kg solid (gm-mols)
CaHCO$_3$ION	3.26E-14		1.49E-14		7.15E-15	
CaION	6.97E-04		7.39E-04		7.55E-04	
CaOHION	3.35E-16		3.09E-16		2.99E-16	
CaPO$_4$ION	2.98E-19		1.26E-19		6.08E-20	
Ce(C$_2$O$_4$)$_2$ION	7.65E-33		4.75E-32		4.61E-31	
Ce(C$_2$O$_4$)$_3$ION	1.44E-32		4.35E-32		2.81E-31	
CeC$_2$O$_4$ION	1.05E-32		9.13E-32		1.05E-30	
CeH$_2$PO$_4$ION					2.66E-35	
CeION	6.66E-34		7.47E-33		9.55E-32	
CeNO$_3$ION	2.25E-34		1.38E-33		9.38E-33	
CeOHION						
Ce(SO$_4$)$_2$ION						
CeSO$_4$ION						
CO$_3$ION	5.66E-17		3.07E-17		2.23E-17	
CrH$_2$PO$_4$ION	1.00E-03		8.77E-04		7.08E-04	
CrH$_2$PO$_4$ION	8.15E-04		6.47E-04		5.05E-04	
CrION	1.53E-03		2.11E-03		2.86E-03	
CrNO$_3$ION	2.38E-03		2.09E-03		1.65E-03	
Cr(OH)$_2$ION	6.18E-16		6.98E-16		8.89E-16	
Cr(OH)$_4$ION	1.42E-30		1.03E-30		1.05E-30	
CrOHION	1.14E-07		1.50E-07		2.04E-07	
Fe$_2$(OH)$_2$ION	2.96E-29		1.19E-28		1.71E-28	
Fe(C$_2$O$_4$)$_2$ION	5.26E-03		1.03E-02		1.49E-02	
Fe(C$_2$O$_4$)$_3$ION	1.11E-01		1.06E-01		1.01E-01	
FeC$_2$O$_4$ION	5.91E-07		1.63E-06		2.79E-06	
FeH$_2$PO$_4$ION	1.52E-12		2.93E-12		3.03E-12	
FeHC$_2$O$_4$ION	1.66E-11		5.16E-11		9.20E-11	
FeHPO$_4$ION	4.89E-14		8.55E-14		8.57E-14	
FeION	1.25E-11		4.48E-11		8.55E-11	
FeNO$_3$ION	2.69E-14		5.21E-14		5.26E-14	
FeOH$_2$ION	9.08E-18		2.28E-17		3.75E-17	
FeOH$_4$ION	1.13E-30		1.80E-30		2.36E-30	
FeOHION	1.52E-13		4.53E-13		8.00E-13	
H$_2$P$_2$O$_7$ION	1.28E-08		7.26E-09		4.11E-09	
H$_2$PO$_4$ION	4.58E-03		4.90E-03		5.24E-03	
H$_3$P$_2$O$_7$ION	8.32E-08		6.57E-08		4.44E-08	
H$_3$SiO$_4$ION	2.88E-11		2.18E-11		1.89E-11	
HCO$_3$ION	3.89E-08		2.95E-08		2.57E-08	
HION	3.50E+00		8.37E+00		1.82E+01	
HOXAIATION	6.75E+00		1.16E+01		2.14E+01	
HP$_2$O$_7$ION	1.36E-13		5.75E-14		2.78E-14	
HPO$_4$ION	9.19E-09		7.01E-09		6.26E-09	

Table N.4, cont.

Constituent	20 L/kg liquid (gm-mols)	20 L/kg solid (gm-mols)	40 L/kg liquid (gm-mols)	40 L/kg solid (gm-mols)	80 L/kg liquid (gm-mols)	80 L/kg solid (gm-mols)
HSO_4ION	9.51E-07		2.26E-06		5.82E-06	
KION	7.54E-02		7.54E-02		7.54E-02	
KSO_4ION	1.75E-09		1.78E-09		2.13E-09	
$Mn(C_2O_4)_2$ION	3.36E-04		1.77E-04		1.28E-04	
$Mn(C_2O_4)_3$ION	3.48E-05		7.54E-06		3.26E-06	
MNiON	2.06E-02		2.18E-02		2.24E-02	
$MnNO_3$ION	9.79E-04		6.12E-04		3.48E-04	
$MnOH_3$ION	5.79E-35		3.47E-35		2.72E-35	
$MnOH_4$ION						
MnOHION	3.17E-12		2.97E-12		2.92E-12	
$NaCO_3$ION	9.09E-18		3.03E-18		1.24E-18	
NaION	5.16E+00		5.20E+00		5.22E+00	
$NaSO_4$ION	1.64E-08		1.68E-08		2.03E-08	
$Ni(C_2O_4)_2$ION	3.36E-04		2.12E-04		1.63E-04	
NiION	2.93E-04		3.72E-04		4.09E-04	
$NiNO_3$ION	2.23E-05		1.66E-05		1.01E-05	
$NiOH_3$ION	5.29E-32		3.78E-32		3.18E-32	
NiOHION	1.11E-13		1.24E-13		1.30E-13	
NO_2ION	4.56E-04		3.48E-04		3.03E-04	
NO_3ION	2.00E+00		2.04E+00		2.06E+00	
OHION	2.21E-12		3.54E-12		6.35E-12	
OXAIATION	1.28E-02		1.58E-02		2.43E-02	
P_2O_7ION	1.29E-20		3.55E-21		1.34E-21	
PO_4ION	2.20E-19		1.22E-19		9.18E-20	
SO_4ION	4.41E-07		7.52E-07		1.62E-06	
$SrHC_2O_4$ION	1.03E-02		1.06E-02		1.08E-02	
SrION	3.39E-03		3.58E-03		3.65E-03	
$SrNO_3$ION	5.06E-04		3.13E-04		1.76E-04	
SrOHION	5.96E-16		5.53E-16		5.36E-16	
$SrPO_4$ION	1.85E-20		7.82E-21		3.77E-21	
Th_2OH_2ION	1.77E-27		1.63E-26		3.56E-26	
$Th(C_2O_4)_3$ION	1.12E-11		2.70E-11		4.40E-11	
$Th(C_2O_4)_4$ION	4.50E-03		4.44E-03		4.34E-03	
ThC_2O_4ION	4.54E-07		2.16E-06		4.98E-06	
$ThH_2(PO_4)_2$ION	8.55E-10		1.80E-09		1.49E-09	
ThH_2PO_4ION	1.83E-10		5.55E-10		7.31E-10	
$ThHC_2O_4$ION	1.56E-05		8.01E-05		1.87E-04	
$ThH(PO_4)_3$ION	2.69E-05		1.69E-05		5.63E-06	
$ThHPO_4$ION	9.36E-13		2.78E-12		3.72E-12	
ThION	3.77E-12		2.17E-11		5.30E-11	
$Th(NO_3)_2$ION	3.05E-34		6.08E-34		4.61E-34	
$Th(NO_3)_3$ION	2.78E-14		3.19E-14		1.32E-14	

Table N.4, cont.

Constituent	20 L/kg liquid (gm-mols)	20 L/kg solid (gm-mols)	40 L/kg liquid (gm-mols)	40 L/kg solid (gm-mols)	80 L/kg liquid (gm-mols)	80 L/kg solid (gm-mols)
ThNO$_3$ION	1.09E-12		3.69E-12		5.04E-12	
Th(OH)$_2$ION	1.22E-18		5.60E-18		1.28E-17	
Th(OH)$_3$ION	3.30E-23		1.27E-22		2.70E-22	
ThOHION	5.75E-16		3.15E-15		7.74E-15	
Th(SO$_4$)$_3$ION	1.79E-28		1.20E-27		5.49E-27	
Th(SO$_4$)$_4$ION					1.49E-35	
ThSO$_4$ION	1.88E-16		1.23E-15		3.94E-15	
(UO$_2$)$_2$(OH)$_2$ION	3.67E-21		6.71E-21		5.91E-21	
(UO$_2$)$_3$(OH)$_5$ION						
UO$_2$(C$_2$O$_4$)$_2$ION	1.27E-04		1.37E-04		1.33E-04	
UO$_2$(C$_2$O$_4$)$_3$ION	4.97E-05		2.19E-05		1.29E-05	
UO$_2$(CO$_3$)$_2$ION	1.91E-27		4.17E-28		9.29E-29	
UO$_2$(CO$_3$)$_3$ION						
UO$_2$ION	4.61E-08		1.04E-07		1.49E-07	
UO$_2$OHION	8.32E-13		1.59E-12		2.11E-12	
UO$_2$(SO$_4$)$_2$ION	7.29E-21		1.58E-20		3.12E-20	
ZrC$_2$O$_4$ION	1.51E-08		1.80E-08		1.93E-08	
ZrION	2.14E-15		2.83E-15		3.08E-15	
Zr(NO$_3$)$_2$ION	5.20E-24		2.60E-24		9.18E-25	
ZrNO$_3$ION	2.56E-22		2.17E-22		1.38E-22	
Zr(OH)$_2$ION	2.24E-17		2.57E-17		2.73E-17	
Zr(OH)$_3$ION	2.72E-20		2.62E-20		2.58E-20	
Zr(OH)$_5$ION	2.23E-29		1.37E-29		1.08E-29	
ZrOHION	1.79E-15		2.07E-15		2.14E-15	
ZrSO$_4$ION	2.59E-25		4.23E-25		6.31E-25	

Table N.5. Speciation of Modeled Sludge After Oxalic Acid Addition at 60 and 100 L/kg

Constituent	60 L/kg liquid (gram-mols)	60 L/kg solid (gram-mols)	100 L/kg liquid (gram-mols)	100 L/kg solid (gram-mols)
H_2O	3.10E+03		5.15E+03	
$Al(OH)_3$	1.23E-16		1.12E-16	
CaC_2O_4	2.18E-05		2.11E-05	
$CaCO_3$	1.12E-26		6.35E-27	
$CaH(C_2O_4)_2$	2.49E-03		2.47E-03	
$CaSO_4$	9.20E-11		1.15E-10	
CO_2	7.34E-03		7.34E-03	
$CrOH_3$	1.05E-20		1.21E-20	
$FeOH_3$	1.02E-21		1.28E-21	
H_2SO_4	1.98E-17		4.37E-17	
HNO_2	6.43E-02		6.43E-02	
HNO_3	1.20E-02		1.31E-02	
$KHSO_4$	5.45E-11		6.96E-11	
MnC_2O_4	3.94E-03		3.85E-03	
$Mn(NO_3)_2$	2.19E-05		8.82E-06	
$Mn(OH)_2$	1.36E-23		1.29E-23	
$MnSO_4$	4.69E-09		5.90E-09	
$NaHCO_3$	1.82E-09		1.05E-09	
$NaNO_3$	3.45E-02		2.17E-02	
NiC_2O_4	1.12E-03		1.13E-03	
$Ni(OH)_2$	3.89E-22		3.83E-22	
$NiSO_4$	7.79E-11		1.01E-10	
OXAlAC	3.63E+01		6.21E+01	
SiO_2	1.66E-02		1.66E-02	
SO_3	2.63E-21		5.78E-21	
SrC_2O_4	3.25E-05		3.16E-05	
$SrH(C_2O_4)_2$	1.07E-02		1.06E-02	
$SrHPO_4$	4.27E-13		2.76E-13	
$Sr(NO_3)_2$	5.63E-06		2.26E-06	
$ThH(PO_2)_4$	1.47E-11		9.91E-12	
$Th(NO_3)_4$	4.87E-15		1.27E-15	
$Th(OH)_4$	2.05E-24		2.97E-24	
$Th(SO_4)_2$	2.65E-20		6.70E-20	
$UO_2C_2O_4$	5.02E-05		5.71E-05	
UO_2CO_3	1.74E-17		1.16E-17	
$UO_2(OH)_2$	4.86E-19		5.40E-19	
UO_2SO_4	1.48E-13		2.18E-13	
$Zr(C_2O_4)_2$	6.31E-03		6.31E-03	
$Zr(OH)_4$	2.67E-24		2.55E-24	

Table N.5, cont.

Constituent	60 L/kg liquid (gram-mols)	60 L/kg solid (gram-mols)	100 L/kg liquid (gram-mols)	100 L/kg solid (gram-mols)
AlBITE				
$Al(NO_3)_3.9H_2O$				
$Ca_3(PO_4)_2$				
$CaC_2O_4.1H_2O$				
$Ca(OH)_2$				
$Ce(NO_3)_3$		8.79E-03		8.79E-03
$Ce(OH)_3$				
$CePO_4$				
$Cr(NO_3)_3$				
Fe_2O_3				
$Fe(NO_3)_3$				
$FePO_4.2H_2O$				
KNO_3				
$Mn_3(PO_4)_2.3H_2O$				
$Mn_3(PO_4)_2.6H_2O$				
$Mn_3(PO_4)_2$				
$MnCO_3$				
Na_2SO_4				
NaH_2PO_4				
$NaNO_2$				
NaOH				
$NiC_2O_4.2H_2O$				
$Ni(NO_3)_2$				
$Sr_3(PO_4)_2$				
$SrCO_3$				
$Sr(OH)_2.8H_2O$				
$Sr(OH)_2$				
$SrSO_4$				
ThO_2				
$(UO_2)_3(PO_4)_2.4H_2O$				
$(UO_2)_3(PO_4)_2$				
$UO_2(NO_3)_2$				
ZrO_2				
$Zr(SO_4)_2.1H_2O$				
$Zr(SO_4)_2.4H_2O$		6.90E-04		6.88E-04
$Zr(SO_4)_2$				
AlION	1.08E-01		1.08E-01	
$Al(OH)_2ION$	5.87E-12		5.57E-12	
$Al(OH)_4ION$	2.57E-22		2.18E-22	
AlOHION	7.75E-07		7.60E-07	
CaH_2PO_4ION	7.45E-07		4.96E-07	
$CaHC_2O_4ION$	3.77E-03		3.78E-03	

Table N.5, cont.

Constituent	60 L/kg liquid (gram-mols)	60 L/kg solid (gram-mols)	100 L/kg liquid (gram-mols)	100 L/kg solid (gram-mols)
CaHCO$_3$ION	9.66E-15		5.68E-15	
CaION	7.50E-04		7.57E-04	
CaOHION	3.02E-16		2.97E-16	
CaPO$_4$ION	8.15E-20		4.87E-20	
Ce(C$_2$O$_4$)$_2$ION	1.73E-31		1.01E-30	
Ce(C$_2$O$_4$)$_3$ION	1.21E-31		5.67E-31	
CeC$_2$O$_4$ION	3.71E-31		2.39E-30	
CeH$_2$PO$_4$ION	1.17E-35		5.07E-35	
CeION	3.27E-32		2.22E-31	
CeNO$_3$ION	4.19E-33		1.77E-32	
CeOHION				
Ce(SO$_4$)$_2$ION				
CeSO$_4$ION				
CO$_3$ION	2.49E-17		2.09E-17	
CrH$_2$PO$_4$ION	7.83E-04		6.47E-04	
CrH$_2$PO$_4$ION	5.64E-04		4.59E-04	
CrION	2.53E-03		3.13E-03	
CrNO$_3$ION	1.85E-03		1.49E-03	
Cr(OH)$_2$ION	8.01E-16		9.62E-16	
Cr(OH)$_4$ION	1.02E-30		1.09E-30	
CrOHION	1.80E-07		2.23E-07	
Fe$_2$(OH)$_2$ION	1.60E-28		1.69E-28	
Fe(C$_2$O$_4$)$_2$ION	1.31E-02		1.60E-02	
Fe(C$_2$O$_4$)$_3$ION	1.03E-01		1.00E-01	
FeC$_2$O$_4$ION	2.33E-06		3.11E-06	
FeH$_2$PO$_4$ION	3.15E-12		2.83E-12	
FeHC$_2$O$_4$ION	7.58E-11		1.03E-10	
FeHPO$_4$ION	8.98E-14		7.97E-14	
FeION	6.89E-11		9.74E-11	
FeNO$_3$ION	5.53E-14		4.86E-14	
FeOH$_2$ION	3.17E-17		4.15E-17	
FeOH$_4$ION	2.15E-30		2.50E-30	
FeOHION	6.60E-13		8.98E-13	
H$_2$P$_2$O$_7$ION	5.20E-09		3.42E-09	
H$_2$PO$_4$ION	5.10E-03		5.36E-03	
H$_3$P$_2$O$_7$ION	5.29E-08		3.83E-08	
H$_3$SiO$_4$ION	1.99E-11		1.84E-11	
HCO$_3$ION	2.69E-08		2.49E-08	
HION	1.33E+01		2.31E+01	
HOXAIATION	1.65E+01		2.63E+01	
HP$_2$O$_7$ION	3.71E-14		2.24E-14	
HPO$_4$ION	6.47E-09		6.16E-09	

Table N.5, cont.

Constituent	60 L/kg liquid (gram-mols)	60 L/kg solid (gram-mols)	100 L/kg liquid (gram-mols)	100 L/kg solid (gram-mols)
HSO_4ION	3.91E-06		7.98E-06	
KION	7.54E-02		7.54E-02	
KSO_4ION	1.95E-09		2.30E-09	
$Mn(C_2O_4)_2$ION	1.43E-04		1.20E-04	
$Mn(C_2O_4)_3$ION	4.34E-06		2.73E-06	
MNiON	2.22E-02		2.25E-02	
$MnNO_3$ION	4.44E-04		2.86E-04	
$MnOH_3$ION	2.95E-35		2.60E-35	
$MnOH_4$ION				
MnOHION	2.93E-12		2.91E-12	
$NaCO_3$ION	1.77E-18		9.56E-19	
NaION	5.21E+00		5.23E+00	
$NaSO_4$ION	1.86E-08		2.20E-08	
$Ni(C_2O_4)_2$ION	1.79E-04		1.55E-04	
NiION	3.97E-04		4.16E-04	
$NiNO_3$ION	1.26E-05		8.42E-06	
$NiOH_3$ION	3.37E-32		3.07E-32	
NiOHION	1.28E-13		1.31E-13	
NO_2ION	3.18E-04		2.95E-04	
NO_3ION	2.05E+00		2.06E+00	
OHION	4.94E-12		7.76E-12	
OXAIATION	1.99E-02		2.88E-02	
P_2O_7ION	1.96E-21		1.03E-21	
PO_4ION	1.01E-19		8.70E-20	
SO_4ION	1.16E-06		2.14E-06	
$SrHC_2O_4$ION	1.08E-02		1.09E-02	
SrION	3.63E-03		3.66E-03	
$SrNO_3$ION	2.25E-04		1.44E-04	
SrOHION	5.41E-16		5.33E-16	
$SrPO_4$ION	5.06E-21		3.02E-21	
Th_2OH_2ION	2.91E-26		3.82E-26	
$Th(C_2O_4)_3$ION	3.73E-11		4.87E-11	
$Th(C_2O_4)_4$ION	4.39E-03		4.31E-03	
ThC_2O_4ION	3.75E-06		5.91E-06	
$ThH_2(PO_4)_2$ION	1.75E-09		1.24E-09	
ThH_2PO_4ION	7.00E-10		7.17E-10	
$ThHC_2O_4$ION	1.41E-04		2.22E-04	
$ThH(PO_4)_3$ION	9.41E-06		3.61E-06	
$ThHPO_4$ION	3.53E-12		3.67E-12	
ThION	3.93E-11		6.34E-11	
$Th(NO_3)_2$ION	5.63E-34		3.71E-34	
$Th(NO_3)_3$ION	2.08E-14		8.65E-15	

Table N.5, cont.

Constituent	60 L/kg liquid (gram-mols)	60 L/kg solid (gram-mols)	100 L/kg liquid (gram-mols)	100 L/kg solid (gram-mols)
$\mathrm{ThNO_3ION}$	4.79E-12		4.95E-12	
$\mathrm{Th(OH)_2ION}$	9.65E-18		1.51E-17	
$\mathrm{Th(OH)_3ION}$	2.09E-22		3.15E-22	
ThOHION	5.72E-15		9.30E-15	
$\mathrm{Th(SO_4)_3ION}$	3.03E-27		8.48E-27	
$\mathrm{Th(SO_4)_4ION}$			2.29E-35	
$\mathrm{ThSO_4ION}$	2.59E-15		5.21E-15	
$\mathrm{(UO_2)_2(OH)_2ION}$	6.56E-21		5.27E-21	
$\mathrm{(UO_2)_3(OH)_5ION}$				
$\mathrm{UO_2(C_2O_4)_2ION}$	1.35E-04		1.32E-04	
$\mathrm{UO_2(C_2O_4)_3ION}$	1.55E-05		1.14E-05	
$\mathrm{UO_2(CO_3)_2ION}$	1.72E-28		5.79E-29	
$\mathrm{UO_2(CO_3)_3ION}$				
$\mathrm{UO_2ION}$	1.33E-07		1.60E-07	
$\mathrm{UO_2OHION}$	1.92E-12		2.23E-12	
$\mathrm{UO_2(SO_4)_2ION}$	2.36E-20		3.87E-20	
$\mathrm{ZrC_2O_4ION}$	1.89E-08		1.96E-08	
ZrION	3.01E-15		3.11E-15	
$\mathrm{Zr(NO_3)_2ION}$	1.45E-24		6.30E-25	
$\mathrm{ZrNO_3ION}$	1.70E-22		1.16E-22	
$\mathrm{Zr(OH)_2ION}$	2.68E-17		2.76E-17	
$\mathrm{Zr(OH)_3ION}$	2.59E-20		2.57E-20	
$\mathrm{Zr(OH)_5ION}$	1.17E-29		1.03E-29	
ZrOHION	2.13E-15		2.15E-15	
$\mathrm{ZrSO_4ION}$	5.38E-25		7.12E-25	

Appendix O

Computer Modeling of Oxalate-Treated Sludge at Thermodynamic Equilibrium: Plots of Predicted Sludge Element Solubility

Note: in this Appendix, the y-axis on many of the plots is % dissolved. In these plots, 0% dissolved means that the amount of the element initially present before acid addition is present. Hence, if precipitation occurs as acid is added, as in the case of aluminum, the % dissolved becomes a negative number.

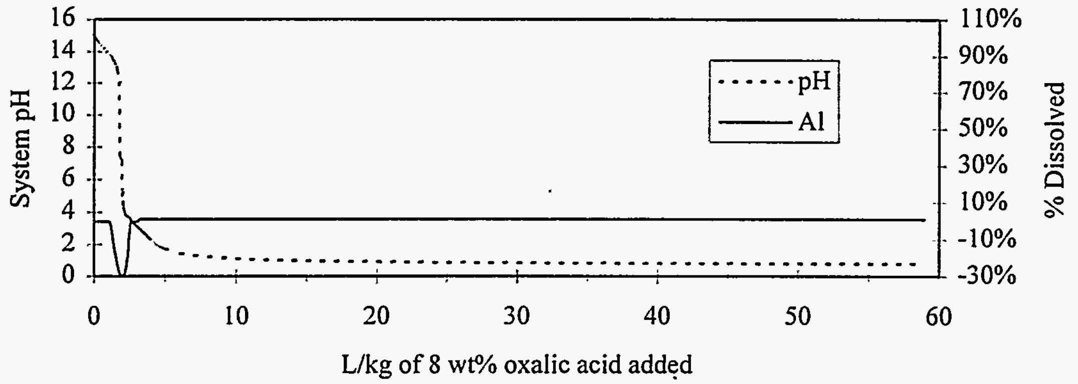

Figure O.1(a). Percent of Modeled Sludge Al Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

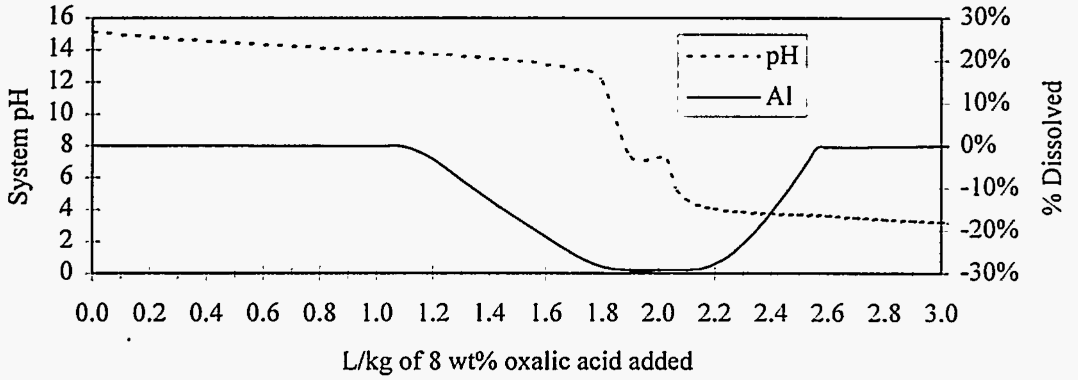

Figure O.1(b). Percent of Modeled Sludge Al Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

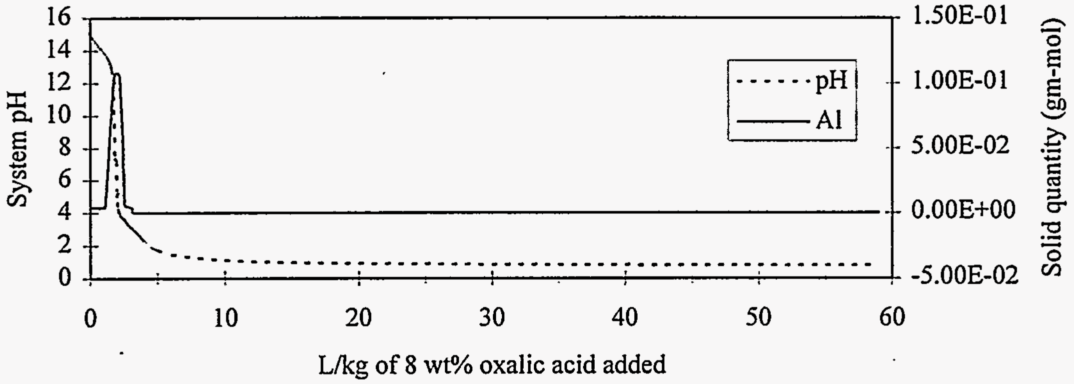

Figure O.1(c). Aluminum Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

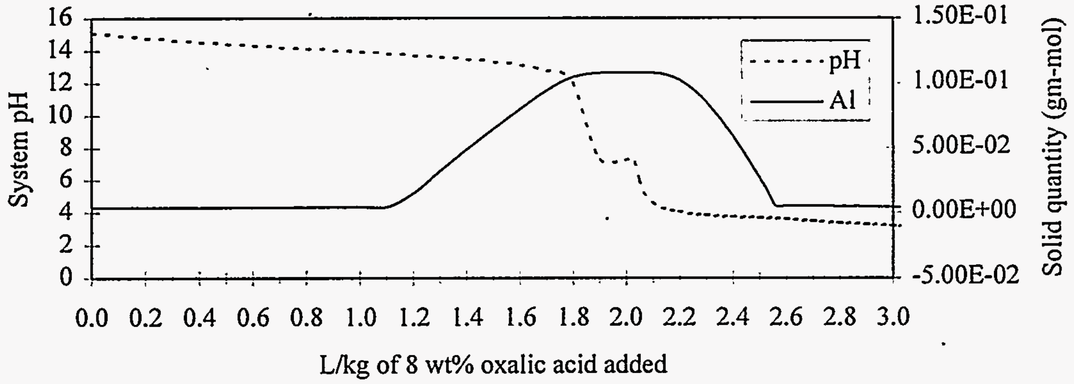

Figure O.1(d). Aluminum Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

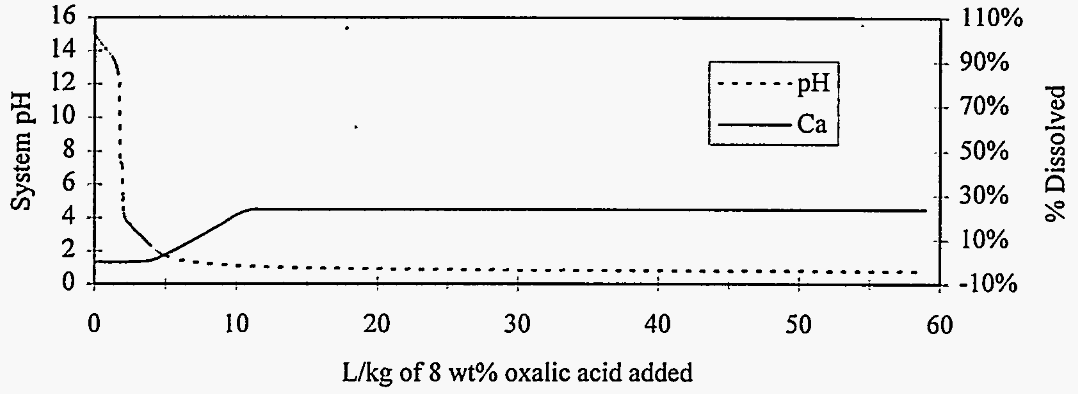

Figure O.2(a). Percent of Modeled Sludge Ca Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

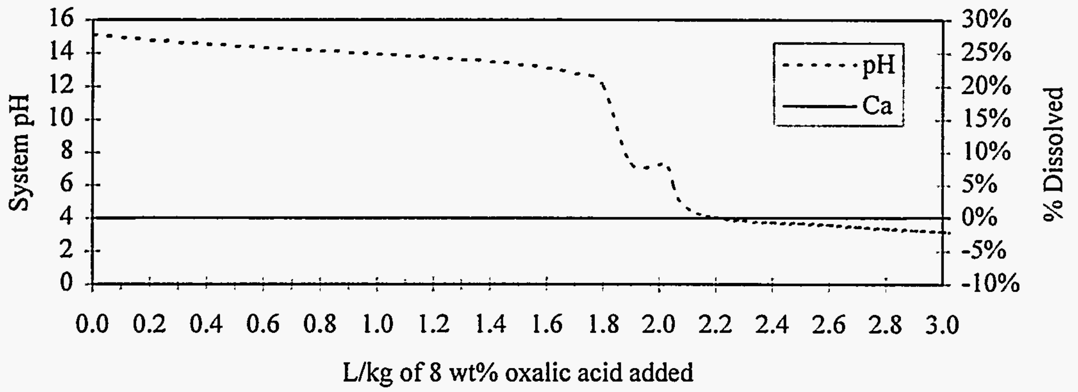

Figure O.2(b). Percent of Modeled Sludge Ca Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

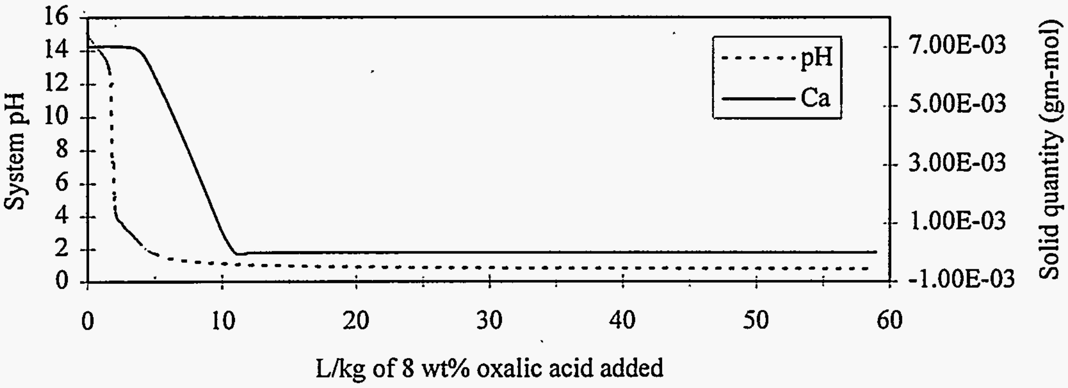

Figure O.2(c). Calcium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

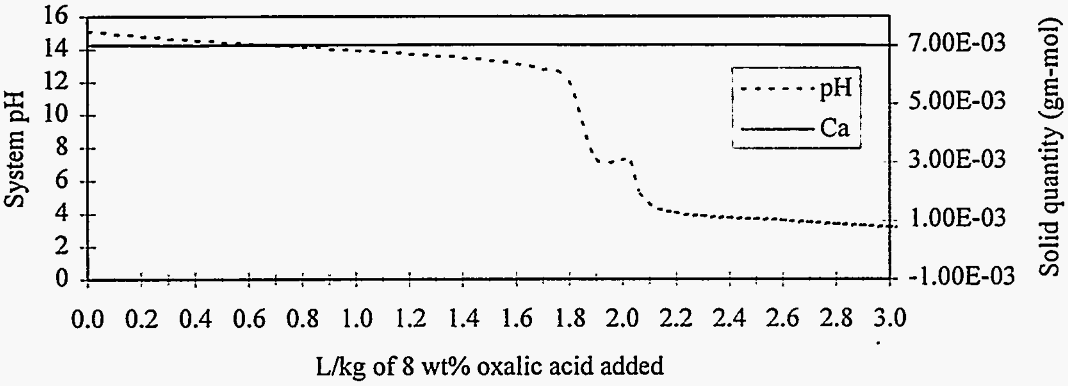

Figure O.2(c). Calcium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

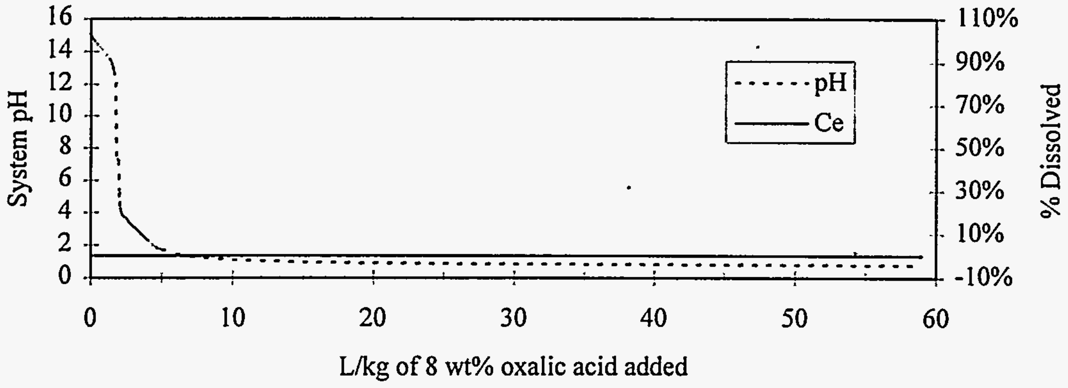

Figure O.3(a). Percent of Modeled Sludge Ce Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

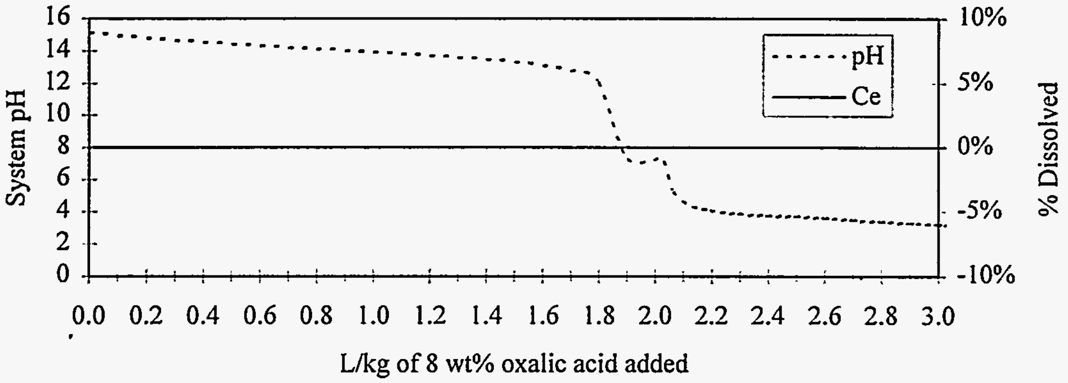

Figure O.3(b). Percent of Modeled Sludge Ce Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

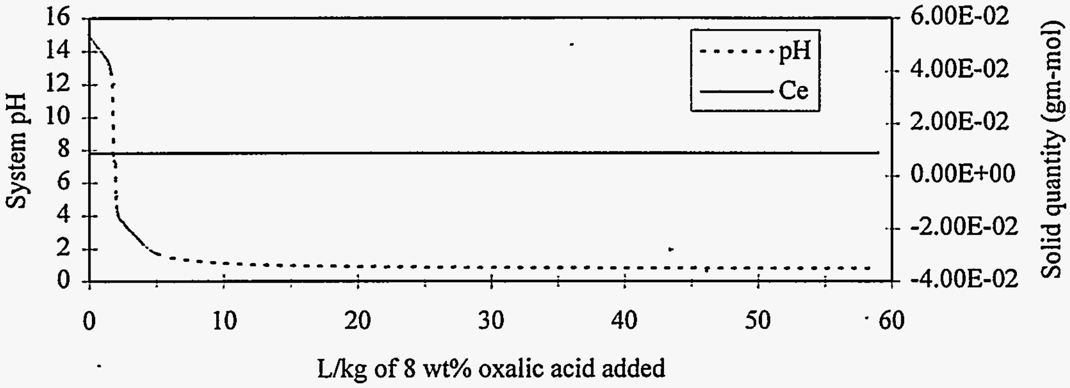

Figure O.3(c). Cerium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

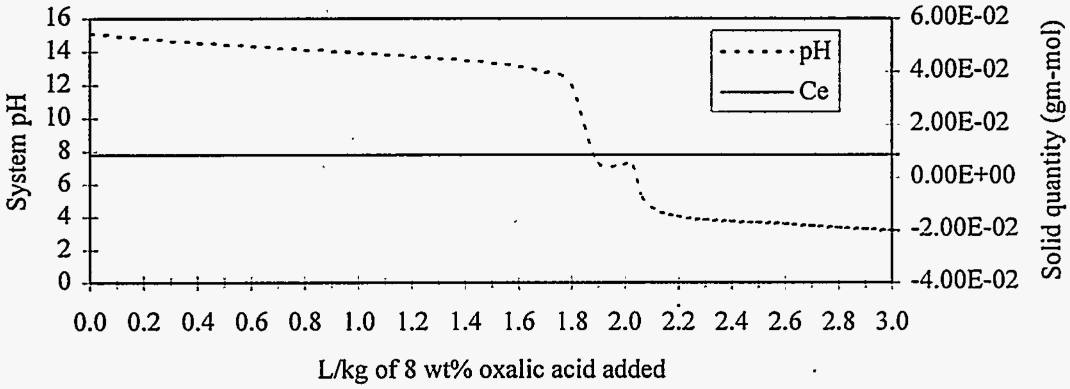

Figure O.3(d). Cerium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

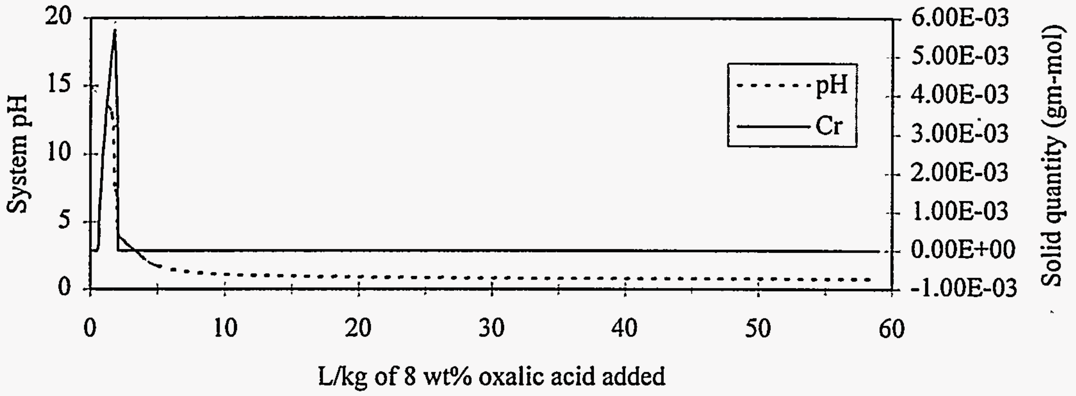

Figure O.4(a). Chromium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

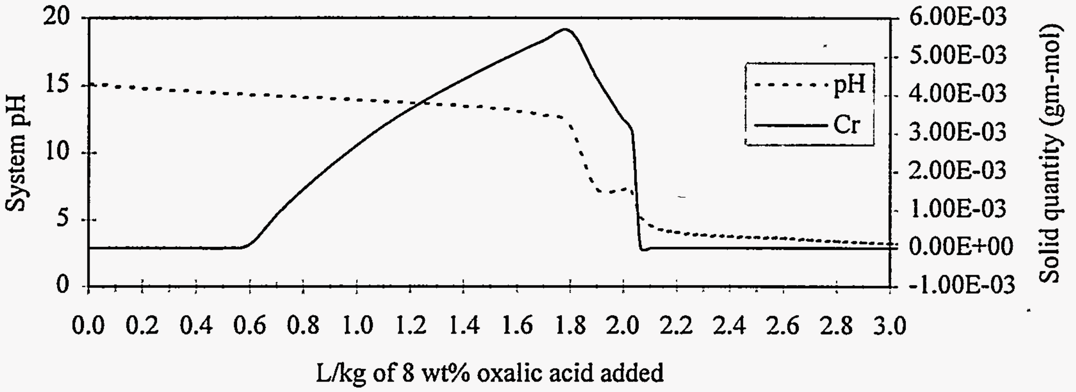

Figure O.4(b). Chromium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

(Note that percent dissolved data was not available for Cr)

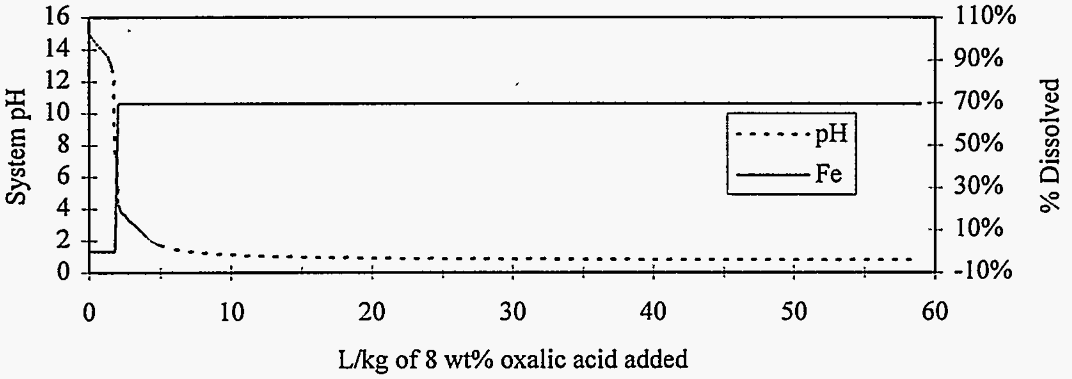

Figure O.5(a). Percent of Modeled Sludge Fe Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

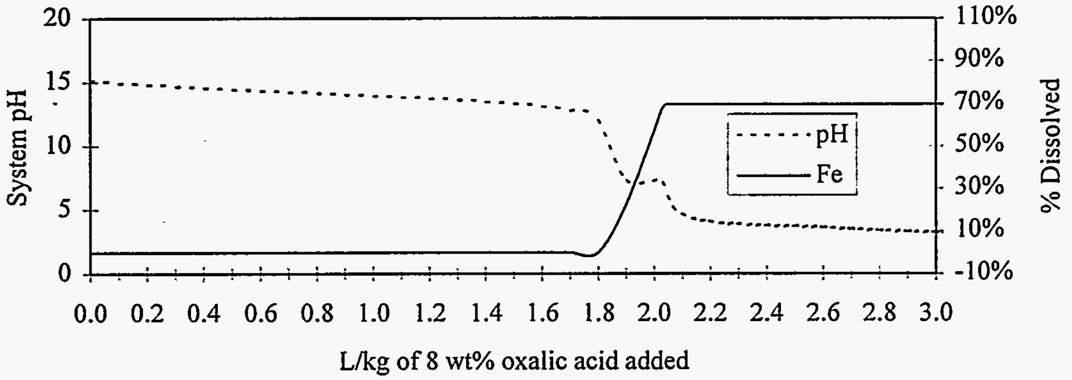

Figure O.5(b). Percent of Modeled Sludge Fe Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

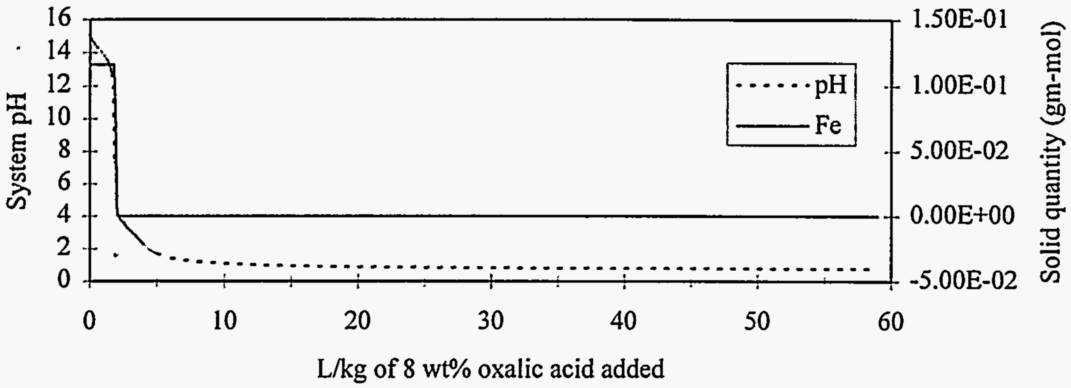

Figure O.5(c). Iron Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

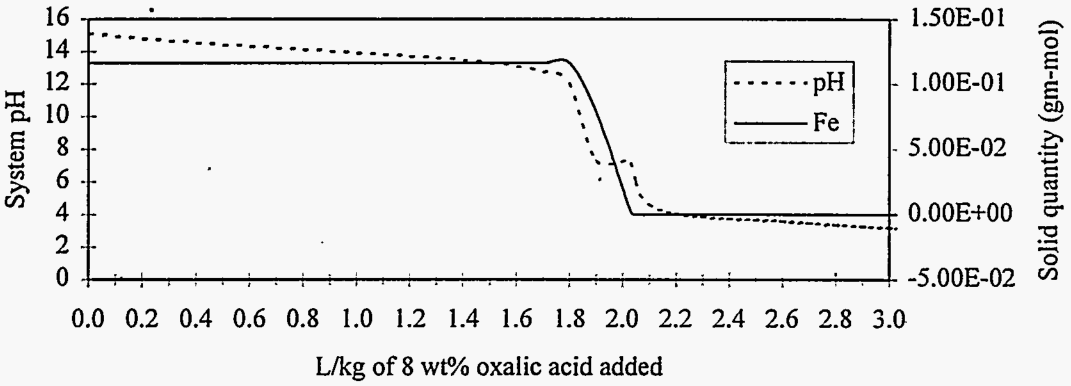

Figure O.5(d). Iron Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

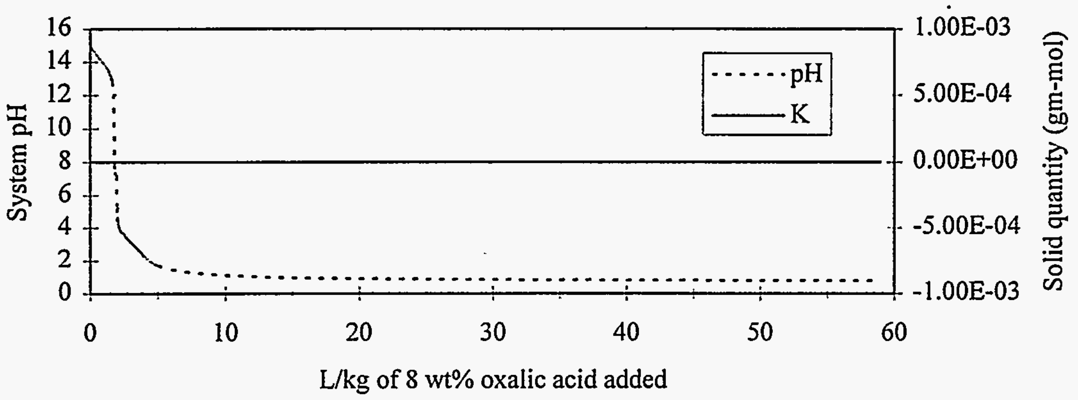

Figure O.6(a). Potassium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

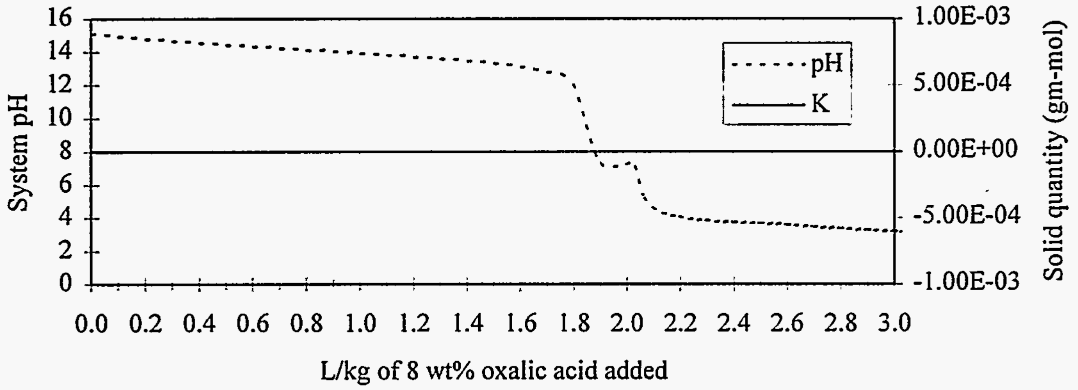

Figure O.6(b). Potassium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

(Note that percent dissolved data was not available for K)

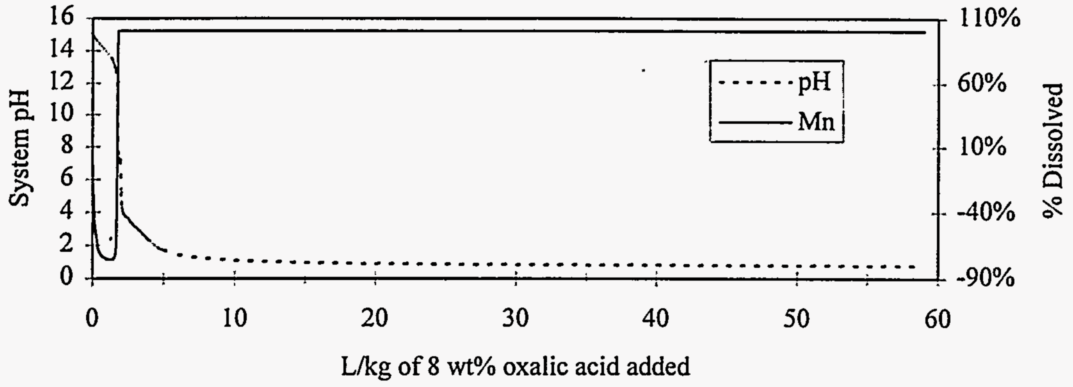

Figure O.7(a). Percent of Modeled Sludge Mn Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

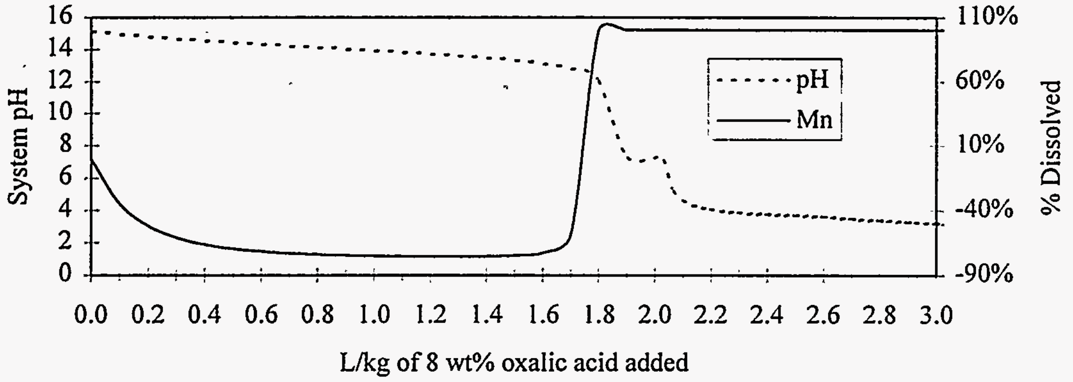

Figure O.7(b). Percent of Modeled Sludge Mn Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

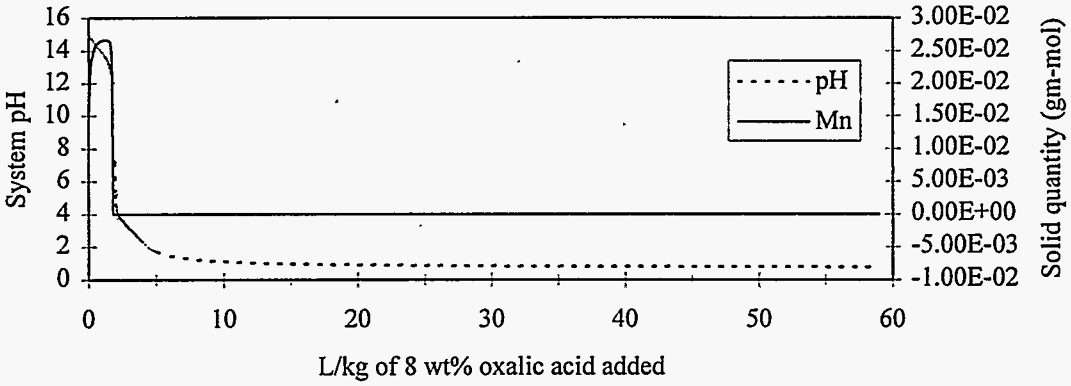

Figure O.7(c). Manganese Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

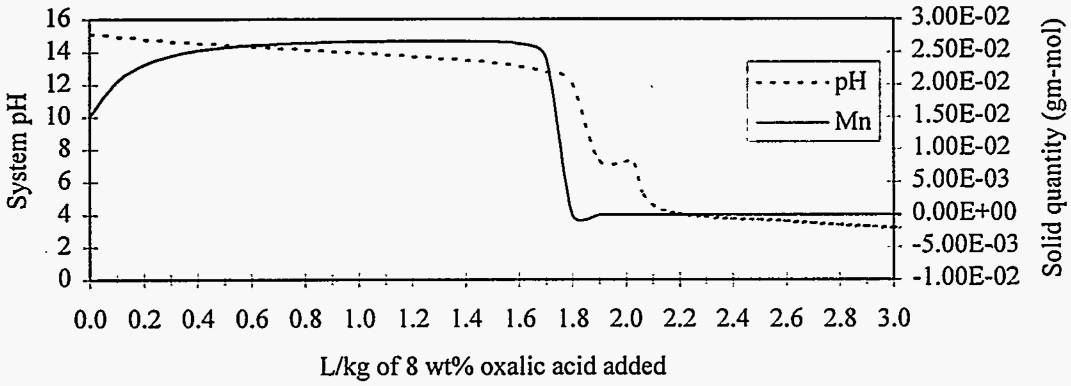

Figure O.7(d). Manganese Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

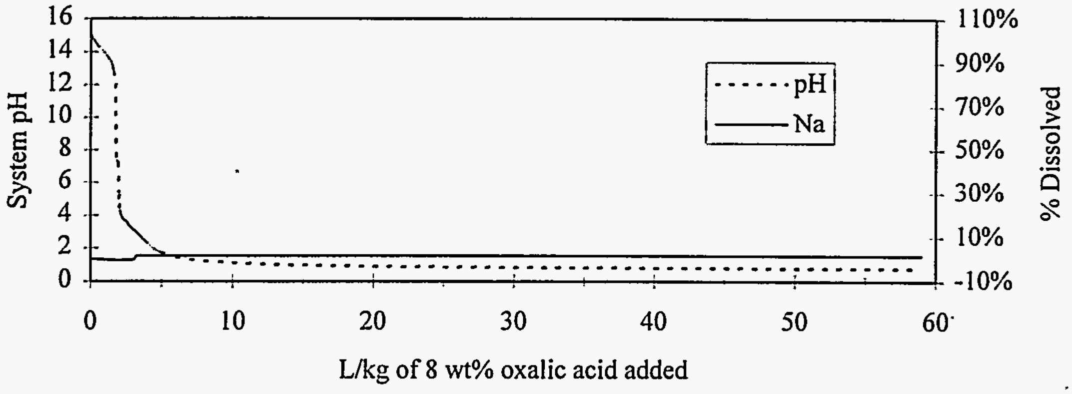

Figure O.8(a). Percent of Modeled Sludge Na Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

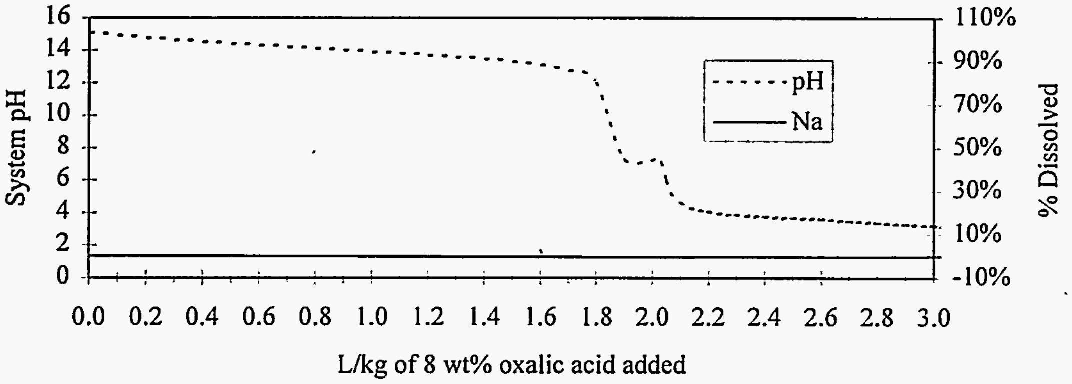

Figure O.8(b). Percent of Modeled Sludge Na Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

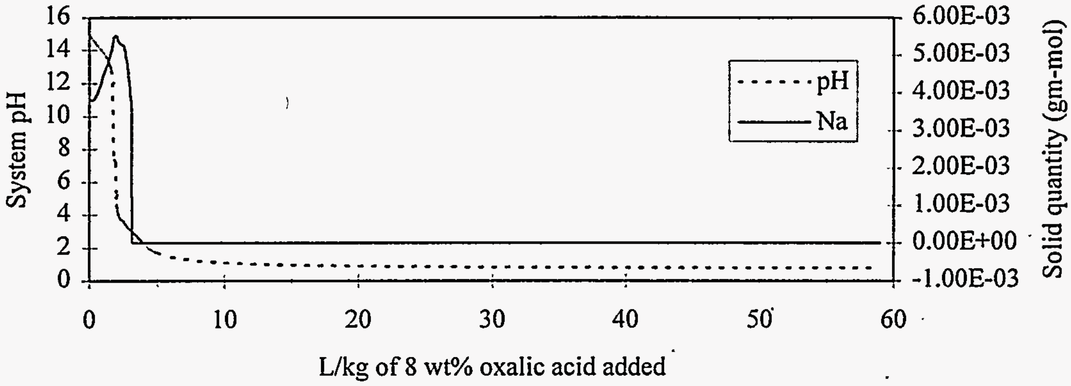

Figure O.8(c). Sodium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

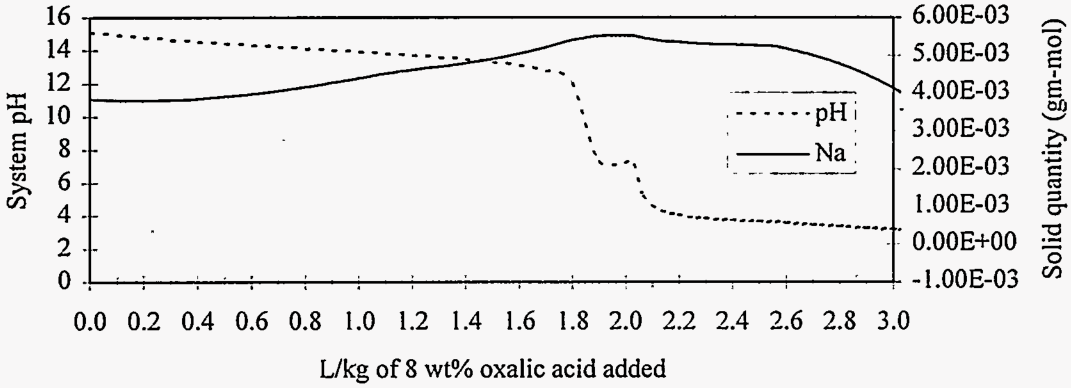

Figure O.8(d). Sodium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

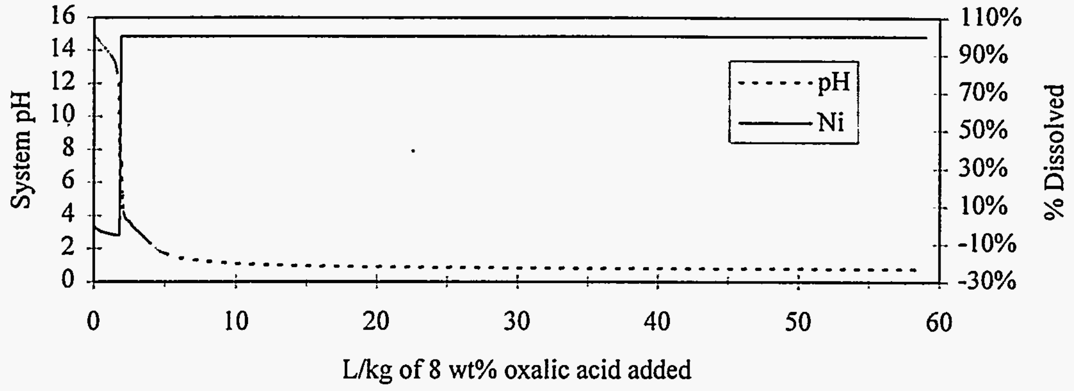

Figure O.9(a). Percent of Modeled Sludge Ni Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

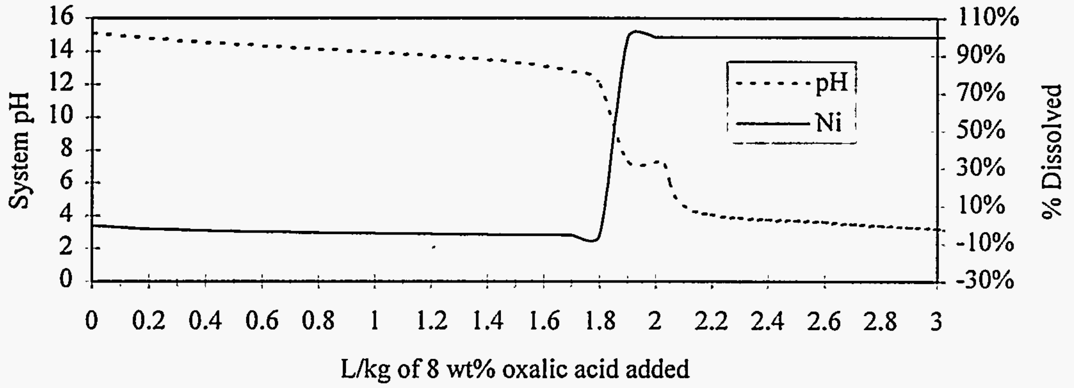

Figure O.9(b). Percent of Modeled Sludge Ni Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

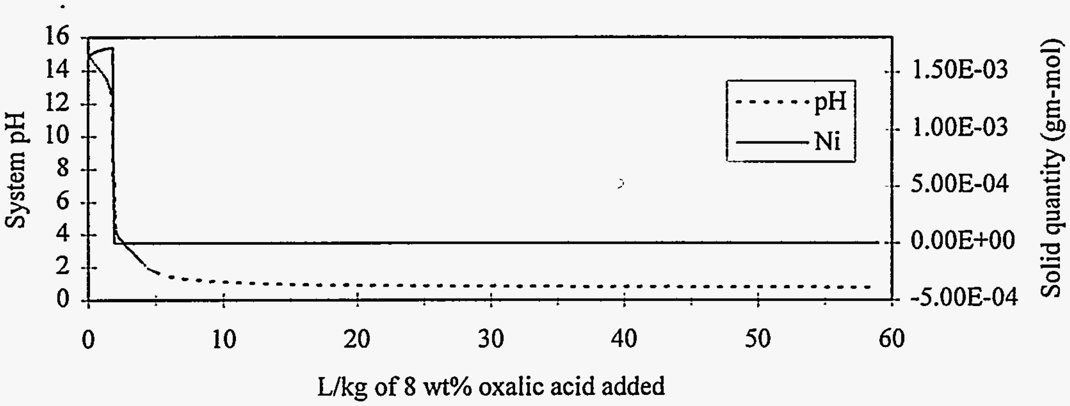

Figure O.9(c). Nickel Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

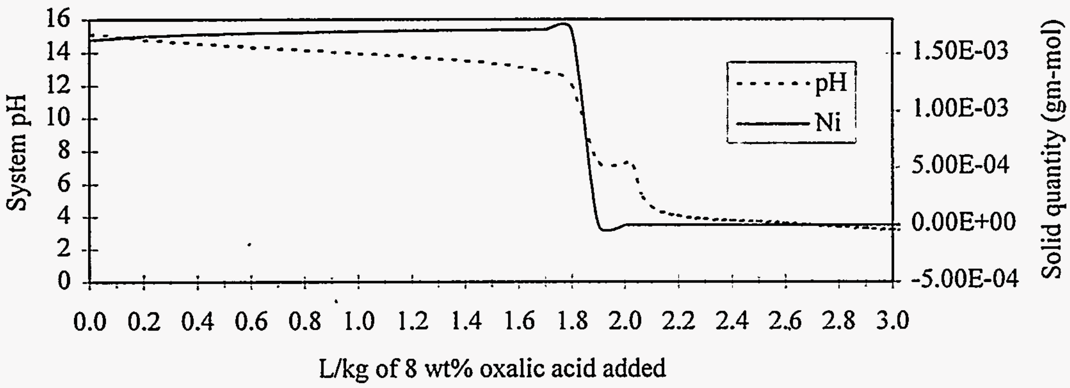

Figure O.9(d). Nickel Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

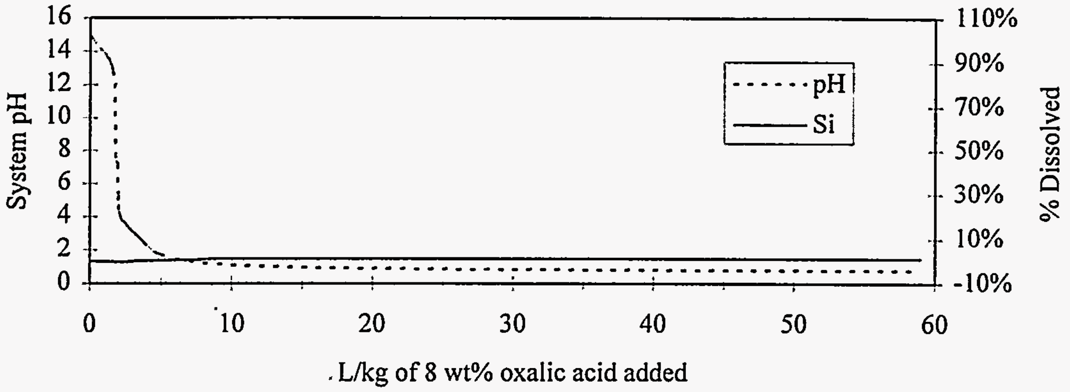

Figure O.10(a). Percent of Modeled Sludge Si Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

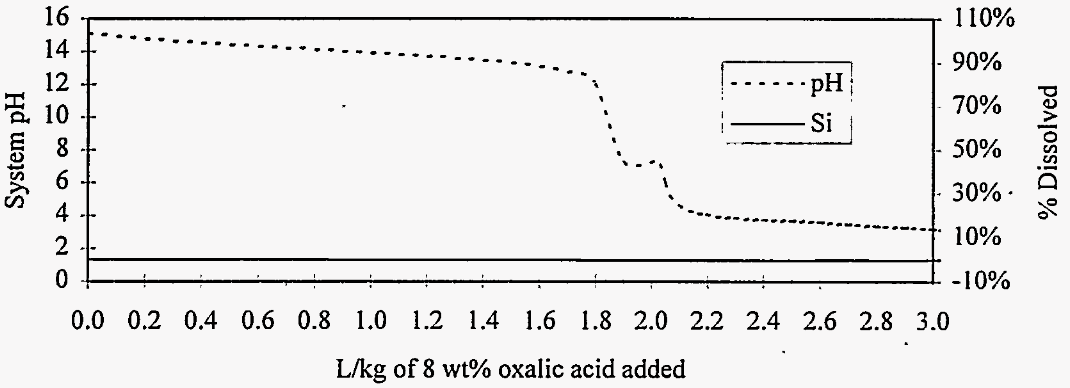

Figure O.10(b). Percent of Modeled Sludge Si Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

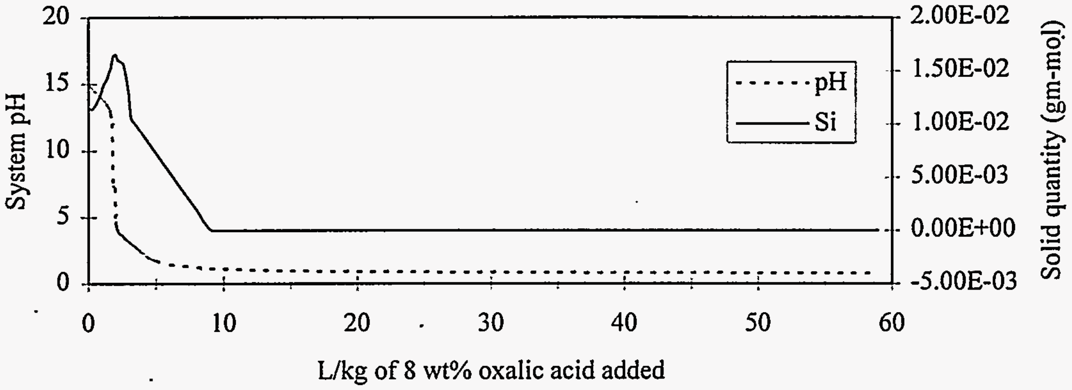

Figure O.10(c). Silicon Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

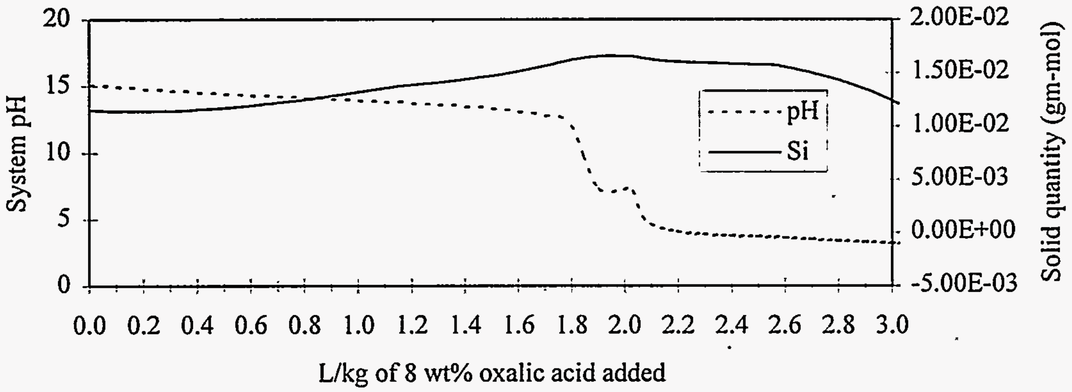

Figure O.10(d). Silicon Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

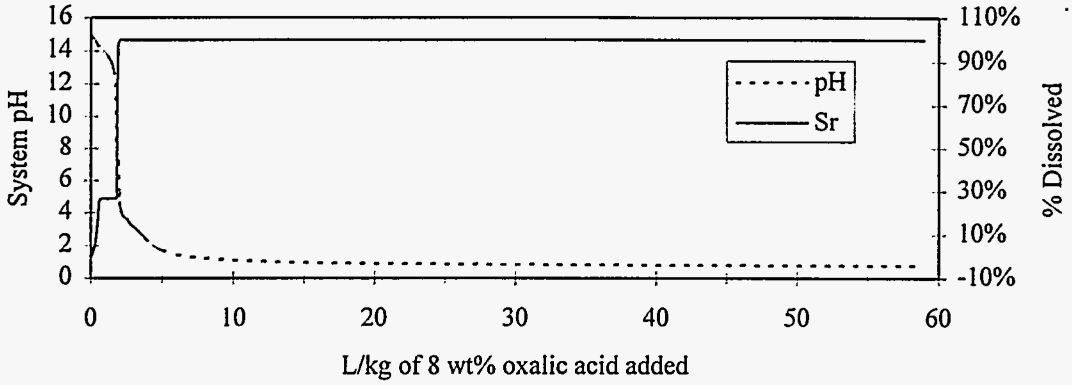

Figure O.11(a). Percent of Modeled Sludge Sr Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

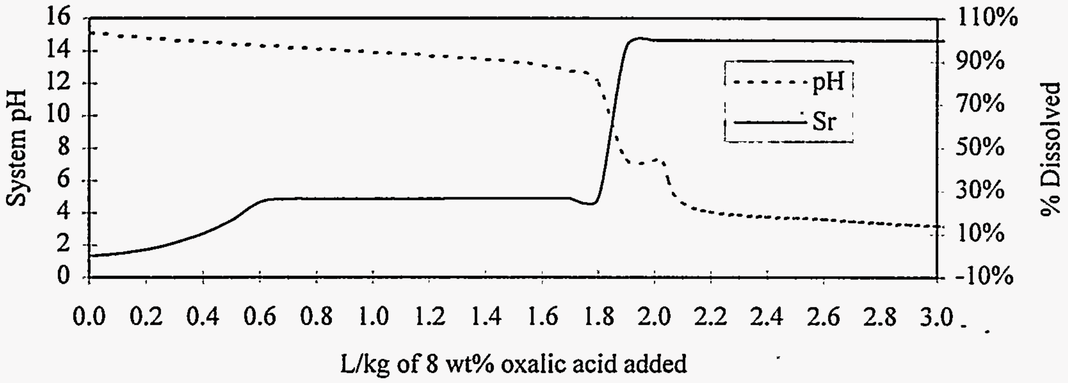

Figure O.11(b). Percent of Modeled Sludge Sr Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

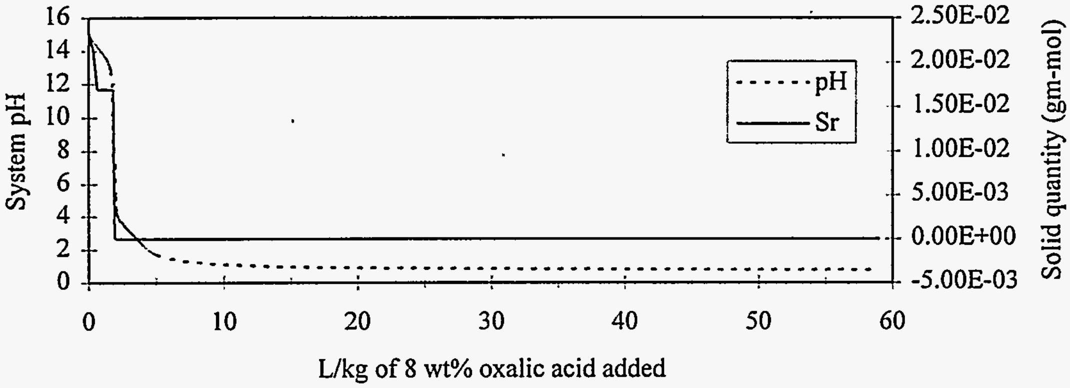

Figure O.11(c). Strontium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

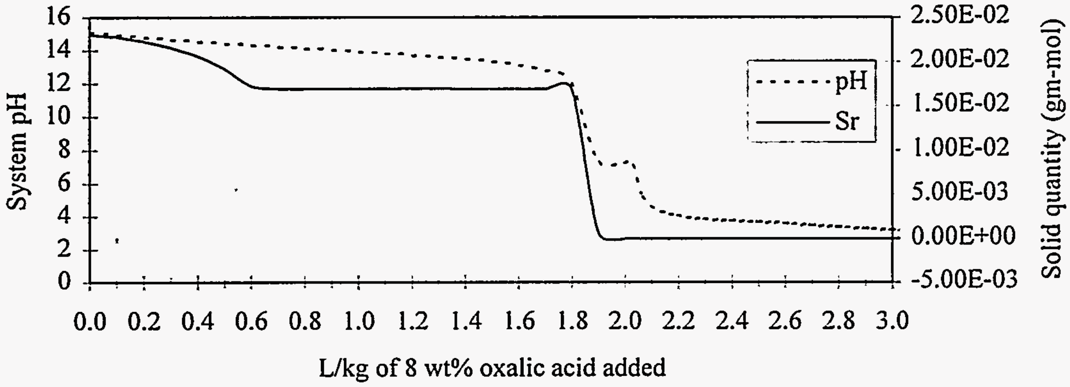

Figure O.11(d). Strontium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

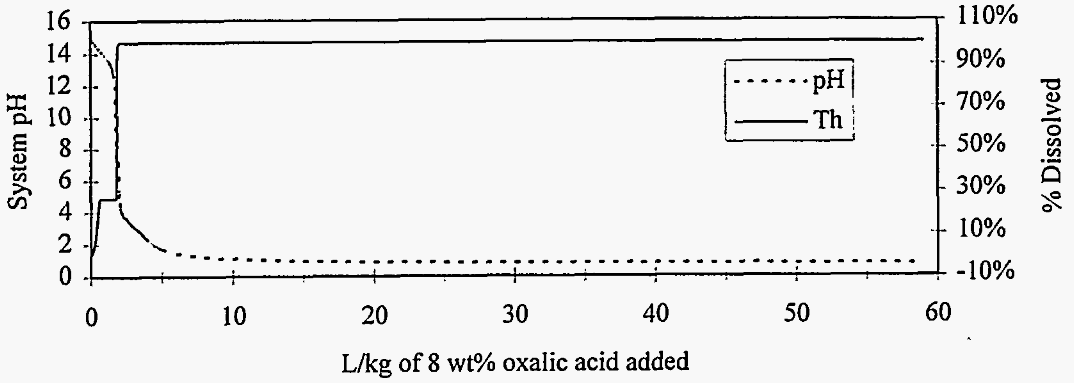

Figure O.12(a). Percent of Modeled Sludge Th Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

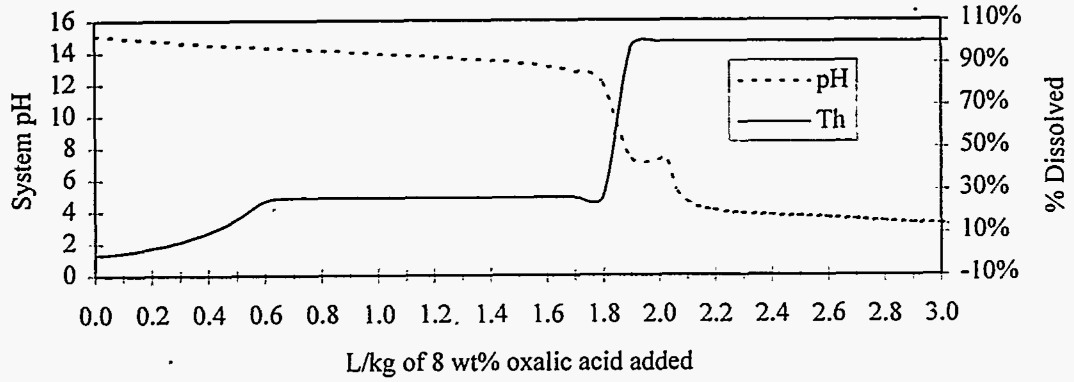

Figure O.12(b). Percent of Modeled Sludge Th Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

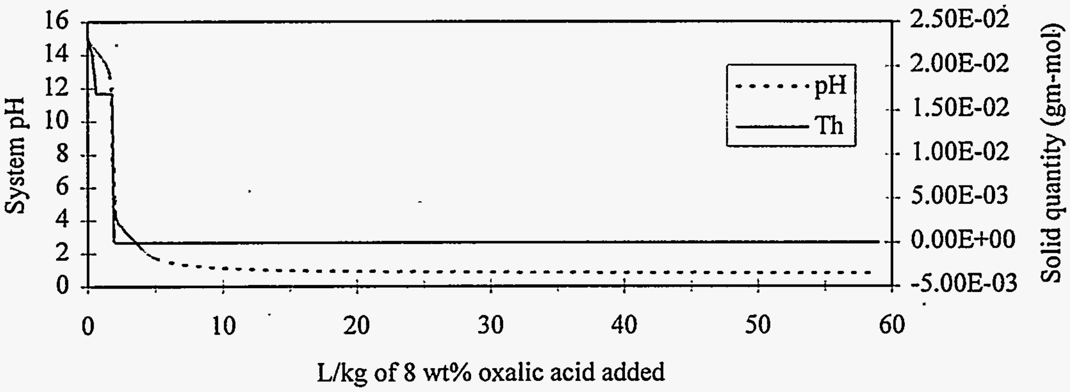

Figure O.12(c). Thorium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

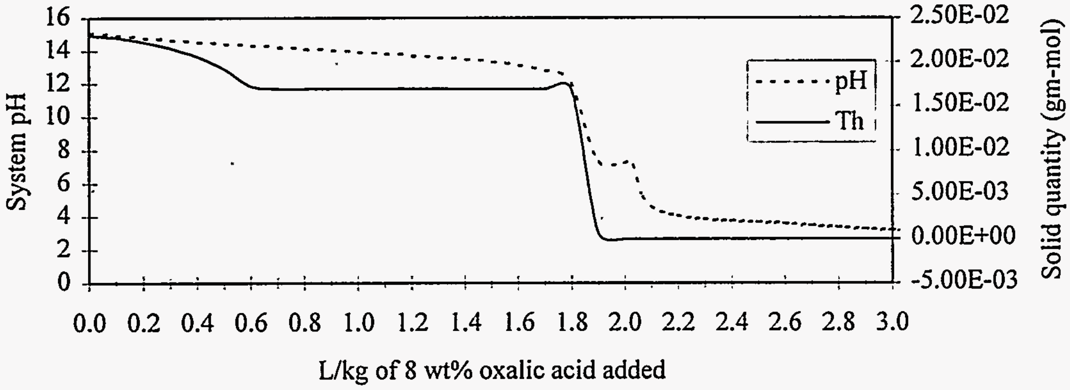

Figure O.12(d). Thorium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

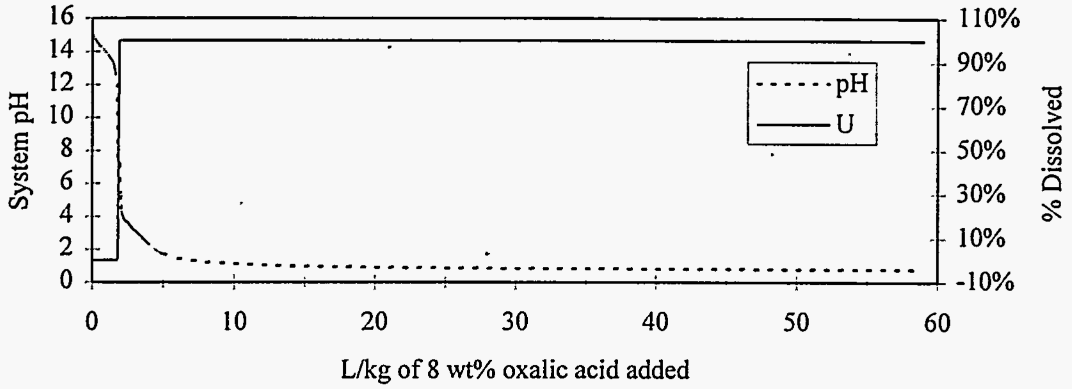

Figure O.13(a). Percent of Modeled Sludge U Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

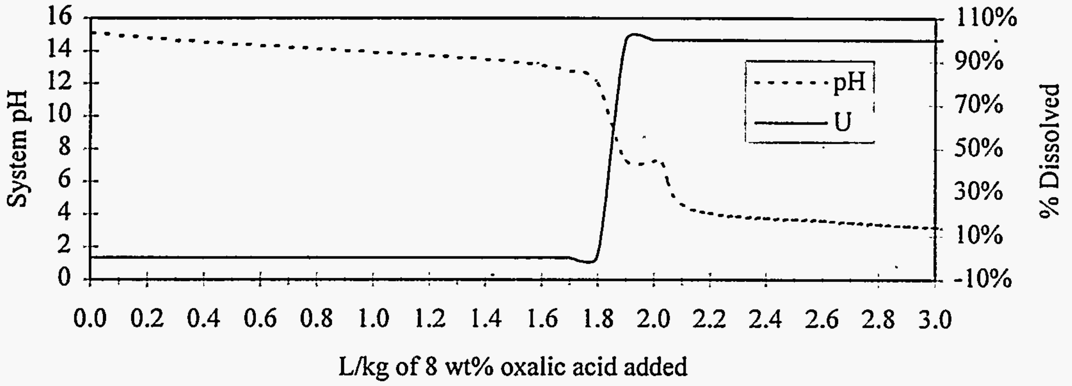

Figure O.13(b). Percent of Modeled Sludge U Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

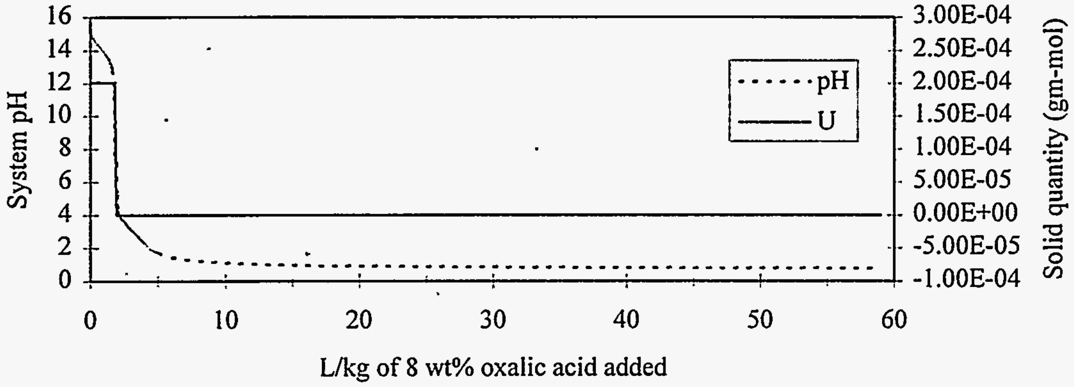

Figure O.13(c). Uranium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

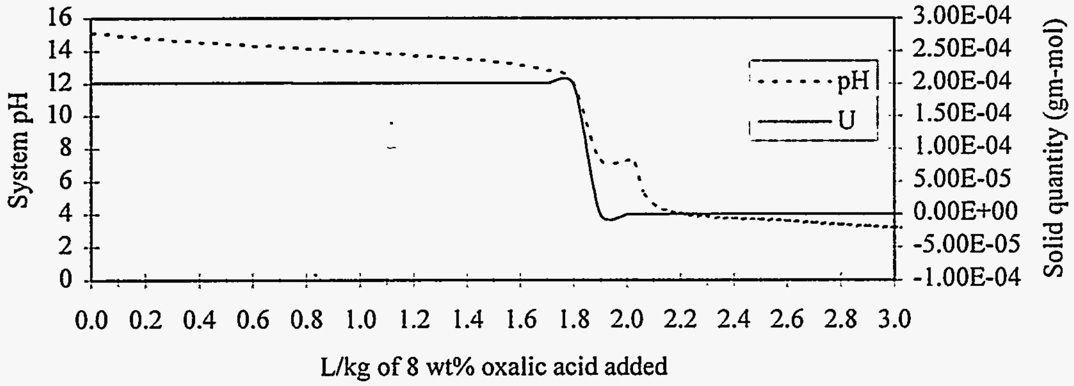

Figure O.13(d). Uranium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

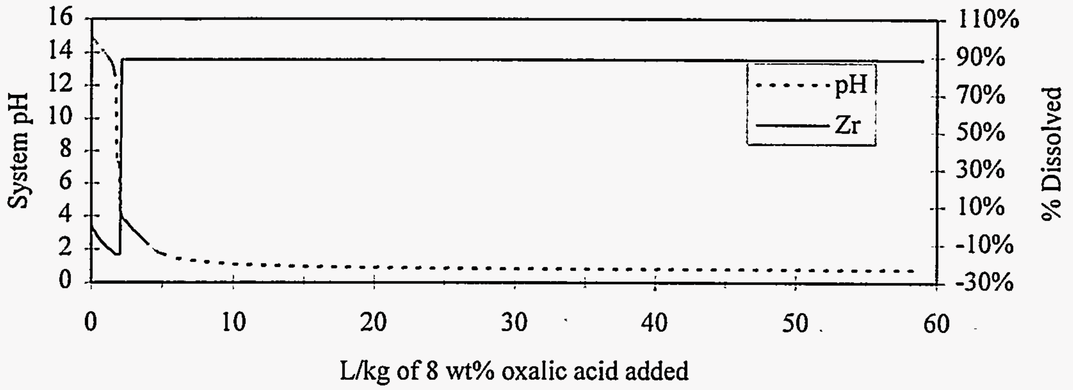

Figure O.14(a). Percent of Modeled Sludge Zr Dissolved at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

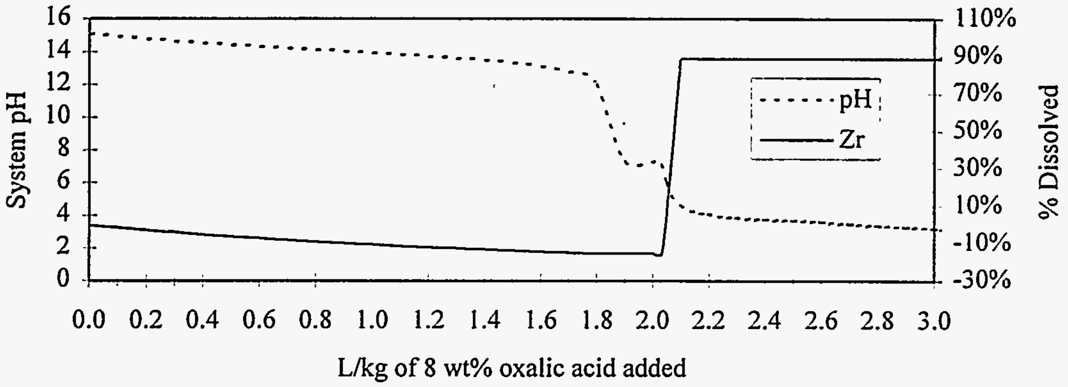

Figure O.14(b). Percent of Modeled Sludge Zr Dissolved at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

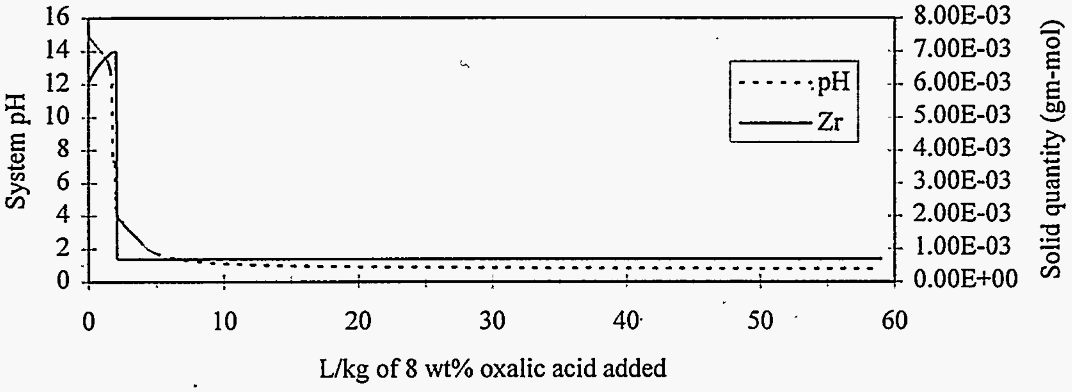

Figure O.14(c). Zirconium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

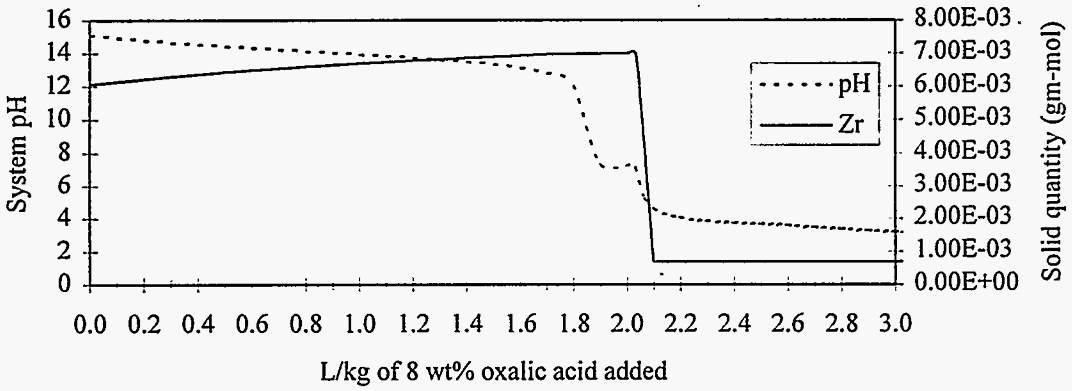

Figure O.14(d). Zirconium Remaining in Modeled Sludge Solids at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

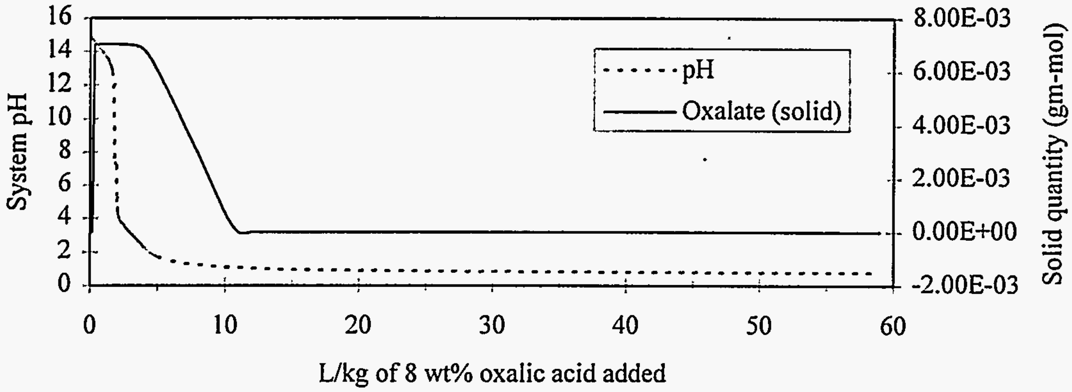

Figure O.15(a). Solid Oxalate Remaining in Modeled Sludge at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

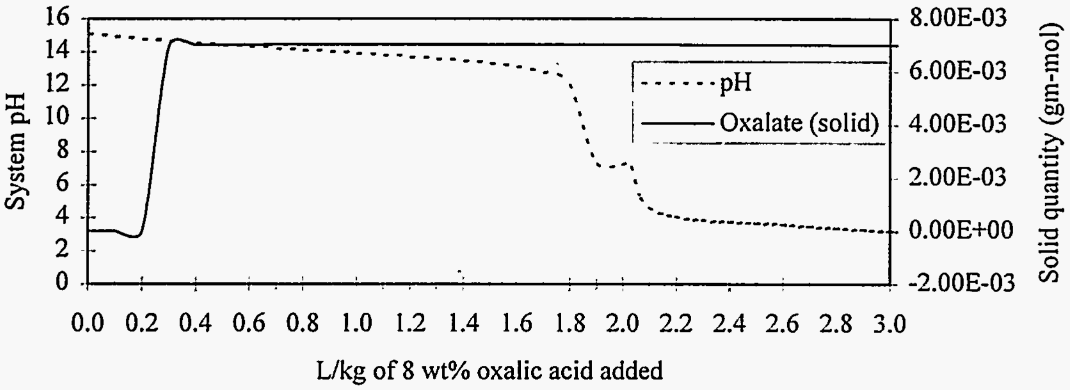

Figure O.15(b). Solid Oxalate Remaining in Modeled Sludge at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

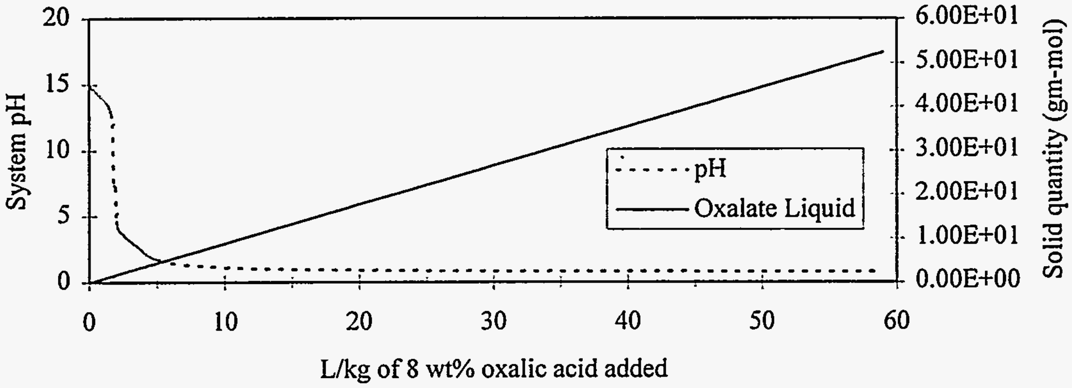

Figure O.16(a). Dissolved Oxalate Remaining in Modeled Sludge at Thermodynamic Equilibrium for 0 - 60 L 8 wt% Oxalic Acid/kg Sludge

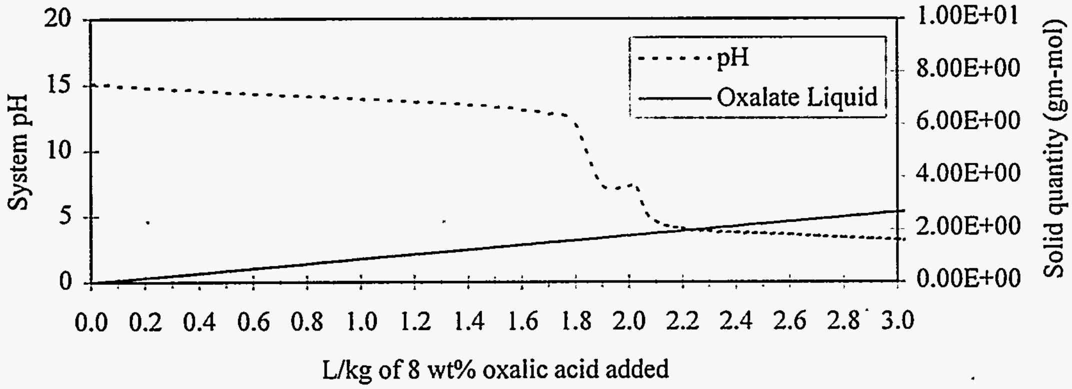

Figure O.16(b). Dissolved Oxalate Remaining in Modeled Sludge at Thermodynamic Equilibrium for 0 - 3 L 8 wt% Oxalic Acid/kg Sludge

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