PNNL-11274
UC-2000

M96050422

RECEIVED
SEP 20 1996
OSTI

Fiscal Year 1996 Laboratory Scale Studies of Cesium Elution in Tank 8D-1

R. L. Russell
G. K. Patello
J. A. Sills

September 1996

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

Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle

Battelle

PNNL-11274

MASTER

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831;
prices available from (615) 576-8401.

Available to the public from the National Technical Information Service,
U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161

PNNL-11274
UC-2000

Fiscal Year 1996 Laboratory Scale Studies of Cesium Elution in Tank 8D-1

R. L. Russell
G. K. Patello
J. A. Sills

September 1996

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

Pacific Northwest National Laboratory
Richland, Washington 99352

QA Level

This work was performed to satisfy WVSP FY 1996 milestone number 0604A and was conducted in accordance with Impact Level II quality assurance requirements as defined by the WVSP Quality Assurance Plan ETD-007 and the PNNL *Quality Assurance Manual* PNNL-MA-70.

Summary

During the vitrification phase of the West Valley Demonstration Project (WVDP) waste remediation, an estimated 95% of the zeolite in tank 8D-1 will be transferred to tank 8D-2, leaving behind residual cesium-loaded zeolite, which will require treatment to remove the cesium.

Oxalic acid has been proposed as a reagent for the elution of cesium from zeolite in tank 8D-1. During FY 1995, nonradioactive laboratory-scale tests were performed to determine optimum conditions for cesium elution in terms of acid-to-zeolite 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. During FY 1996, follow-up studies included examining the effects of reduced amounts of corrosion products (Fe_2O_3), the presence of nitrate and nitrite salts, smaller particle size zeolite, and neutralization by NaOH on cesium elution.

Although the amounts of cesium eluted after 28 hours were essentially the same in all four tests with different amounts of iron, adding larger amounts of Fe_2O_3 tended to slow the overall cesium elution process at the shorter contact times (< 20 hours). Furthermore, 34-39 wt% of the iron added to the system had dissolved after 28 hours of contact, regardless of the absolute amount of Fe_2O_3 present.

Within the bounds of experimental error (± 10%), the presence of nitrate and nitrite salts appears to have no effect on cesium elution. However, at 20 and 28 hours contact, the amount of dissolved iron was significantly lower in the presence of salts than in the control (21 wt% vs. 34 wt%).

Cesium elution levels in the test with the smaller zeolite particles were higher than the control at short contact times; however, after 20 hours of contact, the amounts of cesium eluted in the test and the control were within experimental error.

Neutralizing with NaOH after 20 hours to pH 5 or 12 made little difference in the amount of cesium eluted. However, iron precipitated when the solution was neutralized to pH 12.

Comparison of percentages of aluminum and cesium in solution showed the same trend for each test indicating that there is a correlation between zeolite dissolution and cesium release.

FY 1995 and FY 1996 studies showed that oxalic acid is an effective agent for eluting cesium from zeolite. An extraction efficiency of ~85 wt% in a single contact was achieved. In addition, acid neutralization after cesium elution can limit corrosion of the tank and dissolution of accumulated corrosion products. However, acid neutralization will add additional sodium to the melter feed which may create a concern for West Valley.

Acronyms

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

Contents

Figures

Tables

1.0 Introduction

This report details work performed as part of West Valley Support Project (WVSP) Fiscal Year 1996 Subtask 0604, conducted by the Pacific Northwest National Laboratory (PNNL). This work is intended to support residual waste removal during high-level waste (HLW) tank stabilization activities to be performed by the West Valley Demonstration Project (WVDP). The HLW, approximately 2 million liters, originated from a now inactive commercial nuclear fuel-reprocessing plant at West Valley, New York. It is stored in a carbon-steel tank designated 8D-2. Cesium-loaded zeolite was generated by a supernatant decontamination process involving ion exchange. The exchange columns and the spent zeolite are stored in a carbon-steel tank designated 8D-1 (Cadoff 1991). During the vitrification phase of the WVDP waste remediation, an estimated 95 wt% of the zeolite will be transferred from tank 8D-1 to tank 8D-2. The remaining cesium-loaded zeolite will require treatment to remove the highly radioactive cesium(a).

Oxalic acid has been studied as a decontamination reagent for nuclear reactors and equipment (Meservey 1970). In laboratory studies(b) oxalic acid [0.8M (7 wt%) oxalic acid at ~20 L acid/kg zeolite] eluted cesium from zeolite at a level of 88-90% over 11 days (264 hours).

During FY 1995, studies performed on the use of oxalic acid to elute cesium from zeolite showed that, although no one set of conditions tested could be described as "optimum", 40 L of 8 wt% oxalic acid/ kg zeolite at 50°C in a series of three contacts eluted 99 wt% of the cesium. Most of the cesium elution occurred during the first few hours of contact with oxalic acid; longer times did not significantly increase elution. Addition of Fe_2O_3 to the cesium elution tests to simulate corrosion products present on the bottom of tank 8D-1 did not significantly affect cesium elution. However, ~90 wt% of the iron added to the system dissolved; therefore, as shown in FY 1995, the rust in the tank may be soluble in 8 wt% oxalic acid. Iron oxide dissolution in tank 8D-1 would cause more iron to enter the vitrification process and may increase the number of glass logs generated. Use of lower concentrations of oxalic acid (4 and 1 wt%) decreased iron solubilization to 15-20 wt% of the iron in the system, but cesium elution efficiency also dropped to ~75 wt% with three contacts.

This report summarizes the findings of laboratory experiments conducted at PNNL during FY 1996. The primary objectives of the FY 1996 experiments were as follows:

- to further refine the optimum process conditions identified during the FY 1995 work for the use of oxalic acid to elute cesium from zeolite in tank 8D-1; target value, 99 wt% cesium eluted,
- to minimize iron dissolution during cesium elution,
- to investigate the effect of neutralization on cesium elution,

---

(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.

- to determine the effect of zeolite particle size on cesium elution,
- to determine if aluminum can be used as an indicator for cesium in solution.

These objectives were addressed in 11 tests in which the amount of iron was varied, zeolite particle size was varied, nitrate and nitrite salts were added, and the oxalic acid solution was neutralized. The contact solution from each test was assayed for aluminum, cesium, and iron using ICP-MS. The amount of iron added to the system was decreased in one test to provide a lower bounding case for the amount of iron dissolved assuming that there is less rust present in tank 8D-1 than had been previously estimated. The zeolite particle size was decreased in one test to determine if grinding the zeolite in tank 8D-1 would allow more efficient cesium elution. Nitrate and nitrite salts were added in one test to simulate salts present as interstitial liquid in tank 8D-1 and to determine their affect on cesium elution. The oxalic acid solution was neutralized to simulate quenching of the acid in tank 8D-1 to minimize tank corrosion and to determine the affect on cesium elution.

2.0 Conclusions and Recommendations

2.1 Conclusions

Eleven cesium elution tests were run in three batches. The tests conditions were: (1) control (recommended conditions from FY 1995 testing); (2) a lower concentration of Fe_2O_3, (3) concentrations of $NaNO_2$ and $NaNO_3$ equivalent to those in tank 8D-1, (4) Fe_2O_3 alone (no zeolite), (5) Al_2O_3 alone (no zeolite), (6 and 7) the oxalic acid neutralized with NaOH after 20 hours of contact, (8) zeolite particles ground to < 200 mesh, (9) zeolite alone, (10) a carbon-steel coupon alone, and (11) cesium loading ~ 3 times higher than in previous testing.

Larger quantities of Fe_2O_3 tend to slow the cesium elution process at short contact times (< 20 hours). By the end of the tests (28 hours contact), however, the amounts of cesium eluted were all ~ 90 wt% and within experimental error of one another, regardless of the initial amount of Fe_2O_3 present. After 28 hours, the amounts of iron in solution were within experimental error of each other at 34-39 wt%. However, longer contact times may place larger amounts of iron in solution, as evidenced by the FY 1995 tests, in which 90 wt% of the added iron dissolved after 50 hours contact.

In tests with only zeolite and only carbon-steel coupons much less iron was in solution after 28 hours of contact with oxalic acid. However, in the zeolite only test, after 4 hours of contact, the amount of iron in solution was comparable to the control test. This suggests that iron dissolves at a certain rate regardless of the absolute amount present.

The presence of $NaNO_2$ and $NaNO_3$ in amounts equivalent to the concentration of these compounds in tank 8D-1 had no effect on cesium elution. However, the presence of the salts did decrease the amount of iron in solution by about 40%. Therefore, the amount of iron that would dissolve in tank 8D-1 may not be as high as FY 1995 testing indicated. Tests with higher nitrite and nitrate levels and longer contact times are recommended.

Grinding the zeolite had no significant effect on the overall cesium elution, although elutions were slightly higher at short contact times because a larger surface area of zeolite was in contact with oxalic acid.

When zeolite was loaded with three times more cesium than the control, the percentage of cesium eluted was not significantly different from the control after 20 hours of contact (85 wt% in control vs. 82 wt% in high cesium loading). This shows that all of the data from the lower cesium loaded zeolite tests are still valid.

Neutralizing the oxalic acid with sodium hydroxide after 20 hours had little effect on the amount of cesium eluted, but neutralizing to pH 12.5 decreased the amount of iron in solution. Therefore, neutralization may limit the amount of iron transferred to tank 8D-2 in the supernatant while having minimal impact on cesium elution.

The mechanism for cesium elution may be a process by which aluminum and iron within the zeolite structure dissolve, creating a larger space around the cesium atoms. Aluminum is one of the principal components of zeolite IE-96; therefore, the amount of aluminum in solution might indicate the amount of zeolite dissolved and hence of the level of cesium elution. Comparison of percentages of aluminum and cesium showed the same trend in each test, indicating a correlation. At short contact times, the ratio of moles aluminum to moles cesium is very high. However, as contact time increases, the ratio decreases quickly to a constant value. Thus it may be that initially several aluminum atoms must be eluted from the structure of the zeolite to allow the cesium to escape, but as more aluminum is eluted, cesium escapes from the zeolite at a faster rate.

The results of the current testing, show that cesium can be successfully eluted from zeolite with oxalic acid. The objectives of this task were met by showing that adding salts will minimize the iron dissolution, neutralization with NaOH has no effect on the cesium elution but significantly decreases the amount of iron in solution and reducing zeolite particle size has no effect on the total cesium eluted from the zeolite.

2.2 Recommendations

The findings of this study lead to the following recommendations:

- that West Valley use 8 wt% oxalic acid at 40 L/ kg zeolite to elute the cesium from the remaining zeolite in tank 8D-1.
- a neutralization step with NaOH after 20 hours of acid contact to control dissolution of corrosion products in the tank, however, such neutralization produces a significant amount of precipitate (primarily sodium oxalate).
- three acid contacts be used. With only one acid contact, only ~88 wt% of the cesium can be removed; with three contacts 99 wt% of the cesium can be eluted as shown by the FY 1995 tests.
- several more tests be performed to further refine this process and to gain a thorough understanding of it, including: longer contact times with added salts and higher nitrite and nitrate levels.

3.0 Experimental Procedures

3.1 Zeolite Loading

Zeolite IE-96 (see Table 3.1 for composition), manufactured by UOP[(a)], was loaded to the desired level of cesium (922 µg Cs/g zeolite) by contacting it with $CsNO_3$ solution for 24-hours. The vessel in which the zeolite loading took place was kept at room temperature, and the solution and zeolite were agitated during the entire period to provide maximum contact of the zeolite with the $CsNO_3$ solution. Appendix A contains the calculations of the required levels of cesium loading and the contact conditions for each set. After contact, the solution was decanted and the zeolite was placed in a drying oven at ~105°C for 24 hours. A sample of the zeolite was analyzed by X-ray fluorescence (XRF) for cesium content. In the West Valley tank waste radioactive cesium is 30% of the total cesium on the zeolite. In one test, zeolite was loaded to a level that represented the total cesium loaded on the zeolite. The results for each batch are shown in Table 3.2.

When exposed to air, zeolite absorbs some of the moisture in the air, and for this reason, the loaded zeolite was stored in a drying oven at 110°C. Weights of zeolite referred to in this report are dry weights, and thus no F-factor (dry weight/wet weight) calculation is necessary.

Table 3.1. Analysis of Zeolite IE-96

Compound	wt%
Al_2O_3	15.5
BaO	0.09
CaO	1.1
Fe_2O_3	3.5
MgO	0.7
Na_2O	7.0
MoO_3	0.03
SiO_2	60.1
SrO	0.04
TiO_2	0.03
ZrO_2	0.04
Other	0.87
H_2O	11.0
Total	100

(a) UOP, Tarrytown, NY.

Table 3.2. X-Ray Fluorescence Results for Cesium-Loaded Zeolite

µg Cs/g zeolite	relevant tests
922	30, 31, 32, 35, 37, 40
995	36 (reduced particle size zeolite)
3615	39 (high Cs-loaded zeolite)

3.2 Cesium-Elution Experiments: General Procedure

The objective of this set of cesium elution tests was to refine the optimum process conditions by evaluating several variables. The baseline data were results of the two previous rounds of testing (Sills, et al. 1996) and additional engineering information received from West Valley Nuclear Services. Eight wt% oxalic acid was used in all of these tests. WVNS calculated that the 100 L acid/kg zeolite used in the second round of FY 1995 testing would generate too much waste, and that 20 L/kg would provide too little liquid to cover the zeolite in the tank; 40 to 60 L acid/kg zeolite is a more realistic range for the acid-to-zeolite ratio(a). Therefore, the previously defined optimum process ratio of 40 L acid/kg zeolite was used in these tests. In the FY 1995 testing, 2.77 g of 0.001M NaOH was required to wet the 5 g of zeolite used in these tests (which is equivalent to 0.55 L 0.001M NaOH/kg zeolite). WVNS indicated that a more realistic approach would be to add a volume equivalent to 4 L 0.045 M NaOH/ kg zeolite(b). The FY 1996 tests were scaled up to 10 g zeolite to help reduce the error involved by working with larger quantities. The bath temperature was 50°C and the acid addition temperature was 25°C (room temperature). FY 1995 results showed that most of the cesium elution occurs in the first few hours of contact; therefore, the contact length was shortened to 28 hours, and more sampling events were scheduled at shorter contact times. In previous tests, most of the cesium was eluted during the first acid contact; therefore, these tests were run with only a single acid contact. The test matrix is shown in Table 3.2.

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 4 L of 0.045 M NaOH/kg zeolite (to simulate the basic environment in tank 8D-1). The 8 wt% oxalic acid was then added at room temperature (22 to 25°C). The vessel was sealed with vacuum grease and placed in a heating mantle with a temperature controller programmed to hold at 50°C (the expected temperature of the material in tank 8D-1(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 mixing pumps in tank 8D-1. Tests were run for 28 hours, and sampling was performed at 15 minutes, 30 minutes, 1, 2, 4, 20, and 28 hours of contact time, unless otherwise noted. During sampling, agitation was stopped and the material in the vessel was allowed to settle for five minutes. A 5 mL sample was then drawn from the supernatant and analyzed by inductively coupled plasma mass spectroscopy (ICP-MS) for cesium, iron, and aluminum content.

(a) King, CS, fax to GK Patello dated January 29, 1996.

(b) Schiffhauer, MA, personal communication.

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

Table 3.3. Test Matrix for FY96 Cesium Elution Tests

	Test	Zeolite, g	NaOH Heel, g	Oxalic Acid, g	Fe_2O_3, g
30	control	10	40	407.6	4.00
31	low Fe_2O_3	10	40	407.6	2.00
32*	added salts	10	40	407.6	4.00
33	pure Fe_2O_3	0	0	1630.4	16.00
34**	pure Al_2O_3	0	0	1630.4	0.00
35	neutralization	10	40	407.6	4.00
36	small particles	10	40	407.6	4.00
37	zeolite alone	10	40	407.6	0.00
38	coupon alone	0	40	407.6	0.00
39	high Cs	10	40	407.6	4.00
40	neutralization	10	40	407.6	4.00

* Test 32 contained 0.36 g $NaNO_2$ and 0.11 g $NaNO_3$

** Test 34 contained 16.00 g Al_2O_3

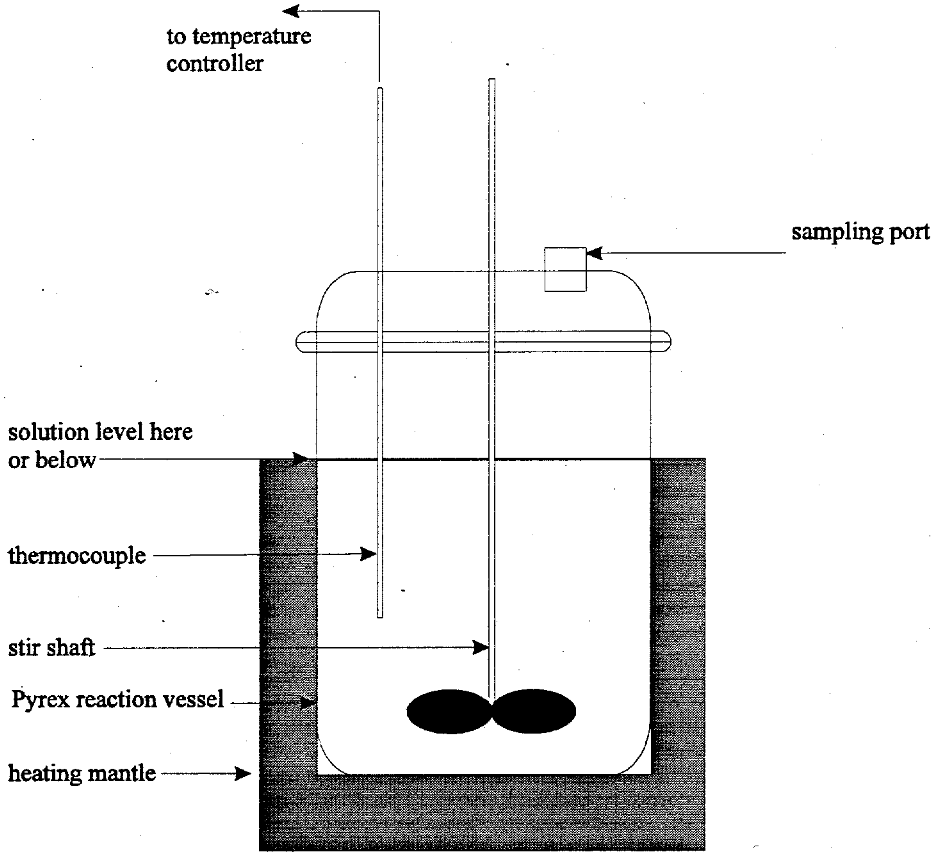

Figure 3.1. Reaction Vessel Assembly Used in Cesium Elution Tests

Control

Test 30 was the control, setting the "standard conditions" for the FY1996 tests. Unlike the FY1995 tests, this control contained 4 g of Fe_2O_3 as a standard condition. The control was run at 40 L of 8 wt% oxalic acid/kg zeolite, and the tests were based on 10 g of zeolite. Final solids were collected for x-ray diffraction (XRD) analysis. In addition, a sample of uncontacted zeolite and a sample of zeolite that had been contacted with oxalic acid for 28 hours with no Fe_2O_3 present were submitted for comparison.

Reduced Fe_2O_3

Test 31 contained half the amount of Fe_2O_3 added to the control to provide a lower bound case for evaluation. All previous tests have examined a "worst case" scenario. In all other respects, this test duplicated the control.

Added Salts

Test 32 was designed to examine the effect of $NaNO_2$ and $NaNO_3$ on cesium elution and iron dissolution. The salts were added to the reaction vessel in amounts that would bring the total NO_2^- and NO_3^- concentrations to the same level as those found in tank 8D-1: 6000 µg NO_2^-/g of solution, and 2000 µg NO_3^-/g of solution[a].

Pure Compounds

Tests 33 and 34 were designed to provide a baseline by examining the dissolution behavior of the pure compounds Fe_2O_3 and Al_2O_3, respectively, in 8 wt% oxalic acid. By comparing these results with the results of the combined reactants, it could be determined if the mixture has an effect on the individual rates of dissolution. These tests were run in two-liter reaction vessels, and amounts of reagents were based on 40 g of zeolite, although no zeolite was actually added to either vessel. Test 33 contained 16 g of Fe_2O_3, and Test 34 contained 16 g of Al_2O_3.

Neutralization

Tests 35 and 40 examined the effect of neutralizing the oxalic acid with NaOH on iron dissolution and cesium elution. The initial test conditions were identical to the control, but after 20 hours of contact with 8 wt% oxalic acid, 10 M NaOH was added to the reaction vessel to raise the pH to ~12 for Test 35, and ~5 for Test 40. Samples were taken at 15 minutes, 30 minutes, 1 hour, 2 hours, and 4 hours after pH stabilization. At the end of the experiments, the remaining solids were collected, weighed, and submitted for XRD analysis.

Reduced Particle Size Zeolite

Test 36 examined the effect of using smaller particles of zeolite. The zeolite was ground in the laboratory, sieved to -200 mesh, and analyzed by XRF to confirm that the cesium loading had not

---

[a] King, CS, fax to GK Patello dated December 8, 1995.

changed significantly. In all other respects, the conditions for this test were identical to the control. After the last sampling event, the remaining solids and a sample of the initial ground zeolite were submitted for particle size analysis.

Zeolite and Coupons

The dissolution of zeolite alone in 8 wt% oxalic acid was examined in Test 37, and the dissolution of a carbon steel coupon in 8 wt% oxalic acid was examined in Test 38. In Test 37, the only source of iron was the zeolite, and in Test 38, the only source of iron was the coupon. These tests were conducted to determine if iron from different sources (i.e., zeolite, carbon steel, and Fe_2O_3) had different rates of dissolution.

High Cesium Loading

Test 39 examined the effect of loading zeolite to about three times the cesium level used in previous experiments. This cesium loading corresponds more closely to the loading of the zeolite in tank 8D-1. In all other respects, the conditions for this test were identical to the control.

3.3 Particle Size Analysis

Particle size analysis with a Microtrac FRA particle size analyzer was performed on the ground zeolite both before and after oxalic acid contact. A representative sample of the zeolite was diluted (~10 times) with deionized water and injected into the sample chamber of the analyzer. This technique was not used on the unground zeolite because the particle size (~200-500 µm) was outside the instrument's capability.

Before and after oxalic acid contact, samples of ground and unground zeolite were sprinkled on a glass slide and examined with an Olympus PMG3 optical microscope with magnification in the range of 10X to 100X. This technique was used in a qualitative manner only to observe changes to the zeolite structure.

4.0 Results and Discussion

4.1 Effect of Amounts and Types of Iron

The amount of cesium eluted in Tests 30 (control), 31 (reduced Fe_2O_3), and 37 (zeolite alone), which contained 4, 2, and 0 g Fe_2O_3, respectively, are shown in Table 4.1, and the data are plotted in Figure 4.1. Test 17 from FY 1995 which also contained 4 g Fe_2O_3 but only 5 g of zeolite is also included for comparison. All values in the tables throughout the report have an estimated ±10% uncertainty due to analytical error. Temperature and pH data for all of the tests are in Appendix C. In Appendix D sample calculations show the method by which raw data were converted to wt% cesium eluted. The data were corrected for sample removal; as shown in the calculations in Appendix D. The amount of cesium eluted after 28 hours contact in Tests 31 and 37 are within experimental error of the control (Figure 4.1). Although the amounts of cesium eluted after 28 hours are essentially the same in all four tests (~90 wt%), larger amounts of Fe_2O_3 slowed the overall cesium elution rate at the shorter contact times (< 20 hours) in the FY 1996 tests. These tests compare favorably with the control from the FY 1995 testing which showed that ~91 wt% of the cesium was eluted from the zeolite after 28 hours even though less zeolite and NaOH were used. However, using less NaOH seems to have increased the rate at which the cesium is eluted at short contact times.

Table 4.1. Weight Percent Cesium Eluted in Tests to Determine Effects of Iron on Cesium Elution

sample	hours	control (FY 1996) Test 30	< Fe Test 31	zeolite Test 37	control (FY 1995) Test 17
T1	0.25	4.45	3.51	1.38	--[(a)]
T2	0.5	9.02	12.5	9.23	--[(a)]
T3	1	21.2	26.9	34.4	--[(a)]
T4	2	38.1	54.4	80.2	68
T5	4	50.9	69.7	89.5	82
T6	20	84.7	86.9	89.5	100
T7	28	87.8	92.3	81.8	91

(a) Not measured.

The amount of iron dissolved in these tests and in Test 33, which contained pure Fe_2O_3, are shown in Table 4.2 and Figure 4.2. In the tests containing added Fe_2O_3 (-325 mesh), 34 to 39 wt% of the total iron in the system dissolved in 28 hours, regardless of the amount of Fe_2O_3 added to the system. In Test 37, which contained zeolite alone, 86 wt% of the iron from the zeolite structure was in solution after 28 hours, which supports FY 1995 findings that 91 wt% of the iron in the zeolite is in solution after 28 hours.

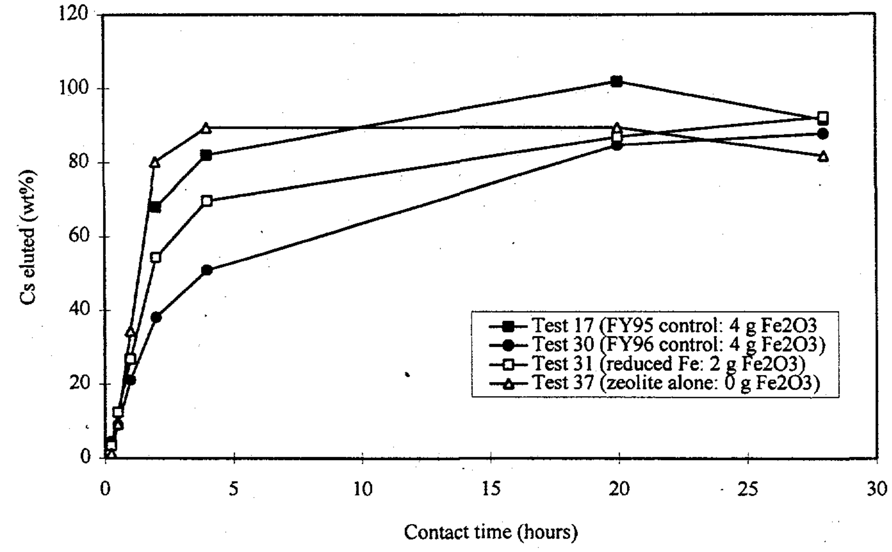

Figure 4.1. Percent Cesium Eluted in Tests Containing Various Amounts of Fe_2O_3 (100% = Total Amount of Cesium Loaded onto Zeolite)

Table 4.2. Percent Iron Dissolved in Tests Containing Iron in Various Amounts and Forms

		control (FY 1996)	< Fe	pure Fe_2O_3	zeolite	control (FY 1995)
	Total Fe	2.7977g	1.3989g	11.20g	0.2448g	2.8886g
sample	hours	Test 30	Test 31	Test 33	Test 37	Test 17
T1	0.25	1.22	2.97	1.56	1.17	--(a)
T2	0.5	1.01	2.99	0.94	11.0	--(a)
T3	1	1.88	4.03	2.63	28.7	--(a)
T4	2	3.31	7.32	4.65	49.9	2.8
T5	4	5.15	9.65	4.80	55.7	6.2
T6	20	24.4	27.0	25.0	104	5.1
T7	28	33.7	38.6	38.5	85.5	63

(a) Not measured.

The FY 1996 results are also compared to those of FY 1995 in Figure 4.2. In Test 17, conducted in FY 1995, 63 wt% of the iron had dissolved after 28 hours of contact. It is most likely that either this point or the 20 hour contact point is aberrant because the FY 1996 results show a maximum iron dissolution of 39 wt% for similar tests. Longer contact times under the conditions examined in the FY 1996 tests may result in more dissolution of iron because the iron concentrations are still increasing slightly after 28 hours.

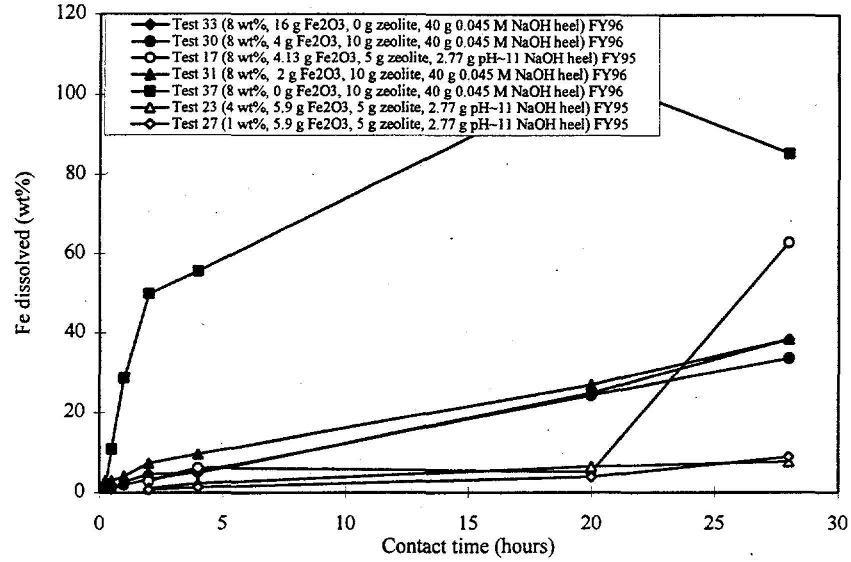

Figure 4.2. Percent Iron Dissolved in Tests Containing Iron in Various Forms and Amounts (100% = Total Amount of Iron in the System)

In comparing the results of FY 1995 and FY 1996 testing, it is important to note some basic differences in the two sets of tests. The NaOH heel used in FY1996 tests was much larger--40 g of 0.045 M NaOH, as opposed to 2.77 g of 0.001M NaOH used in the FY 1995 tests. Also, FY 1995 tests were scaled to 5 g zeolite; FY 1996 tests, to 10 g. Therefore, a total of 7.2 mg NaOH/g zeolite in the FY 1996 tests must be compared to 0.02 mg NaOH/g zeolite in the FY 1995 tests (see Appendix A for calculations). Later results in Section 4.2 show that added salts decrease iron dissolution. Adding NaOH to the system may have a similar effect and thereby explain the finding that a lower percent iron was dissolved at 28 hours in the FY 1996 tests than in the FY 1995 tests.

Additionally, FY1995 Tests 23 and 27 contained more iron per gram of zeolite than the FY 1996 tests, but Test 23 was run in 4 wt% oxalic acid, and Test 27 was run in 1 wt% oxalic acid. These tests show that increasing the concentration of oxalic acid from 1 to 8 wt% increases iron dissolution.

The amount of iron in solution (µg iron/g solution) at each sampling event is shown in Table 4.3. The data are also plotted in Figure 4.3 to compare the amount of iron in solution from the coupons and from the other tests. In Tests 37 and 38, which contained only zeolite and only coupons, respectively, much less iron than in the control was in solution after 28 hours of contact. Thus, most of the iron in solution apparently comes from dissolved Fe_2O_3, which may be present as rust in the bottom of the tanks, and not from the zeolite. In the zeolite only test, the amount of iron in solution until 4 hours contact is close to the control level. This finding may indicate that iron dissolves at a certain rate regardless of the absolute amount present. The zeolite does not keep pace with the control at 20 and 28 hours contact because the iron in the zeolite is depleted (85.5 wt% of the iron was eluted after 28 hours in the zeolite only test). The close comparison between the pure Fe_2O_3 test and the control throughout the test further suggests that most of the dissolved iron comes from the added iron (simulated corrosion product) and not from the zeolite. This is expected because the amount of corrosion product far exceeds the amount of iron present in the zeolite.

Table 4.3. Concentrations of Iron in Solution (µg/g) in Tests Containing Iron in Various Forms

		control	< Fe	pure Fe_2O_3	zeolite	coupons
sample	hours	Test 30	Test 31	Test 33	Test 37	Test 38
T1	0.25	83.3	109	107	6.41	2.60
T2	0.5	68.5	110	64.4	60.7	6.93
T3	1	129	149	181	160	15.3
T4	2	230	274	322	280	26.4
T5	4	361	364	332	313	35.0
T6	20	1750	1040	1740	598	196
T7	28	2430	1500	2690	485	300

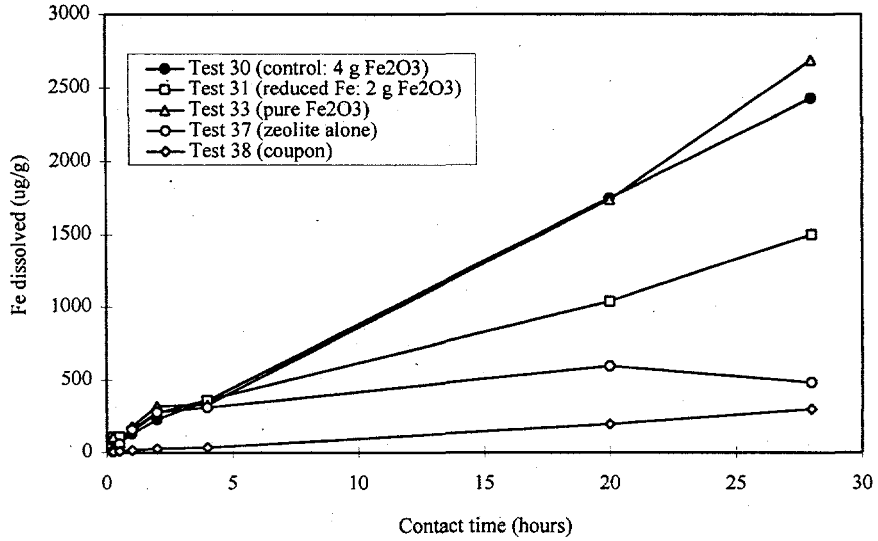

Figure 4.3. Concentrations of Iron in Solution (µg/g) in Tests Containing Iron in Various Amounts and Forms

4.2 Effect of the Presence of Salts

Test 32 contained $NaNO_2$ and $NaNO_3$ in amounts equivalent to the nitrite and nitrate levels found in tank 8D-1. The amount of cesium eluted in Tests 30 (control) and 32 (added salts) are shown in Table 4.4 and plotted in Figure 4.4. Within the bounds of experimental error (±10%), the presence of salts appears to have no effect on cesium elution.

Table 4.4. Weight Percent Cesium Eluted in Tests 30 and 32 to Determine the Effect of Salts on Cesium Elution

sample	hours	control Test 30	salts Test 32
T1	0.25	4.45	1.97
T2	0.5	9.02	9.33
T3	1	21.2	22.4
T4	2	38.1	40.9
T5	4	50.9	50.0
T6	20	84.7	78.7
T7	28	87.8	87.8

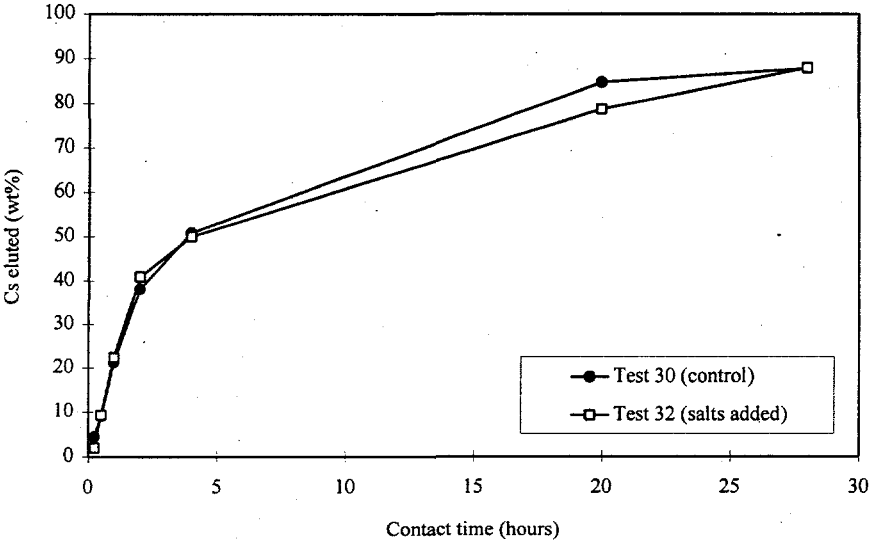

Figure 4.4. Weight Percent Cesium Eluted in Tests 30 and 32 to Determine the Effect of $NaNO_2$ and $NaNO_3$ Salts on Cesium Elution

The effect of salts on the dissolution of iron is shown in Table 4.5 and in Figure 4.5 by comparison to the control (Test 30). During the first four hours of contact, the presence of salts has no significant effect on the dissolution of iron; however, at 20 and 28 hours contact, significantly less iron was dissolved in the presence of salts (Test 32) than in the control (Test 30). After 28 hours contact, only 21 wt% of the iron had dissolved in the salts test, compared to 34 wt% in the control. Since nitrate and nitrite are present in tank 8D-1 in the concentrations used in Test 32, these data suggest that the amount of iron dissolved during cesium elution with oxalic acid may not be as high as FY 1995 testing indicated. This may have positive implications for rust dissolution. Not as much iron as previously thought may be going into the melter feed flowsheet. It is recommended to run these tests for longer times to determine whether the presence of salts would slow iron dissolution or halt the process after a certain time.

Table 4.5. Percentage of Iron Dissolved in Tests 30 and 32 to Determine the Effect of Salts on Cesium Elution

sample	hours	control Test 30	salts Test 32
T1	0.25	1.22	1.85
T2	0.5	1.01	1.17
T3	1	1.88	2.07
T4	2	3.31	2.98
T5	4	5.15	4.56
T6	20	24.4	12.5
T7	28	33.7	21.0

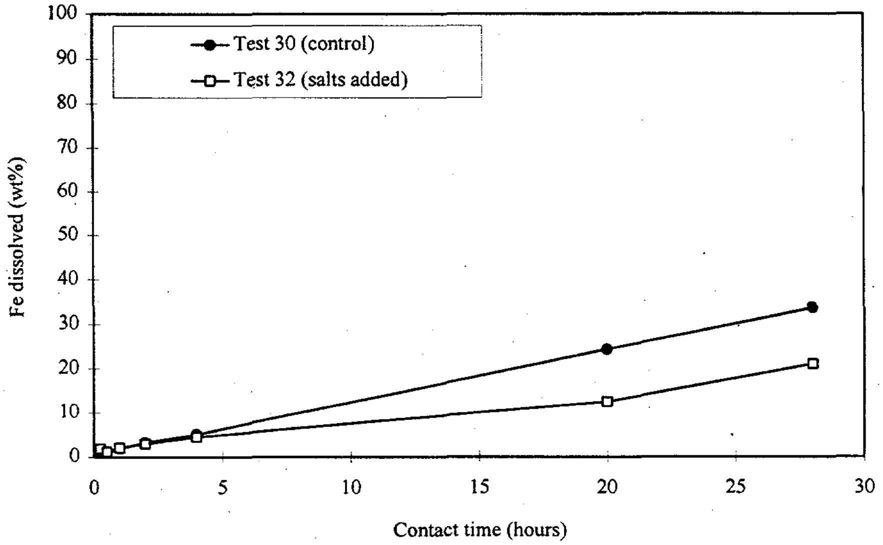

Figure 4.5. Percentage of Iron Dissolved in Tests 30 and 32 to Determine the Effect of $NaNO_2$ and $NaNO_3$ Salts on Cesium Elution

4.3 Effect of Reduced Particle Size Zeolite

Test 36 examined the effect of reducing the particle size of the loaded zeolite on cesium elution. The loaded zeolite (estimated particle size range of 240-420 µm), was ground and passed through a sieve so that only particles less than 75 um (-200 mesh) were used. The zeolite used in the West Valley columns was ~300 µm[a] . Table 4.6 and Figure 4.6 compare the cesium elution data from Test 36 with that of Test 30 (control). Higher cesium elution levels in the test with the ground zeolite at short contact times indicate that grinding the zeolite initially increases the rate of cesium elution. However, after 20 hours,

---

(a) Schiffhauer, MA, personal communication.

Table 4.6. Weight Percent Cesium Eluted in Test Containing Reduced Particle Size Zeolite

sample	hours	control Test 30	red. part. Test 36
T1	0.25	4.45	23.6
T2	0.5	9.02	34.0
T3	1	21.2	47.3
T4	2	38.1	55.4
T5	4	50.9	64.2
T6	20	84.7	80.7
T7	28	87.8	86.1

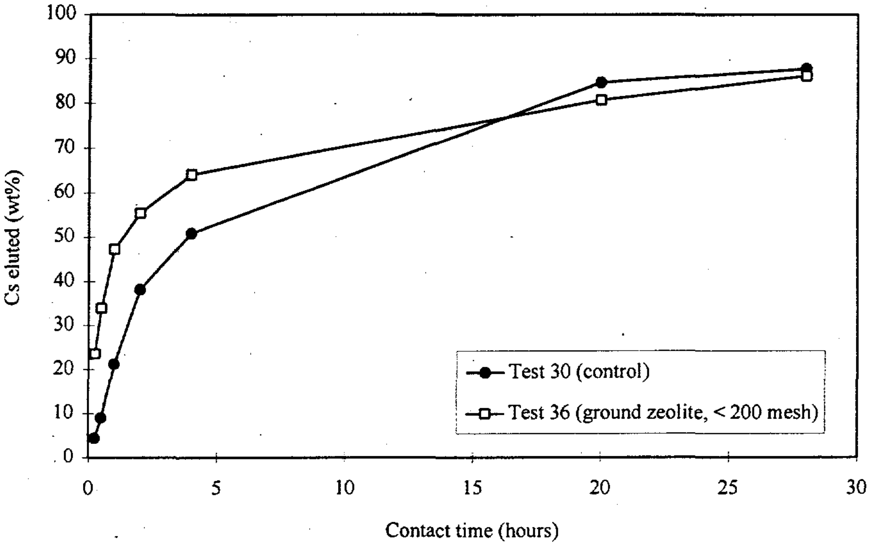

Figure 4.6. Weight Percent Cesium Eluted in Test Containing Reduced Particle Size Zeolite

the amount of cesium eluted in the reduced particle size test was within experimental error of the control. The greater cesium elution on the ground zeolite at shorter contact times is not surprising because a much greater surface area of zeolite is exposed to the oxalic acid solution. At longer times, however, an equilibrium is reached and therefore, the amount of cesium eluted is approximately the same.

Iron dissolution in Test 36 (reduced particle size zeolite) and Test 30 (control) is shown in Table 4.7 and plotted in Figure 4.7. The amount of iron in solution for the ground zeolite test is within experimental error of the control. Grinding the zeolite was not anticipated to affect the dissolution of iron because most of the iron is suspected to be coming from the Fe_2O_3 and not the zeolite.

Table 4.7. Effect of Reduced Particle Size Zeolite on Iron Dissolution During Cesium Elution Tests

sample	hours	control Test 30	red. part. Test 36
T1	0.25	1.22	1.53
T2	0.5	1.01	1.79
T3	1	1.88	3.19
T4	2	3.31	2.93
T5	4	5.15	4.42
T6	20	24.4	23.2
T7	28	33.7	31.8

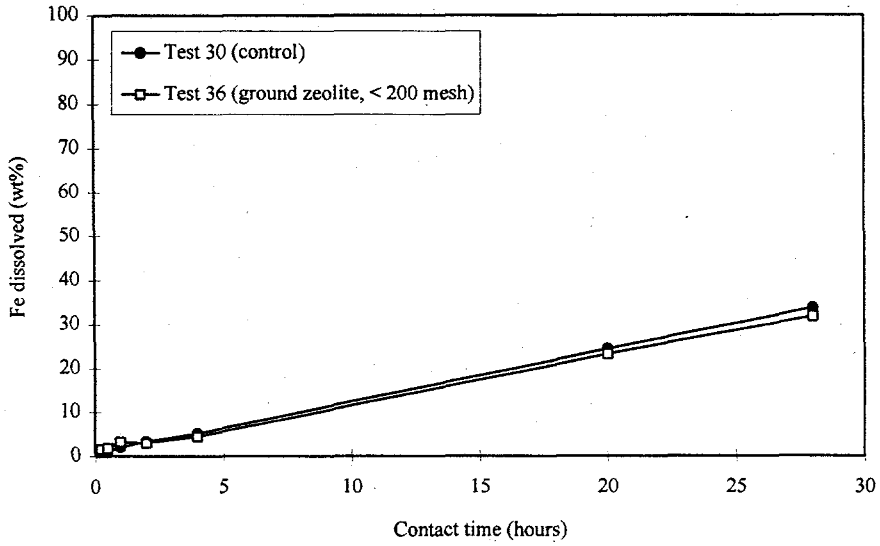

Figure 4.7. Effect of Reduced Particle Size Zeolite on Iron Dissolution During Cesium Elution Tests

4.4 Particle Size Analysis of the Zeolite

Particle size analysis was performed on the ground zeolite before and after contact with oxalic acid. The particle size distribution before and after oxalic acid contact based on an equivalent spherical diameter is plotted in Figures 4.8 and 4.9. The distribution curves before and after acid contact seem to be trimodal and bimodal, respectively. Before contact with oxalic acid, 10% of the particles are smaller than 2.80 μm, 50% are smaller than 22.33μm and 90% are smaller than 66.67 μm. After contact, 10% of the particles are smaller than 3.72 μm, 50% are smaller than 20.88μm and 90% are smaller than 57.01 μm. Thus the acid may dissolve some of the finer zeolite dust from grinding but it does not reduce

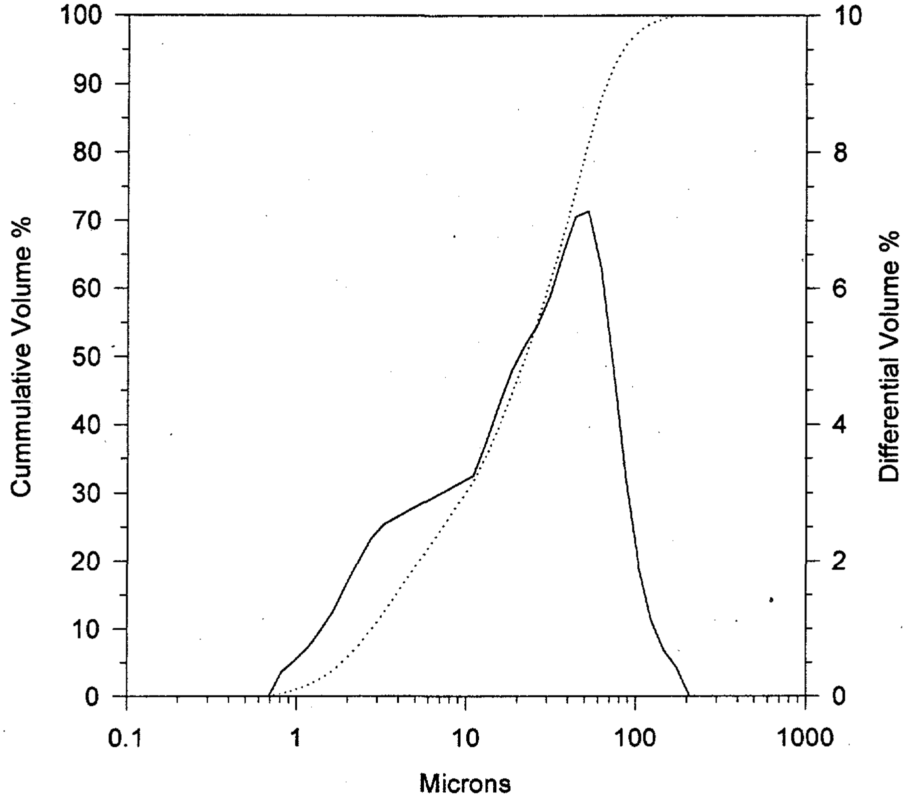

Figure 4.8. Zeolite Particle Size Before Acid Dissolution

4.5 Zeolite Loaded to a Higher Level of Cesium

Test 39 examined the effect on cesium elution of increasing cesium loading on the zeolite. The zeolite was loaded to 3615 µg cesium/g zeolite in Test 39 versus 922 µg cesium/g zeolite in most other experiments. The results of this test and the control (Test 30) are shown in Table 4.8 and Figure 4.10. This test became necessary after Lane Bray of PNNL revealed that only cesium-137 had been addressed in the cesium loading calculations and that it accounted for only ~30% of total cesium[(a)]. Therefore, the calculated cesium level was only about one-third of the actual cesium level in tank 8D-1. Test 39 was run to determine whether the cesium elution behavior was affected by increasing the cesium loading of the zeolite to the proper level. Figure 4.10 shows that the amount of cesium eluted in Test 39 is within experimental error of the control.

The amounts of iron dissolved in Test 39 and in Test 30 (control) in Table 4.9 and Figure 4.11. At longer contact times (20 and 28 hours), the amount of iron dissolved was slightly higher in the test than in the control. This result is unexpected as the amount of cesium in the zeolite should not affect the dissolution of loose Fe_2O_3, the source of most of the iron in solution. At shorter contact times, the amount of iron dissolved in Test 39 was within experimental error of the control.

(a) Bray, LA, personal communication.

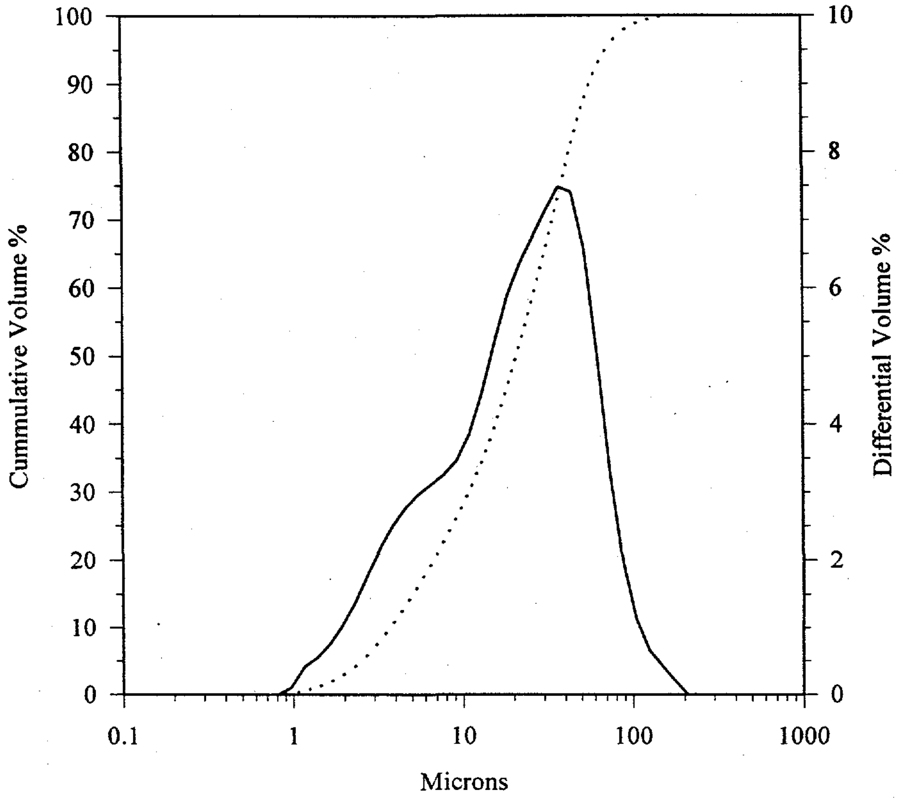

Figure 4.9. Zeolite Particle Size After Acid Dissolution

Table 4.8. Weight Percent Cesium Eluted in Test to Determine the Effect of Using Higher Cesium-Loaded Zeolite on Cesium Elution

sample	hours	control Test 30	high Cs Test 39
T1	0.25	4.45	1.73
T2	0.5	9.02	5.57
T3	1	21.2	13.5
T4	2	38.1	24.4
T5	4	50.9	47.2
T6	20	84.7	81.8
T7	28	87.8	78.5

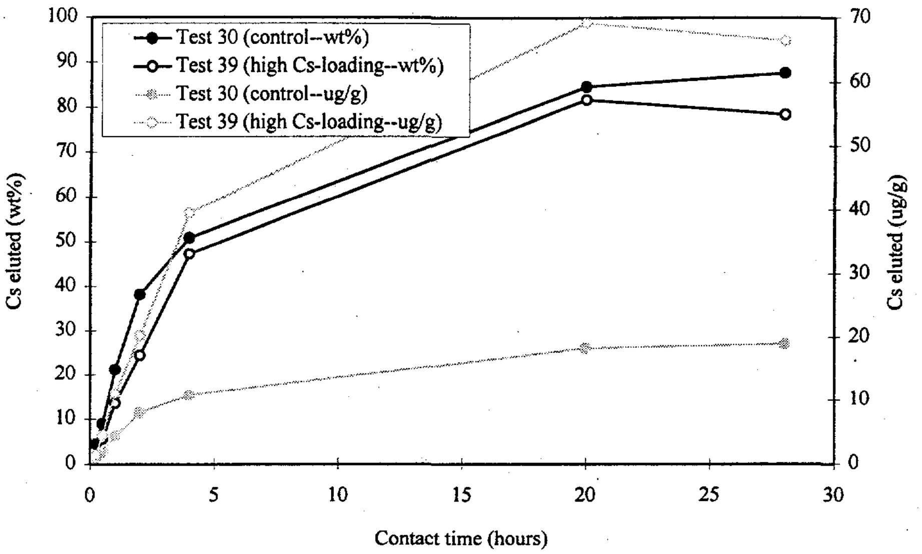

Figure 4.10. Weight Percent Cesium Eluted in Test to Determine the Effect of Using Higher Cesium-Loaded Zeolite on Cesium Elution

Table 4.9. Weight Percent Iron Dissolved in Tests to Determine the Effect of Using Higher Cesium-Loaded Zeolite in Cesium Elution Tests

sample	hours	control Test 30	high Cs Test 39
T1	0.25	1.22	1.79
T2	0.5	1.01	1.12
T3	1	1.88	1.82
T4	2	3.31	2.97
T5	4	5.15	5.65
T6	20	24.4	34.6
T7	28	33.7	48.2

4.6 Neutralization Tests

Tests 35 and 40 were designed to evaluate how neutralizing the oxalic acid with NaOH affected the amount of iron and cesium in solution. This test was done to simulate quenching the actual operations in tank 8D-1 to halt corrosion of the tank by the acid. These tests differed from the control in the following ways: after the 20-hour sample, the pH was recorded and 10 M NaOH was slowly added to the reaction vessel until the desired pH (12 in Test 35 and 5 in Test 40) was reached. Samples were then drawn at 15 minutes, 30 minutes, 1 hour, 2 hours, and 4 hours after neutralization. Cesium data from these tests are shown in Table 4.10 and Figure 4.12. As Figure 4.12 shows, neutralization has little effect on the amount of cesium eluted with the cesium concentration in solution being within experimental error of the control test. The amount of cesium eluted increases slightly after the test is neutralized which may be due to an equilibrium shift in the solution; however, this shift is very small and is within experimental error.

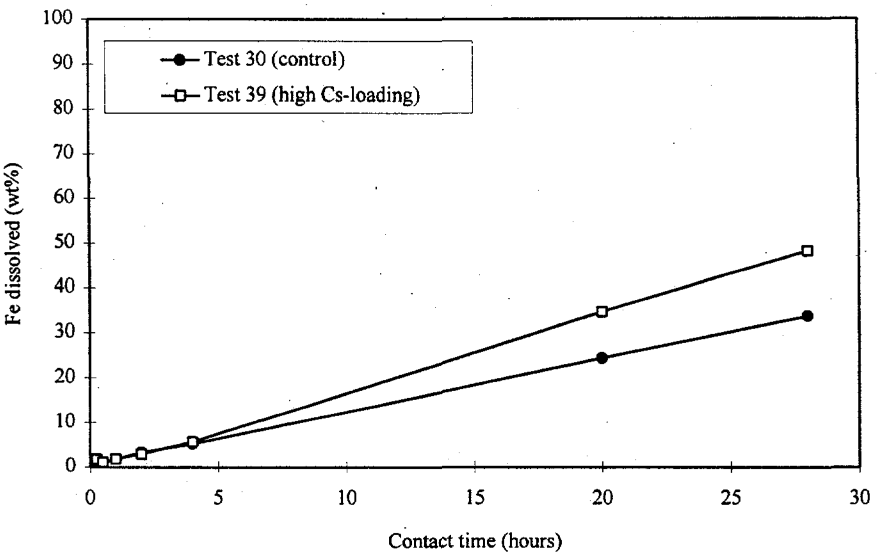

Figure 4.11. Weight Percent Iron Dissolved in Tests to Determine the Effect of Using Higher Cesium-Loaded Zeolite in Cesium Elution Tests

Table 4.10. Weight Percent Cesium Eluted in Neutralization Tests

sample	hours	Test 30 (control)	Test 35 (neutralize to pH 12)	Test 40 (neutralize to pH 5)
T1	0.25	4.45	3.58	--[a]
T2	0.5	9.02	9.64	--[a]
T3	1	21.21	38.73	--[a]
T4	2	38.14	48.91	--[a]
T5	4	50.88	65.13	--[a]
T6	20	84.66	80.25	86.86
N1	20.5	--[a]	88.97	92.52
N2	20.75	--[a]	84.70	93.04
N3	21.25	--[a]	87.72	92.78
N4	22.25	--[a]	82.34	92.52
N5	24.25	--[a]	89.44	94.57
T7	28	87.76	90.61	--[a]

(a) Not measured.

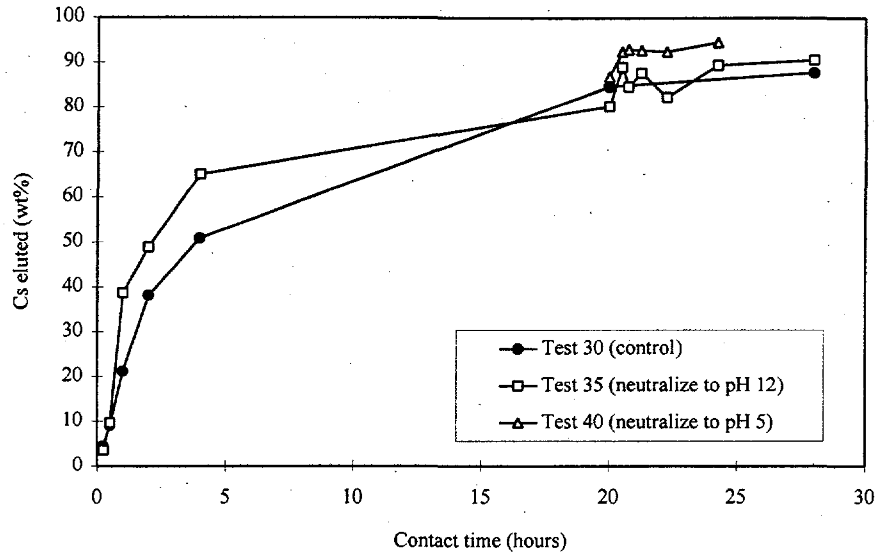

Figure 4.12. Weight Percent Cesium Eluted in Neutralization Tests

The amount of iron in solution in Tests 30, 35, and 40 is shown in Table 4.11 and Figure 4.13. The figure shows that the amount of iron in solution decreases at pH 12. However, at pH 5, the amount of iron in solution does not seem to be affected. In the control, about 33 wt% of the added iron is in solution after 28 hours. In Test 40, which was neutralized to pH 5, about 27 wt% of the iron remains four hours after neutralization, and in Test 35, which was neutralized to pH 12, only about 7 wt% of the iron remains in solution four hours later. Neutralization may decrease the amount of iron transferred to tank 8D-2 in the oxalic acid solution while having minimal impact on cesium elution.

Table 4.11. Weight Percent Iron Dissolved in Neutralization Tests

sample	hours	Test 30 (control)	Test 35 (pH 12.5)	Test 40 (pH 5)
T1	0.25	1.22	3.04	--(a)
T2	0.5	1.01	2.82	--(a)
T3	1	1.88	3.70	--(a)
T4	2	3.31	4.35	--(a)
T5	4	5.15	9.25	--(a)
T6	20	24.4	24.40	27.77
N1	20.5	--[a]	25.23	26.58
N2	20.75	--[a]	25.69	27.06
N3	21.25	--[a]	11.09	27.69
N4	22.25	--[a]	20.12	26.44
N5	24.25	--[a]	7.33	27.21
T7	28	33.7	9.55	--(a)

(a) Not measured.

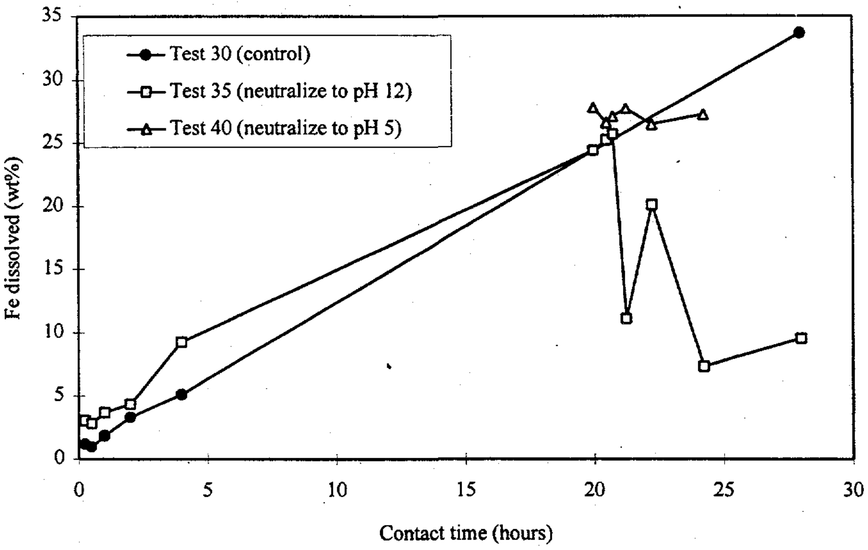

Figure 4.13. Weight Percent Iron Dissolved in Neutralization Tests

In Test 35, 149.8 g of 10 M NaOH were required to attain a pH of 12.5. The neutralization resulted in a large amount of precipitate on the bottom of the reaction vessel. The contents of the reaction vessel were filtered and dried, and 31.5 g of precipitate were recovered in contrast to 9.5 g recovered in the control. It is estimated that an additional 2-3 g were lost due to caking on the funnel and the bottom of the reaction vessel. X-ray diffraction analysis indicated that approximately 30 vol% of the precipitate was amorphous. Semi-quantitative analysis of the crystalline fraction showed 65 wt% $Na_2C_2O_4$ (sodium oxalate), and 35 wt% Fe_2O_3 (hematite). Table 4.12 summarizes the x-ray diffraction results for the precipitates recovered in the neutralization tests and the control.

In Test 40, 48.33 g of 10 M NaOH were required to attain a pH of 5. Neutralizing to a pH of 5 produced only 10.7 g of precipitate in contrast to 9.5 g for the control and >31.5 g for Test 35, in which the vessel contents were neutralized to a pH of 12. The amount of material recovered in Test 40 and in the control is less than the amount of material originally added to the reaction vessel (10 g zeolite and 4 g Fe_2O_3). X-ray diffraction analysis of the recovered solids indicated that approximately 45 vol% of the material was amorphous. Semi-quantitative analysis of the crystalline fraction showed it to contain 92 wt% Fe_2O_3 (hematite).

4.7 Relationship Between Cesium Eluted and Aluminum in Solution

The oxalic acid solutions were also analyzed for aluminum to determine the existence of a correlation between the amount of cesium eluted and the amount of aluminum in solution. The mechanism for cesium elution is a process by which aluminum and iron within the zeolite structure dissolve, creating a larger space around the cesium atom. Aluminum is one of the principal components of zeolite IE-96 (see Table 3.1 for zeolite composition), therefore, the amount of aluminum in solution could reflect the amount of zeolite dissolved and hence the level of cesium elution.

Table 4.12. X-Ray Diffraction Analysis of Precipitates in Neutralization Tests and Control

Test	Amorphous Content	Crystalline Fraction*
30 (control)	60 vol%	97 wt% Hematite, Fe_2O_3
35 (neutralize to pH 12.5)	30 vol%	65 wt% $Na_2C_2O_4$ 35 wt% Hematite, Fe_2O_3
40 (neutralize to pH 5)	45 vol%	92 wt% Hematite, Fe_2O_3 3 wt% Whewellite**, $CaC_2O_4 \cdot H_2O$

* semi-quantitative only

** tentative ID

Percentages of aluminum and cesium showed the same trend in each test, confirming a correlation (see Figure E.1). Figure 4.14 shows a plot of the molar ratio of aluminum to cesium in solution vs. contact time, and Figure 4.15 shows a plot of moles of aluminum vs. moles of cesium. The data for the higher cesium loading test were normalized to those of the other tests for comparison. The data plotted in these figures can be found in Appendix E. As Figure 4.14 shows, at short contact times the molar ratio of aluminum to cesium is high. However, as contact time increases, the ratio decreases quickly to a constant value. This suggests that initially several aluminum atoms must be eluted from the structure of the zeolite so that the cesium can escape. But as more and more aluminum is eluted, cesium escapes at a faster rate until a constant value is reached. Grinding placed more surface area in contact with the oxalic acid, so that both aluminum and cesium eluted at a faster rate. Therefore, the aluminum to cesium ratio for Test 36 reaches the limiting ratio more quickly. This is also shown in Figure 4.15, where concentration of cesium increases more rapidly with increasing concentration of aluminum.

Although there seems to be a definite correlation between the amount of aluminum dissolved and the amount of cesium eluted, it is recommended that the aluminum concentration not be used as an indicator of the cesium concentration until the mechanism is completely understood. Therefore, more studies should be performed to elucidate the mechanism that controls cesium elution and aluminum dissolution.

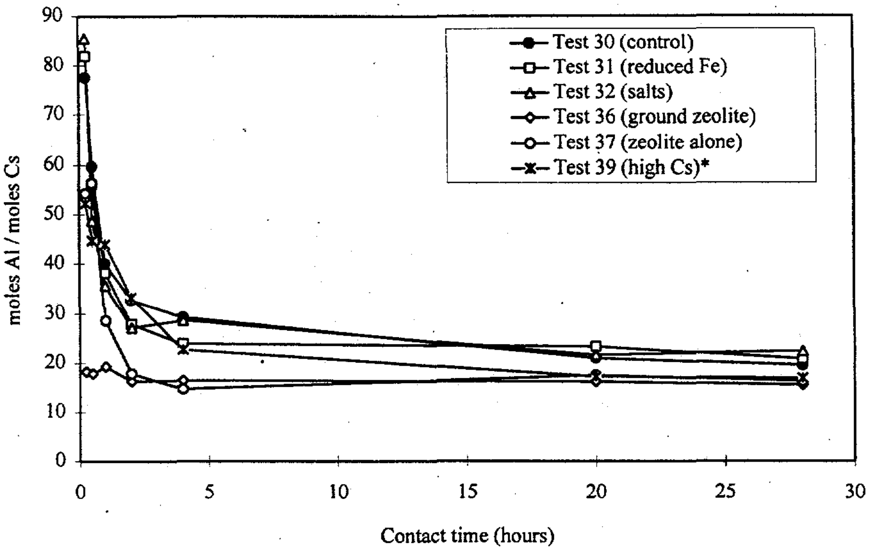

Figure 4.14. Moles of Aluminum/Moles of Cesium vs. Contact Time for Various Cesium Elution Tests
(* Normalized to control cesium loading)

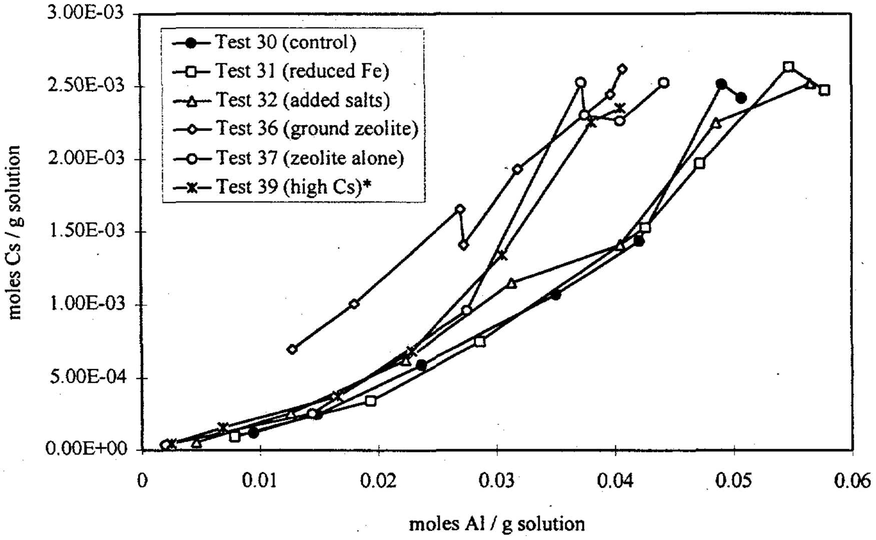

Figure 4.15. Moles of Cesium vs. Moles of Aluminum for Various Cesium Elution Tests
(* Normalized to control cesium loading)

5.0 References

Sills, JA, MR Elmore, GK Patello, WG Richmond, JS Roberts, RL Russell, and KD Wiemers. 1996. "Fiscal Year 1995 Laboratory Scale Studies of Cs Elution in Tank 8D-1 and Sludge Dissolution in Tank 8D-2." PNNL-10945. Pacific Northwest National Laboratory, Richland, WA.

Appendix A

Contact Conditions

Appendix A

Contact Conditions

Calculations for Loading Zeolite with Cesium

Need to determine how much cesium must be loaded onto zeolite to simulate cesium 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 pounds of zeolite in tank 8D-1 (at completion of Thorex transfer and third sludge wash) is 154,800 pounds of (this is based on 43 cv and 3600 pounds of (dry weight) zeolite/cv)
- total estimated Curies of cesium in tank 8D-1 (at end of third sludge wash) is 5534 kCi cesium
- 5534 kCi / 154,800 lb = 35.75 Ci/lb zeolite

Calculations: goal is to calculate grams cesium 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% will be)
Assume also that this represents all of the Cs present in the tank (no ^{133}Cs, etc.)

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

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

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

Then N can be calculated from

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

$$N = 1.8157 \times 10^{21}\ \text{atoms} \times \frac{1\ \text{mole}}{6.0225 \times 10^{23}\ \text{atoms}} \times \frac{132.9\ \text{g}}{\text{mole}} = 0.401\ \text{g}\ ^{137}\text{Cs / lb zeolite}$$

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

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.**

Conversations with LA Bray revealed that only ^{137}Cs was figured into these calculations. Since ^{137}Cs is only ~30% abundant, the cesium level in the zeolite is only about 1/3 of the actual cesium content. For Test 39, zeolite was loaded with the actual amount of cesium to determine whether elution behavior would be affected by a change in cesium loading.

If a loading of 0.886 g Cs / kg zeolite yields 30% of the amount of Cs that is in the West Valley zeolite, then the amount required for 100% loading is 0.886/.30 = 2.954 g Cs/kg zeolite. Calculations are for 25 g of zeolite,

$$2.954\ \text{g Cs/kg zeolite} \times 0.025\ \text{kg zeolite} = 7.385 \times 10^{-2}\ \text{g Cs}$$

7.385×10^{-2} g Cs per 25 g zeolite

$$7.385 \times 10^{-2}\ \text{g Cs} \times \frac{1\ \text{mole}}{132.9054\ \text{g Cs}} = 5.557 \times 10^{-4}\ \text{moles Cs}$$

Cs will be in the form of $CsNO_3$, with a formula weight of 194.909 g/mole
We will require 5.557×10^{-4} moles of $CsNO_3$ for 5.557×10^{-4} moles Cs

$$5.557 \times 10^{-4}\ \text{moles} \times \frac{194.909\ \text{g}}{\text{mole}} = 0.10831\ \text{g}\ CsNO_3$$

So for 25 g zeolite (dry weight), will require 0.11 g $CsNO_3$ to get the correct loading.

Contact Conditions for Cs-Loading

Table A.1. Conditions for Cesium-Loading of Zeolite

Batch	1	2*	3
Used for Tests	30,31,32,35,37,40	36	39
Cs Loading, µg/g	922	995	3615
Zeolite, g (dry weight)	125	125	25
Deionized Water, g	500.1	500.1	100.1
$CsNO_3$, g	0.1631	0.1631	0.11

* *this was material from batch 1, ground and re-analyzed for Cs level*

Comparison of Amount of NaOH Used in FY 1995 and FY 1996

FY 1996: used 40g of 0.045M NaOH per 10g zeolite. (assume density = 1.00g/mL)

(0.045 moles NaOH/L)(.04L/10g zeolite)(40g NaOH/mole NaOH) = 7.2 mg NaOH/g zeolite

FY 1995: used 2.77g of 0.001M NaOH per 5g zeolite. (assume density = 1.00 g/mL)

(0.001 moles NaOH/L)(.00277L/5g zeolite)(40g NaOH/mole NaOH) = 0.02 mg NaOH/g zeolite

Appendix B

Cesium Elution Test Conditions

Appendix B

Cesium Elution Test Conditions

Test Conditions

Constants:
oxalic acid concentration: 8 wt%
oxalic acid:zeolite ratio: 40 L/kg zeolite
amount of zeolite: 10 g, dry weight
amount of 0.045 M NaOH (heel): 4 L/kg zeolite
acid addition temperature: 25°C
bath temperature: 50°C
number of contacts: 1
time per contact: 28 hours
agitation: stir

Sampling Schedule:
15 minutes contact
30 minutes contact
1 hour contact
2 hours contact
4 hours contact
20 hours contact
28 hours contact

Table B.1. Test Matrix Used for Cs Elution Tests 30 - 40

Test	30	31	32	33	34	35	36	37	38	39	40
zeolite, g	10	10	10	0	0	10	10	10	0	10	10
NaOH*, L/kg	4	4	4	0	0	4	4	4	4	4	4
Fe_2O_3, g	4	2	4	16	0	4	4	0	0	4	4
Al_2O_3, g	0	0	0	0	16	0	0	0	0	0	0
$NaNO_3$, g	0	0	0.11	0	0	0	0	0	0	0	0
$NaNO_2$, g	0	0	0.36	0	0	0	0	0	0	0	0
coupon present									●		
neutralize**						●					●
Analyses:											
ICP-MS for Cs	●	●	●	●	●	●	●	●	●	●	●
ICP-MS for Fe	●	●	●	●	●	●	●	●	●	●	●
ICP-MS for Al	●	●	●	●	●	●	●	●	●	●	●
XRD on zeolite	●					●					●
particle size	●						●				

* 0.045 M NaOH

** neutralization uses 10 M NaOH

Appendix C

Temperature and pH Data for Cesium Elution Tests

Appendix C
Temperature and pH Data for Cesium Elution Tests

Table C.1. Temperature and pH Data for Cs Elution Tests 30 - 34 and 36-39

	Test 30		Test 31		Test 32		Test 33		Test 34	
	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
time 0	1.230	22	1.060	22	0.818	22	1.380	20	1.403	20
15 minutes	--	46	--	38	--	42	--	48	--	33
30 minutes	--	53	--	51	--	62	--	61	--	52
1 hour	--	51	--	49	--	57	--	61	--	56
2 hours	--	50	--	48	--	49	--	54	--	51
4 hours	--	50	--	49	--	51	--	49	--	49
20 hours	--	51	--	49	--	49	--	49	--	49
28 hours	0.858	49	0.818	49	0.863	49	0.760	49	0.769	49

Test	Test 36		Test 37		Test 38		Test 39	
	pH	T (°C)	pH	T (°C)	pH	T (°C)	pH	T (°C)
time 0	1.488	22	1.445	22	1.330	22	1.051	22
15 minutes	--	46	--	41	--	53	--	45
30 minutes	--	66	--	53	--	56	--	55
1 hour	--	61	--	53	--	52	--	53
2 hours	--	49	--	49	--	51	--	49
4 hours	--	49	--	49	--	49	--	49
20 hours	--	51	--	50	--	51	--	49
28 hours	0.732	48	0.656	50	0.627	53	0.683	49

Table C.2. Temperature and pH Data for Cs Elution Tests 35 and 40

Test	Test 35		Test 40	
	pH	T (°C)	pH	T (°C)
time 0	1.042	20	0.701	22
15 minutes	--	47	--	--
30 minutes	--	62	--	--
1 hour	--	58	--	--
2 hours	--	52	--	--
4 hours	--	49	--	--
neutralization (20 hours)	0.810	52	0.684	51
15 min	12.554	50	5.366	53
30 min	12.653	48	5.471	50
1 hour	12.444	58	5.571	51
2 hours	12.449	50	5.681	51
4 hours	12.383	53	5.764	48
28 hours	13.123	53	--	--

-- = not measured

Appendix D

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

Appendix D

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

Table D.1. Sample Calculations and Formulas Used in Determination of Percent Cs Eluted in Cs Elution Tests 30 - 40

	A	B	C
1			
2	**Assembly number**	**Test 35**	**source of value**
3	**Contact 1, time 0: initial conditions**		
4	initial amount of zeolite in reaction vessel, g	10.0	measured in laboratory
5	F factor of zeolite (=dry weight/wet weight)	1.0	dry zeolite used; F-factor = 1
6	amount of dry zeolite used, g	10.0	=C4*C5
7	μg element/g dry zeolite	922.0	XRF measurement
8	total amount element initially present in vessel, μg	9220.0	=C6*C7
9	weight of oxalic acid initially present, g	407.6	measured in laboratory
10	weight of NaOH initially present, g	40.0	measured in laboratory
11	weight of salts initially present, g	0.0	measured in laboratory
12	weight of Al2O3 initially present, g	0.0	measured in laboratory
13	weight of Fe2O3 initially present, g	4.0	measured in laboratory
14	total initial solution weight, g	447.6	=C9+C10
15	weight of empty vessel, g	1995.7	measured in laboratory
16	weight of loaded vessel, g	2457.3	=C15+C6+SUM(C9:C13)
17	weight loss per hour, g	0.0	measured in laboratory
18	**Time 1 = 15 minutes**	0.25	measured in laboratory
19	Weight of material present at T1, g	447.6	=C14-(C18*C17)
20	Weight of sample removed at T1, g	5.0	measured in laboratory
21	element concentration, run 1, μg element/g sample	0.7	ICP-MS measurement
22	element concentration, run 2, μg element/g sample	--	not run in duplicate
23	average element concentration, μg/g	0.7	=C21
24	amount of element in solution at C1T1, μg	330.3	=C19*C23
25	**percentage element dissolved at T1**	**3.6**	**=(C24/C8)*100**
26	**Time 2 = 30 minutes**	0.5	measured in laboratory
27	Weight of material present at T2, g	442.6	=C19-C20-(C26-C18)*C17
28	Weight of sample removed at T2, g	5.1	measured in laboratory
29	element concentration, run 1, μg element/g sample	2.0	ICP-MS measurement
30	element concentration, run 2, μg element/g sample	--	not run in duplicate
31	average element concentration, μg/g	2.0	=C29
32	amount of element in solution at T2, μg	885.2	=C27*C31
33	amount of element removed during T1 sampling, μg	3.7	=C20*C23
34	total amount of element dissolved at T2, μg	888.9	=C32*C33
35	**percentage of element dissolved at T2**	**9.6**	**=(C34/C8)*100**
36	**Time 3 = 1 hour**	1.0	measured in laboratory
37	weight of material present at T3, g	437.5	=C27-C28-(C36-C26)*C17
38	weight of sample removed at T3, g	5.0	measured in laboratory

Table D.1 (contd)

	A	B	C
39	element concentration, run 1, µg element/g sample	8.1	ICP-MS measurement
40	element concentration, run 2, µg element/g sample	--	not run in duplicate
41	average element concentration, µg/g	8.1	=C39
42	amount of element in solution at T3, µg	3556.9	=C37*C41
43	amount of element removed during T1 sampling, µg	3.7	=C33
44	amount of element removed during T2 sampling, µg	10.2	=C28*C31
45	total amount of element removed during sampling, µg	13.9	=SUM(C43:C44)
46	total amount of element dissolved at T3, µg	3570.7	=C42+C45
47	**percentage of element dissolved at T3**	**38.7**	**=(C46/C8)*100**
48	**Time 4 = 2 hours**	2.0	measured in laboratory
49	weight of material present at T4, g	432.5	=C37-C38-(C48-C36)*C17
50	weight of sample removed at T4, g	5.1	measured in laboratory
51	element concentration, run 1, µg element/g sample	10.3	ICP-MS measurement
52	element concentration, run 2, µg element/g sample	--	not run in duplicate
53	average element concentration, µg/g	10.3	=C51
54	amount of element in solution at T4, µg	4455.0	=C49*C53
55	amount of element removed during T1 sampling, µg	3.7	=C33
56	amount of element removed during T2 sampling, µg	10.2	=C44
57	amount of element removed during T3 sampling, µg	40.5	=C38*C41
58	total amount of element removed during sampling, µg	54.4	=SUM(C55:C57)
59	total amount of element dissolved at T4, µg	4509.3	=C54+C58
60	**percentage of element dissolved at T4**	**48.9**	**=(C59/C8)*100**
61	**Time 5 = 4 hours**	4.0	measured in laboratory
62	weight of material present at T5, g	427.4	=C49-C50-(C61-C48)*C17
63	weight of sample removed at T5, g	5.1	measured in laboratory
64	element concentration, run 1, µg element/g sample	13.8	ICP-MS measurement
65	element concentration, run 2, µg element/g sample	--	not run in duplicate
66	average element concentration, µg/g	13.8	=C64
67	amount of element in solution at T5, µg	5898.4	=C62*C66
68	amount of element removed during T1 sampling, µg	3.7	=C33
69	amount of element removed during T2 sampling, µg	10.2	=C44
70	amount of element removed during T3 sampling, µg	40.5	=C57
71	amount of element removed during T4 sampling, µg	52.5	=C50*C53
72	total amount of element removed during sampling, µg	106.9	=SUM(C68:C71)
73	total amount of element dissolved at T5, µg	6005.3	=C67+C72
74	**percentage of element dissolved at T5**	**65.1**	**=(C73/C8)*100**
75	**Time 6 = 20 hours**	20.0	measured in laboratory
76	weight of material present at T6, g	422.3	=C62-C63-(C75-C61)*C17
77	weight of sample removed at T6, g	5.1	measured in laboratory
78	element concentration, run 1, µg element/g sample	17.1	ICP-MS measurement
79	element concentration, run 2, µg element/g sample	--	not run in duplicate
80	average element concentration, µg/g	17.1	=C78
81	amount of element in solution at T6, µg	7221.8	=C76*C80
82	amount of element removed during T1 sampling, µg	3.7	=C33
83	amount of element removed during T2 sampling, µg	10.2	=C69
84	amount of element removed during T3 sampling, µg	40.5	=C70

Table D.1 (contd)

	A	B	C
85	amount of element removed during T4 sampling, µg	52.5	=C71
86	amount of element removed during T5 sampling, µg	70.2	=C63*C66
87	total amount of element removed during sampling, µg	177.1	=SUM(C82:C86)
88	total amount of element dissolved at T6, µg	7399.0	=C81+C87
89	**percentage of element dissolved at T6**	**80.2**	**=(C88/C8)*100**
90	**Neutralization**		
91	time to reach pH ~ 5	0.25	measured in laboratory
92	weight of NaOH added to neutralize	149.8	measured in laboratory
93	Neutralization time 1 = 15 minutes	20.5	=C75+C91+0.25
94	weight of material present at N1	567.0	=C76-C77-(C93-C75)*C17+C92
95	weight of sample removed at N1	5.3	measured in laboratory
96	element concentration, run 1, µg element/g sample	14.5	ICP-MS measurement
97	element concentration, run 2, µg element/g sample	13.5	duplicate ICP-MS measurement
98	average element concentration, µg/g	14.0	=(C96+C97)/2
99	*amount of element in solution at N1, µg*	7938.6	=C94*C98
100	amount of element removed during T1 sampling, µg	3.7	=C33
101	amount of element removed during T2 sampling, µg	10.2	=C69
102	amount of element removed during T3 sampling, µg	40.5	=C70
103	amount of element removed during T4 sampling, µg	52.5	=C71
104	amount of element removed during T5 sampling, µg	70.2	=C86
105	amount of element removed during T6 sampling, µg	87.0	=C77*C80
106	total amount of element removed during sampling, µg	264.2	=SUM(C100:C105)
107	total amount of element dissolved at N1, µg	8202.7	=C99+C106
108	**percentage of element dissolved at N1**	**89.0**	**=(C107/C8)*100**
109	**Neutralization time 2 = 30 minutes**	20.75	=C93+0.25
110	weight of material present at N2	561.7	=C94-C95-(C109-C93)*C17
111	weight of sample removed at N2	5.4	measured in laboratory
112	element concentration, run 1, µg element/g sample	13.3	ICP-MS measurement
113	element concentration, run 2, µg element/g sample	--	not run in duplicate
114	average element concentration, µg/g	13.3	=C112
115	amount of element removed during T1-N1, µg	338.9	=C106+(C95*C98)
116	amount of element in solution at N2, µg	7470.6	=C110*C114
117	total amount of element dissolved at N2, µg	7809.5	=C115+C116
118	**percentage of element dissolved at N2**	**84.7**	**=(C117/C8)*100**
119	**Neutralization time 3 = 1 hour**	21.25	=C109+0.5
120	weight of material present at N3	556.3	=C110-C111-(C119-C109)*C17
121	weight of sample removed at N3	5.4	measured in laboratory
122	element concentration, run 1, µg element/g sample	13.8	ICP-MS measurement
123	element concentration, run 2, µg element/g sample	–	not run in duplicate
124	average element concentration, µg/g	13.8	=C122
125	amount of element removed during T1-N2, µg	410.3	=C115+(C111*C114)
126	amount of element in solution at N3, µg	7677.4	=C120*C124
127	total amount of element dissolved at N3, µg	8087.7	=C125+C126
128	**percentage of element dissolved at N3**	**87.7**	**=(C127/C8)*100**
129	**Neutralization time 4 = 2 hours**	22.25	=C119+1

Table D.1 (contd)

	A	B	C
130	weight of material present at N4	551.0	=C120-C121-(C129-C119)*C17
131	weight of sample removed at N4	5.3	measured in laboratory
132	element concentration, run 1, µg element/g sample	12.9	ICP-MS measurement
133	element concentration, run 2, µg element/g sample	--	not run in duplicate
134	average element concentration, µg/g	12.9	=C132
135	amount of element removed during T1-N3, µg	484.5	=C125+(C121*C124)
136	amount of element in solution at N4, µg	7107.4	=C130*C134
137	total amount of element dissolved at N4, µg	7591.8	=C135+C136
138	**percentage of element dissolved at N4**	**82.3**	**=(C137/C8)*100**
139	**Neutralization time 5 = 4 hours**	24.25	=C129+2
140	weight of material present at N5	545.6	=C130-C131-(C139-C129)*C17
141	weight of sample removed at N5	5.3	measured in laboratory
142	element concentration, run 1, µg element/g sample	14.1	ICP-MS measurement
143	element concentration, run 2, µg element/g sample	--	not measured in duplicate
144	average element concentration, µg/g	14.1	=C142
145	amount element removed during T1-N4, µg	553.2	=C135+(C131*C134)
146	amount of element in solution at N5, µg	7693.4	=C140+C144
147	total amount of element dissolved at N5, µg	8246.6	=C145+C146
148	**percentage of element dissolved at N5**	**89.4**	**=(C147/C8)*100**
149	**Time 7 = 28 hours**	28.0	measured in laboratory
150	weight of material present at T7, g	540.3	=C140-C141-(C149-C139)*C17
151	weight of sample removed at T7, g	5.3	measured in laboratory
152	element concentration, run 1, µg element/g sample	14.3	ICP-MS measurement
153	element concentration, run 2, µg element/g sample	--	not run in duplicate
154	average element concentration, µg/g	14.3	=C152
155	amount of element removed during T1-T7, µg	628.2	=C145+(C141*C144)
156	amount of element in solution at T7, µg	7726.4	=C150+C154
157	total amount of element dissolved at T7, µg	8354.7	=C155+C156
158	**percentage of element dissolved at T7**	**90.6**	**=(C157/C8)*100**

Appendix E

Data: Cesium:Aluminum

Appendix E

Data: Cesium:Aluminum

Table E.1. Weight Percent Cesium Eluted and Aluminum Dissolved in Experiments to Determine the Effect of the Type and Amount of Iron Present on Cesium Elution

sample	hours	control		< Fe		zeolite	
		Al, Test 30	Cs, Test 30	Al, Test 31	Cs, Test 31	Al, Test 37	Cs, Test 37
T1	0.25	19.1	4.45	15.9	3.51	4.15	1.38
T2	0.5	29.9	9.02	38.9	12.5	28.9	9.23
T3	1	47.3	21.2	57.2	26.9	54.8	34.4
T4	2	69.6	38.1	84.6	54.4	80.1	80.2
T5	4	83.1	50.9	93.4	69.7	73.8	89.5
T6	20	99.5	84.7	113	86.9	87.1	89.5
T7	28	96.4	87.8	107	92.3	74.4	81.8

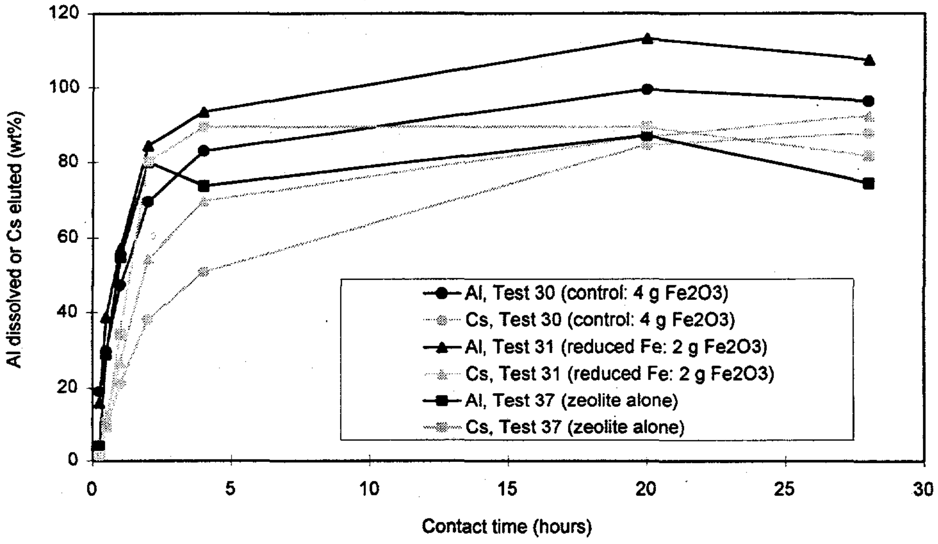

Figure E.1. Percentage of Aluminum Dissolved in Tests Containing Various Amounts of Iron

Table E.2. Concentration of Aluminum in Solution in μg/g

sample	hours	control Test 30	pure Al_2O_3 Test 34	zeolite Test 37
T1	0.25	351	0.563	76
T2	0.5	550	0.863	534
T3	1	877	0.125	1020
T4	2	1300	2.08	1500
T5	4	1560	2.53	1380
T6	20	1880	4.40	1640
T7	28	1820	5.28	1390

Note that more aluminum was dissolved in the control (Test 30), in the presence of Fe_2O_3, than in Test 37, which contained only zeolite and no Fe_2O_3. Also note that pure Al_2O_3 does not appear to be soluble in oxalic acid, and that the phase of Al_2O_3 used in Test 34 is obviously not the same phase of Al_2O_3 in zeolite.

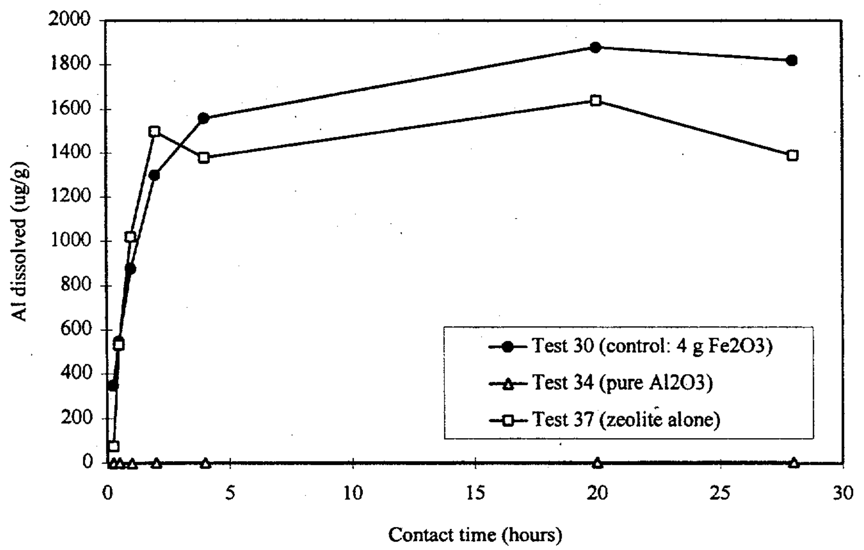

Figure E.2. Concentration of Aluminum in Solution, in μg/g

Table E.3. Weight Percent Cesium Eluted and Aluminum Dissolved in Tests to Determine Effect of Added Salts on Cesium Elution

sample	hours	control		salts	
		Al, Test 30	Cs, Test 30	Al, Test 32	Cs, Test 32
T1	0.25	19.1	4.45	9.33	1.97
T2	0.5	29.9	9.02	25.3	9.33
T3	1	47.3	21.2	44.5	22.4
T4	2	69.6	38.1	62.1	40.9
T5	4	83.1	50.9	79.8	50.0
T6	20	99.5	84.7	95.0	78.7
T7	28	96.4	87.8	110	87.8

The presence of added salts appears to have no effect on aluminum dissolution.

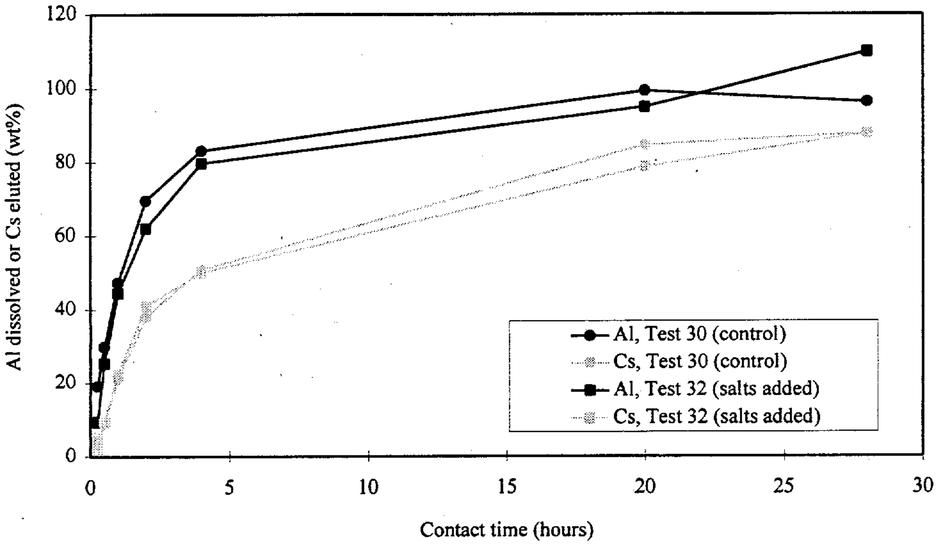

Figure E.3. Weight Percent Cesium Eluted and Aluminum Dissolved in Tests to Determine Effect of Added Salts on Cesium Elution

Table E.4. Weight Percent Cesium Eluted and Aluminum Dissolved in Tests to Determine Effect of Reduced Particle Size Zeolite on Cesium Elution

sample	hours	control		reduced particle size	
		Al, Test 30	Cs, Test 30	Al, Test 36	Cs, Test 36
T1	0.25	19.1	4.45	25.8	23.6
T2	0.5	29.9	9.02	36.3	34.0
T3	1	47.3	21.2	54.6	47.3
T4	2	69.6	38.1	54.1	55.4
T5	4	83.1	50.9	63.4	64.2
T6	20	99.5	84.7	78.3	80.7
T7	28	96.4	87.8	80.3	86.1

Figure E.4 and Table E.4 show that the amount of Al dissolved in the reduced particle size test (Test 36) was less than the amount dissolved in the control (Test 30). Because of the grinding, the Al to Cs ratio is already at the constant value since the structure of the zeolite has been broken down to release the Cs more readily.

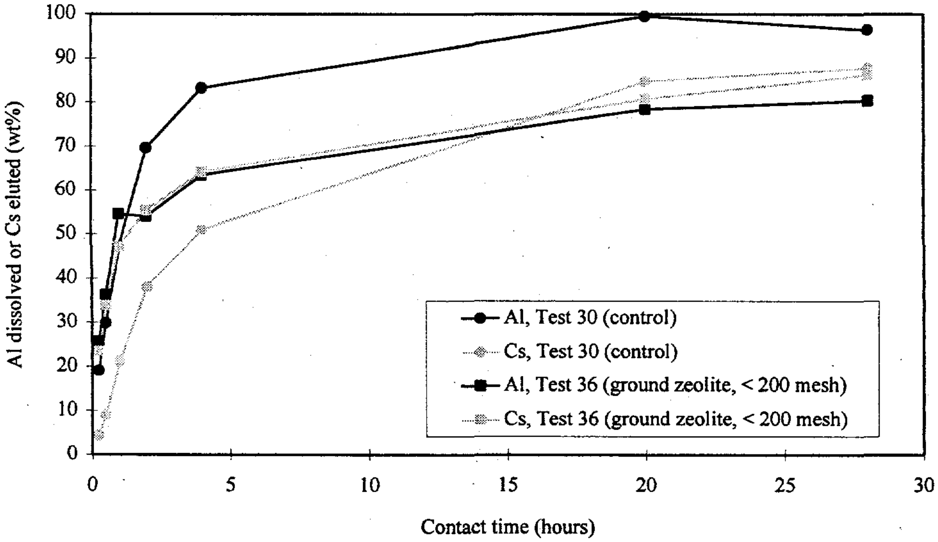

Figure E.4. Weight Percent Cesium Eluted and Aluminum Dissolved in Tests to Determine Effect of Reduced Particle Size Zeolite on Cesium Elution

Table E.5. Weight Percent Cesium Eluted and Aluminum Dissolved in Tests to Determine Effect of Using Higher-Loaded Zeolite on Cs Elution

sample	hours	control		high Cs	
		Al, Test 30	Cs, Test 30	Al, Test 39	Cs, Test 39
T1	0.25	19.1	4.45	5.02	1.73
T2	0.5	29.9	9.02	13.8	5.57
T3	1	47.3	21.2	33.0	13.5
T4	2	69.6	38.1	45.2	24.4
T5	4	83.1	50.9	60.0	47.2
T6	20	99.5	84.7	78.9	81.8
T7	28	96.4	87.8	74.2	78.5

Although the amounts of Cs eluted in Tests 30 and 39 were essentially the same, the amounts of Al dissolved were significantly higher in the control than in the test containing higher level Cs-loaded zeolite.

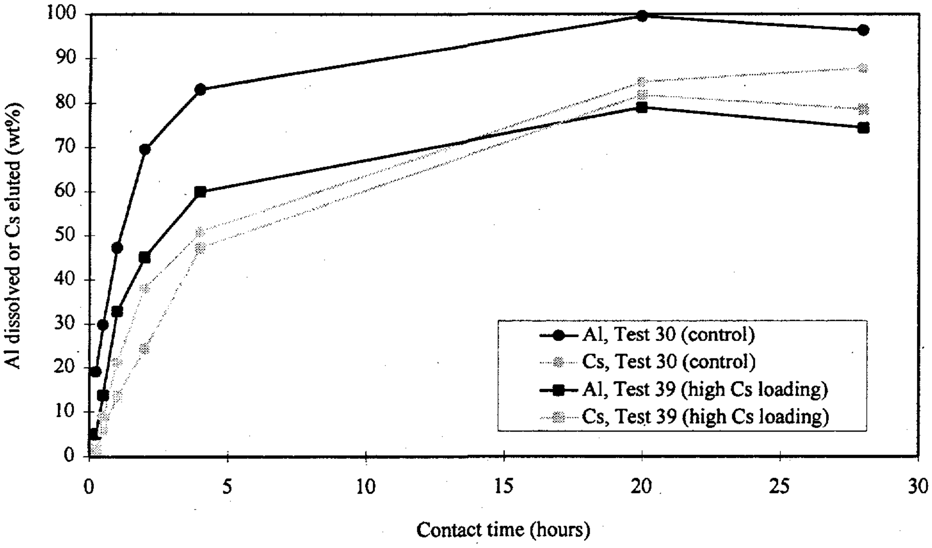

Figure E.5. Weight Percent Cesium Eluted and Aluminum Dissolved in Tests to Determine Effect of Using Higher-Loaded Zeolite on Cesium Elution

Table E.6. Weight Percent Aluminum Dissolved in Neutralization Tests

sample	hours	Al, Test 30 (control)	Cs, Test 30 (control)	Al, Test 35 (neutralize to pH 12)	Cs, Test 35 (neutralize to pH 12)	Al, Test 40 (neutralize to pH 5)	Cs, Test 40 (neutralize to pH 5)
T1	0.25	19.12	4.45	13.09	3.58		
T2	0.5	29.88	9.02	23.78	9.64		
T3	1	47.31	21.21	49.00	38.73		
T4	2	69.61	38.14	54.22	48.91		
T5	4	83.14	50.88	67.77	65.13		
T6	20	99.47	84.66	67.77	80.25	92.17	86.86
N1	20.5			83.01	88.97	87.76	92.52
N2	20.75			73.77	84.70	88.95	93.04
N3	21.25			71.05	87.72	88.95	92.78
N4	22.25			70.52	82.34	86.05	92.52
N5	24.25			53.49	89.44	85.48	94.57
T7	28	96.35	87.76	44.99	90.61		

The figure shows that, as in the case with Fe dissolution, the amount of Al in solution decreases with increased pH.

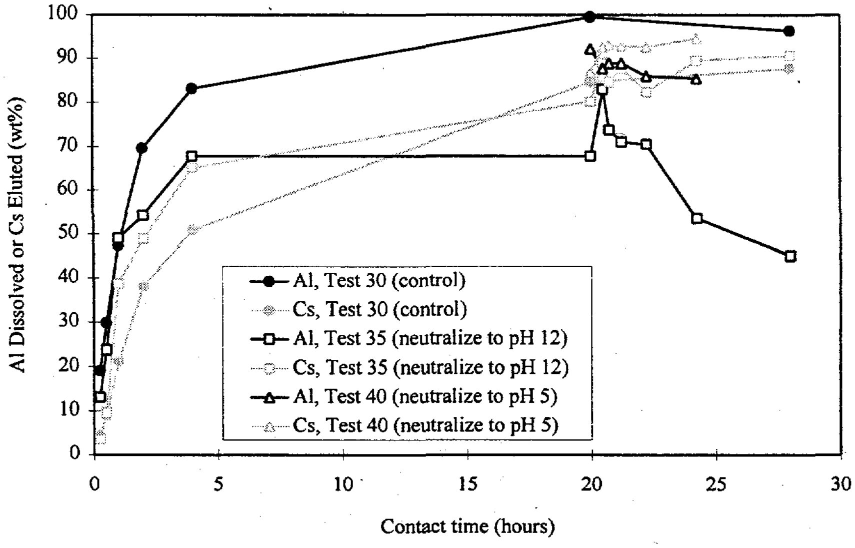

Figure E.6. Weight Percent Aluminum Dissolved and Percent Cesium Eluted in Neutralization Tests

Table E.7(a). Test 30: Data Used in Figure 4.13, Plotting Moles Al/Moles Cs

	Test 30: control				
	Al	Al	Cs	Cs	Al/Cs
hours	µg/g	mol/g	µg/g	mol/g	
0.25	351	9.46E-03	0.917	1.22E-04	77.60
0.5	550	1.48E-02	1.87	2.49E-04	59.71
1	877	2.37E-02	4.44	5.90E-04	40.10
2	1300	3.51E-02	8.05	1.07E-03	32.79
4	1560	4.21E-02	10.8	1.44E-03	29.32
20	1880	5.07E-02	18.2	2.42E-03	20.97
28	1820	4.91E-02	18.9	2.51E-03	19.55

Table E.7(b). Test 31: Data Used in Figure 4.13, Plotting Moles Al/Moles Cs

	Test 31: reduced Fe				
	Al	Al	Cs	Cs	Al/Cs
hours	µg/g	mol/g	µg/g	mol/g	
0.25	292	7.88E-03	0.724	9.62E-05	81.88
0.5	717	1.93E-02	2.59	3.44E-04	56.20
1	1060	2.86E-02	5.63	7.48E-04	38.22
2	1580	4.26E-02	11.5	1.53E-03	27.89
4	1750	4.72E-02	14.8	1.97E-03	24.01
20	2140	5.77E-02	18.6	2.47E-03	23.36
28	2030	5.48E-02	19.8	2.63E-03	20.81

Table E.7(c). Test 32: Data Used in Figure 4.13, Plotting Moles Al/Moles Cs

	Test 32: added salts				
	Al	Al	Cs	Cs	Al/Cs
hours	µg/g	mol/g	µg/g	mol/g	
0.25	171	4.61E-03	0.406	5.40E-05	85.51
0.5	467	1.26E-02	1.94	2.58E-04	48.87
1	827	2.23E-02	4.70	6.25E-04	35.72
2	1160	3.13E-02	8.64	1.15E-03	27.26
4	1500	4.05E-02	10.6	1.41E-03	28.73
20	1800	4.86E-02	16.9	2.25E-03	21.62
28	2095	5.65E-02	19.0	2.52E-03	22.44

Table E.7(d). Test 36: Data Used in Figure 4.13, Plotting Moles Al/Moles Cs

	Test 36: ground zeolite				
hours	Al µg/g	Al mol/g	Cs µg/g	Cs mol/g	Al/Cs
0.25	472	1.27E-02	5.24	6.96E-04	18.29
0.5	668	1.80E-02	7.57	1.01E-03	17.91
1	1010	2.73E-02	10.6	1.41E-03	19.34
2	1000	2.70E-02	12.5	1.65E-03	16.31
4	1180	3.18E-02	14.5	1.93E-03	16.52
20	1470	3.97E-02	18.4	2.45E-03	16.22
28	1510	4.07E-02	19.7	2.62E-03	15.56

Table E.7(e). Test 37: Data Used in Figure 4.13, Plotting Moles Al/Moles Cs

	Test 37: zeolite alone				
hours	Al µg/g	Al mol/g	Cs µg/g	Cs mol/g	Al/Cs
0.25	76	2.05E-03	0.284	3.77E-05	54.33
0.5	534	1.44E-02	1.92	2.55E-04	56.46
1	1020	2.75E-02	7.22	9.60E-04	28.68
2	1500	4.05E-02	17.0	2.26E-03	17.91
4	1380	3.72E-02	19.0	2.53E-03	14.75
20	1640	4.43E-02	19.0	2.53E-03	17.52
28	1390	3.75E-02	17.3	2.30E-03	16.31

Table E.7(f). Test 39: Data Used in Figure 4.13, Plotting Moles Al/Moles Cs

	Test 39: high Cs				
hours	Al µg/g	Al mol/g	Cs µg/g	Cs mol/g	Al/Cs
0.25	92	2.48E-03	1.40	1.86E-04	13.34
0.5	255	6.88E-03	4.53	6.02E-04	11.43
1	614	1.66E-02	11.1	1.48E-03	11.23
2	846	2.28E-02	20.2	2.68E-03	8.50
4	1130	3.05E-02	39.5	5.25E-03	5.81
20	1500	4.05E-02	69.2	9.20E-03	4.40
28	1410	3.80E-02	66.4	8.82E-03	4.31

Table E.8. Data Used in Figure 4.14, Plotting Moles Al versus Moles Cs

moles Al per g solution	moles Cs per g solution					
	Test 30	**Test 31**	**Test 32**	**Test 36**	**Test 37**	**Test 39**
2.05E-03					3.77E-05	
2.48E-03						1.86E-04
4.61E-03			5.40E-05			
6.88E-03						6.02E-04
7.88E-03		9.62E-05				
9.46E-03	1.22E-04					
1.26E-02			2.58E-04			
1.27E-02				6.96E-04		
1.44E-02					2.55E-04	
1.48E-02	2.49E-04					
1.66E-02						1.48E-03
1.80E-02				1.01E-03		
1.93E-02		3.44E-04				
2.23E-02			6.25E-04			
2.28E-02						2.68E-03
2.37E-02	5.90E-04					
2.70E-02				1.65E-03		
2.73E-02				1.41E-03		
2.75E-02					9.60E-04	
2.86E-02		7.48E-04				
3.05E-02						5.25E-03
3.13E-02			1.15E-03			
3.18E-02				1.93E-03		
3.51E-02	1.07E-03					
3.72E-02					2.53E-03	
3.75E-02					2.30E-03	
3.80E-02						8.82E-03
3.97E-02				2.45E-03		
4.05E-02			1.41E-03			
4.05E-02					2.26E-03	
4.05E-02						9.20E-03
4.07E-02				2.62E-03		
4.21E-02	1.44E-03					
4.26E-02		1.53E-03				
4.43E-02					2.53E-03	
4.72E-02		1.97E-03				
4.86E-02			2.25E-03			
4.91E-02	2.51E-03					
5.07E-02	2.42E-03					
5.48E-02		2.63E-03				
5.65E-02			2.52E-03			
5.77E-02		2.47E-03				

PNNL-11274
UC-2000

Distribution

No. of Copies

OFFSITE

2 DOE/Office of Scientific and Technical Information

6 C. S. King
West Valley Nuclear Services Co., Inc.
P.O. Box 191-10282 Rock Springs Road
West Valley, NY 141710191

ONSITE

1 **U.S. Department of Energy Richland Operations Office**

R. A. Gilbert	K6-51

4 **Westinghouse Hanford Company**

R. A. Kirkbride	K5-27
P. S. Schaus	G3-21
G. E. Stegen	H5-27
J. A. Voogd	H5-03

No. of Copies

29 **Pacific Northwest National Laboratory**

L. A. Bray	P7-28
K. P. Brooks	P7-43
J. L. Buelt	P7-41
M. L. Elliott	P7-41
M. R. Elmore	K3-75
J. G. H. Getting	P7-19
L. K. Holton	K9-73
D. E. Kurath	P7-20
E. V. Morrey	P7-19
G. K. Patello (4)	P7-28
J. M. Perez	P7-41
W. G. Richmond	K6-51
R. L. Russell	P7-28
G. L. Smith	P7-41
H. D. Smith	P7-28
J. M. Tingey	P7-25
K. D. Wiemers	K6-51
K. R. Savard	K9-04
Information Release (5)	K1-06