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Release of Radionuclides and Chelating Agents From Cement- Solidified Decontamination Low-Level Radioactive Waste Collected From the Peach Bottom Atomic Power Station Unit 3

Prepared by
D. W. Akers, N. C. Kraft, J. W. Mandler

Idaho National Engineering Laboratory
EG&G Idaho, Inc.

Prepared for
U.S. Nuclear Regulatory Commission

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Prepared by
D. W. Akers, N. C. Kraft, J. W. Mandler

Idaho Engineering Laboratory
EG&G Idaho, Inc.
Idaho Falls, ID 83415

Prepared for
Division of Regulatory Applications
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
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ABSTRACT

As part of a study being performed for the Nuclear Regulatory Commission (NRC), small-scale waste-form specimens were collected during a low oxidation-state transition-metal ion (LOMI)-nitric permanganate (NP)-LOMI solidification performed in October 1989 at the Peach Bottom Atomic Power Station Unit 3. The purpose of this program was to evaluate the performance of cement-solidified decontamination waste to meet the low-level waste stability requirements defined in the NRC's "Technical Position on Waste Form," Revision 1. The samples were acquired and tested because little data have been obtained on the physical stability of actual cement-solidified decontamination ion-exchange resin waste forms and on the leachability of radionuclides and chelating agents from those waste forms. The Peach Bottom waste-form specimens were subjected to compressive strength, immersion, and leach testing in accordance with the NRC's "Technical Position on Waste Form," Revision 1.

Results of this study indicate that the specimens withstood the compression tests (>500 psi) before and after immersion testing and leaching, and that the leachability indexes for all radionuclides, including ^{14}C , ^{99}Tc , and ^{129}I , are well above the leachability index requirement of 6.0, required by the NRC's "Technical Position on Waste Form," Revision 1.

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EXECUTIVE SUMMARY

During light water reactor operation, the in-core irradiation of fuel rod cladding and other reactor structural surfaces and the subsequent corrosion of these components introduces activation products such as ^{54}Mn , ^{55}Fe , ^{60}Co , ^{63}Ni , and transuranics into the primary coolant. These activation products are transported by the primary coolant throughout the primary system. Some fraction of these activation products adheres to internal primary system surfaces and, over time, can result in the buildup of deposited activity and substantial radiation fields in the vicinity of these surfaces. In order to minimize occupational exposure during primary system maintenance and inspection activities, chemical decontamination methods are now commonly employed to remove activation products from primary system internal surfaces.

The low oxidation-state transition-metal ion-nitric permanganate-low oxidation-state transition-metal ion (LOMI-NP-LOMI) process is among the chemical processes most frequently used to decontaminate primary system components. A principal reagent used in this process is the chelating agent, picolinic acid. Chelating agents are used in reactor system decontamination formulas because they form strong complexes with actinides, lanthanides, heavy metals, and transition metals and help them to stay in solution. These chemical decontamination solutions, once used, are treated with ion-exchange resins to extract soluble metals and chemicals; these resins constitute the final waste to be processed and disposed from the decontamination process.

The resin wastes contain quantities of chelates or complexing agents in addition to inventories of radioactive corrosion products. A potential problem with the chelated decontamination wastes is the potential for increased solubility of the organo-radionuclide complexes in groundwaters at low-level-waste repositories.

The purpose of this research study is to evaluate the stability of actual low-level radioactive

ion-exchange resin wastes solidified in cement using the tests from the U.S. Nuclear Regulatory Commission's "Technical Position on Waste Form," Revision 1, for compression testing and leachability and to determine the release rates of radionuclides, chelating agents, and transition metals from solidified ion-exchange resin wastes that have been immersed in deionized water.

In this study, untreated ion-exchange resin wastes and small-scale Portland Type I-P waste-form specimens were collected from solidification vessel's during a solidification at the Peach Bottom Atomic Power Station Unit 3 (Peach Bottom-3). LOMI-NP-LOMI decontaminations were performed on the primary coolant recirculation system and on the reactor water cleanup system at Peach Bottom-3 during late December 1987 and early January 1988. The ion-exchange resins generated as part of this process were solidified in cement in 1989.

Two tests were performed to evaluate waste form structural stability and leachability; the ASTM C39 compression testing procedure was used to assess structural stability, and the ANSI/ANSI 16.1 leach test procedure was used to assess leachability. Compression tests were performed both before and after the 7-day immersion testing and after the 90-day leach test as specified and described in Appendix A of the "Technical Position on Waste Form," Revision 1.

During the leach test that was performed according to the ANSI/ANSI 16.1 standard, measurements were performed for pH and conductivity of the leachate; for the concentrations of radionuclides, stable metals, and picolinic acid in the waste form; and for the releases of these constituents from the waste form into the leachant. The releases to the leachant are quantified in terms of the absolute and fractional release rates, cumulative fractional release, effective diffusivity, and leachability index.

The primary conclusions of this study relate to the structural stability of the Peach Bottom-3 waste form and the releases of radionuclides,

stable metals, and the chelating agent (picolinic acid) from the waste form. In this study, comparisons have been made with degraded waste forms such as the FitzPatrick waste form from a prior part of this study and with other LOMI waste form research.

Key conclusions of the compression tests performed to assess structural integrity are that the Peach Bottom-3 waste forms meet the requirements for waste form integrity identified in the "Technical Position on Waste Form," Revision 1 (500 psi or 3.4×10^3 kPa). Post-immersion, compression-testing results indicate that the compressive strengths ranged from 960 to 1,370 psi for all leachants with specimens tested in deionized water and seawater having compressive strengths near 1,350 psi. This is lower than the compressive strengths of laboratory specimens of solidified decontamination ion-exchange resin waste forms solidified using the LOMI process.

In the evaluation of the leach test results, the primary conclusion was that the Peach Bottom-3 waste form meets the leachability index requirements specified in the NRC's "Technical Position on Waste Form," Revision 1. In addition, pH effects on leachability and the characteristics of radionuclide and stable metal releases from the waste form were also assessed. The pH data suggest that the pH of the leachate is affected within a few hours and probably within a few minutes by the chemistry of the waste form. Other studies have indicated that the cement chemistry will control the leachate pH unless magnesium, a constituent of seawater, is present in the leachate in significant concentrations. It has been further suggested that the ion strength of seawater may be a more important parameter and have a greater effect on radionuclide solubilities in the leachate than the pH.

Primary radionuclides for which measurements were performed were ^{14}C , ^{55}Fe , ^{60}Co , ^{63}Ni , ^{99}Tc , ^{129}I , and transuranics. Iron-55, ^{60}Co , and ^{63}Ni are neutron activation products that are concentrated by the decontamination process and are of primary concern in this study. The results for ^{137}Cs , ^{99}Tc , and ^{129}I , fission products, have

been added for comparison purposes and because of the importance of possible releases of ^{129}I and ^{99}Tc to the environment.

The summed radionuclide content in the Peach Bottom-3 waste forms is 7.8 $\mu\text{Ci/g}$ of waste form. The primary decontamination radionuclides present in the resins based on their measured concentration are ^{54}Mn , ^{65}Zn , ^{60}Co , ^{55}Fe , ^{63}Ni , and ^{14}C . The summed activity of these radionuclides is 7.7 $\mu\text{Ci/g}$ waste form or about 98% of the total activity. Carbon-14 makes up about 58% of the total activity. The dominant decontamination radionuclides, ^{60}Co and ^{55}Fe , make up about 31% and 1.9% of the total activity in the resin waste, respectively. In contrast, the fission products ^{90}Sr , ^{99}Tc , ^{129}I , and ^{137}Cs collectively constitute about 0.3% of the total activity. The concentrations of the transuranic isotopes are also low and sum to a total of $1.6 \times 10^{-3} \mu\text{Ci/g}$ (0.02% of the total activity). Greater than 87% of the transuranic activity was ^{241}Pu .

Among the stable metals whose concentrations were measured in the resin wastes, the iron concentration was highest at 3,000 $\mu\text{g}/\text{gram}$ resin. This was followed by nickel and chromium. In addition, analyses were performed for sulfate and phosphate. Neither type of ion was detectable in the waste form or the resin samples.

A comparison of the leachability characteristics of picolinic acid from the Peach Bottom-3 and decomposed FitzPatrick samples indicates that the average absolute and fractional release rates of picolinic acid for both waste forms are similar. The similarity of the release rates of the picolinic acid from the two waste forms (one that remained intact and one that disintegrated) indicates that structural integrity is not a factor in releases from the waste form. These data suggest that chemical mechanisms either in the waste form or in the resin itself control the release rate from the waste form and that compressive strength does not affect retention in the waste form.

A comparison of the absolute and fractional release rate data for ^{55}Fe , ^{60}Co , and ^{63}Ni , the three decontamination radionuclides from the

Peach Bottom-3 and FitzPatrick samples, indicates that the waste form structural stability does not appear to affect releases from the waste form. The average fractional release rate of ^{63}Ni from the Peach Bottom-3 samples is higher than that for the other decontamination radionuclides and indicates that this radionuclide is released at a higher rate than the others. The higher fractional release rates associated with ^{63}Ni may be due to the increased stability of nickel complexes, as the Irving-Williams correlation indicates that the stability of transition metal complexes fall in the order $\text{Ni}^{\text{II}} > \text{Co}^{\text{II}} > \text{Fe}^{\text{II}}$. These data suggest that the stability of the nickel complex with a chelating agent may result in the higher average fractional release rates of ^{63}Ni relative to the other transition metals.

The best value cumulative fractional release (CFR) for all radionuclides ranged from 8.8×10^{-5} (^{14}C) to 7.7×10^{-2} (^{129}I). The best value CFR for ^{55}Fe was 1.42×10^{-3} and is similar to the CFR for the radionuclide for the degraded FitzPatrick sample and is similar to that for ^{60}Co . In contrast to these data, the CFR for ^{63}Ni was 5.9×10^{-3} which is about a factor of three to four higher than the other two radionuclides. These data suggest that ^{55}Fe and ^{60}Co exhibit similar cumulative release behavior that is different than the releases of ^{63}Ni .

Other radionuclides for which analyses of the waste form and chelating agents were performed and measurable results were obtained were ^{14}C , ^{99}Tc , ^{129}I , ^{90}Sr , and ^{241}Pu . Carbon-14, ^{99}Tc , and ^{129}I were detectable in most samples, whereas ^{90}Sr was detectable in only a few of the leachate samples, and ^{241}Pu was not detectable in the leachates. The weighted average fractional release rates of ^{14}C are the lowest of any of the radionuclides measured. This is consistent with the results of Krishnamoorthy, who attributes the low release rate of ^{14}C measured in his study to the formation of insoluble hydrates and carbonates, which slow the release of this radionuclide. Further, he suggests that the fractional release rate of ^{60}Co should be slower than ^{14}C . This is inconsistent with our results, in which ^{60}Co is released at

a weighted average fractional release rate 20 times faster than ^{14}C .

This weighted average fractional release rate of ^{99}Tc is statistically the same as that for ^{55}Fe and ^{60}Co and suggests similar chemical and release rate behavior for ^{99}Tc . This might be expected because technetium is a metal and would be expected to form complexes with organic compounds such as chelating agents.

The fractional release rates of iodine and cesium are the highest of all radionuclides present in the waste form and are statistically the same as that for picolinic acid ($5.8 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$), which suggests that the maximum diffusional fractional release rate from the waste form is between $5 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$ and $1 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$.

Evaluation of the release rate data for the stable metals indicates that iron and chromium have similar release rate characteristics and that the release rates (absolute and fractional) of these metals from the intact Peach Bottom-3 waste form are similar to those from the degraded FitzPatrick waste form. These data further confirm the fact that the release of nickel and other elements are not dependant on waste form structural stability. In addition, the average fractional release rate of nickel ($9 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$) is statistically the same as that for picolinic acid. These data suggest that the release of nickel may not be controlled by the release of picolinic acid as it is released at a similar rate to the picolinic acid.

In contrast to nickel, the average fractional release rate of iron ($6.4 \times 10^{-13} \text{ cm}^2 \cdot \text{s}^{-1}$) is about a factor of 100 less than the weighted average fractional release of ^{55}Fe ($6.8 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$) and indicates that ^{55}Fe is released faster than elemental iron from the waste form. A comparison of the average and diffusion-driven fractional release rates indicates that ^{55}Fe release rate is higher during the initial leach periods, whereas the iron release rates are relatively similar throughout the leach test. These findings suggest that ^{55}Fe is in a different chemical form than the elemental iron; however, kinetics suggest that ^{55}Fe should be equilibrium with whatever

chemical form the elemental iron is in. No explanation for this behavior is apparent.

Absolute release rates for iron, nickel, and chromium are within a factor of four for all elements. These data and the fractional release rate data, which indicate a low fractional release rate for iron, suggest that the release of these elements from the waste form is not dependent on the inventory in the waste form and that other chemical mechanisms may be controlling factors for releases of stable metals.

The CFRs for nickel (2.9×10^{-2}) and chromium (1.6×10^{-2}) are within a factor of two, whereas the CFR for iron (4.6×10^{-5}) is about 0.1% of the CFR for nickel. These data indicate that the iron is retained in the waste form to a much greater extent than the other metals, which may be due to the greater complexing capability of nickel and the other metals. These data suggest that a complexing effect is present, which enhances the release of nickel and possibly chromium.

The concentrations of radionuclides, chelating agents, and stable metals in the liner were calculated as Ci/liner for radionuclides or kg/liner of stable metals or chelates. The summed radionuclide content of the Peach Bottom-3 liner is 56 Ci. The primary decontamination radionuclides present in the liner, based on their measured concentrations, are ^{54}Mn , ^{65}Zn , ^{60}Co , ^{55}Fe , ^{63}Ni , and ^{14}C . The summed activity of these radionuclides is 55 Ci or about 98% of the total activity.

To assess possible radionuclide releases from the liner, the order of the leachability indexes are summarized below from lowest to highest: $^{129}\text{I} > ^{137}\text{Cs} > ^{90}\text{Sr} > ^{63}\text{Ni} > ^{99}\text{Tc} > ^{60}\text{Co} > ^{55}\text{Fe} > ^{14}\text{C}$. As expected, the cations and anions had the lowest leachability indexes (8-9), and ^{14}C had the highest.

Comparisons of the releases from the Peach Bottom-3 LOMI waste forms that have been leached as part of this study and LOMI waste forms leached as part of previous studies indicate that the inventory of picolinic acid in the waste

form does not appear to affect the fractional release rate. The inventory of picolinic acid in an Indian Point sample was an order of magnitude less than the other waste forms and yet the fractional release rate was greater than those for FitzPatrick and Peach Bottom-3. However, it should be noted that although all samples were solidified using the LOMI process, the formulations used for Indian Point and FitzPatrick were considerably different than those used for Peach Bottom-3. The fact that the CFR of picolinic acid for Peach Bottom-3 is considerably lower than that observed for other waste forms suggests that recent changes in the formulation of the LOMI waste form may have improved the leaching properties of the waste form.

In summary, the key conclusion from the Peach Bottom-3 study is that the leachability indexes of all radionuclides meet the requirements of the NRC's "Technical Position on Waste Form" Revision 1. Other primary conclusions are that the release rates of radionuclides, stable metal, and chelating agents do not appear, in general, to be affected by the structural stability of the waste form. These data suggest that waste form compression testing is of limited value in assessing actual waste form stability. Other key points are that the apparent higher release rate for ^{63}Ni may be due to greater stability of complexes formed by this radionuclide and further suggests that there are chelant effects on the transition metals. These effects are less apparent for ^{55}Fe and ^{60}Co , which form complexes that are less stable than those formed by ^{63}Ni . Also, the release rate behavior of ^{99}Tc is similar to that of ^{55}Fe and ^{60}Co and suggests similar chemistry for this radionuclide.

Another principal observation is that ^{14}C , which had the highest inventory (58%) of any radionuclide in the waste form, had the lowest leachability index, and indicates that this radionuclide is strongly retained in the cement matrix, probably as an insoluble hydrate or carbonate. In contrast, ^{129}I , a mobile anion, is released at rates similar to those for ^{137}Cs and has the lowest leachability index (8.4) or the highest release rate of all radionuclides measured.

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ACRONYMS

ANSI/ANS	American National Standards Institute/American Nuclear Society	INEL	Idaho National Engineering Laboratory
AP	alkaline permanganate	ISO	International Standards Organization
ASTM	American Society for Testing and Materials	LOMI	low oxidation-state transition-metal ion
CFR	Code of Federal Regulations	NP	nitric permanganate
CFR	cumulative fractional release	NRC	U.S. Nuclear Regulatory Commission
CNSI	Chem Nuclear Systems Incorporated	PCP	process control program
HPGe	hyperpure germanium	Peach Bottom-3	Peach Bottom Atomic Power Station Unit 3
ICP-AES	inductively coupled plasma-atomic emission spectroscopy	TSP	trisodium phosphate

Release of Radionuclides and Chelating Agents from Cement-Solidified Decontamination Low-Level Radioactive Waste Collected from the Peach Bottom Atomic Power Station Unit 3

INTRODUCTION

During light water reactor operation, the in-core irradiation of fuel rod cladding and other reactor structural surfaces, and the subsequent corrosion of these components introduces activation products such as ^{54}Mn , ^{55}Fe , ^{60}Co , and ^{63}Ni into the primary coolant. These activation products are transported by the primary coolant throughout the primary system. Some fraction of these activation products adheres to internal primary system surfaces and, over time, can result in the buildup of deposited activity and substantial radiation fields in the vicinity of these surfaces. In order to minimize occupational exposure during primary system maintenance and inspection activities, chemical decontamination methods are now commonly employed to remove activation products from primary system internal surfaces.

The LOMI-NP-LOMI process is among the chemical processes most frequently used to decontaminate primary system components. This process is described in Appendix A. A principal reagent used in this process is the chelating agent, picolinic acid. Chelating agents are used in reactor system decontamination formulas because they form strong complexes with actinides, lanthanides, heavy metals, and transition metals, and help them to stay in solution. These chemical decontamination solutions, once used, are treated with ion-exchange resins to extract soluble metals and chemicals; these resins constitute the final waste to be processed and disposed from the decontamination process. The resin wastes contain quantities of chelates or complexing agents in addition to inventories of radioactive corrosion products and lesser inventories of fission products. A potential problem with the chelated decontamination wastes is the potential for increased solubility of the organo-radionuclide

complexes in groundwaters at low-level waste repositories.

The U.S. Nuclear Regulatory Commission (NRC) is concerned with the safe disposal of these chemical decontamination wastes and has made provisions for their disposal in "Licensing Requirements for Land Disposal of Radioactive Waste" (U.S. Code of Federal Regulations Standard 10, Part 61). Section 61.56 provides requirements for the stability of waste forms that must be met for the waste form to be acceptable for near-surface disposal. Additional requirements for the disposal of chelated wastes are also given in burial site regulations. In Section 61.54, the wastes are classified as Class A, B, or C, based on the concentrations of radionuclides in the wastes. Class A wastes have lower concentrations and may be disposed of without stabilization; however, Class A wastes buried with Class B and C wastes must be stabilized. Class B and C wastes must be structurally stabilized to ensure that the waste form does not degrade and does not promote slumping, collapse, or failure of the cap or cover of the near-surface disposal trench. In addition, the stability of the waste form limits exposure to inadvertent intruders. Class B and C low-level wastes from light water reactors (LWRs) may be solidified at LWR sites using cement to meet the stability requirements of the NRC's "Technical Position on Waste Form," Revision 1. Solidification of these wastes is intended to provide the structural stability needed to ensure that no collapse of the disposal trench occurs and that the release of radionuclides via leaching is minimized.

Test procedures to demonstrate waste form stability and to quantify leachability for the low-level wastes from LWRs have been specified by

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the NRC in the Low-Level Waste Management Branch's "Technical Position on Waste Form,"¹ published in 1983, and in the "Technical Position on Waste Form," Revision 1,² published in January 1991. Both revisions of the Technical Position specify that small-scale waste-form specimens must be prepared and tested to certify the stability of the full-scale waste form. A range of tests must be performed that include compression testing and leach testing of small-scale waste-form specimens.

The NRC's "Technical Position on Waste Form," Revision 1, stipulates that the small-scale waste-form specimen, after having been immersed in deionized water for a minimum of 90 days, should have a compressive strength of at least 500 psi (3.4×10^3 kPa) when tested in accordance with the American Society for Testing and Materials (ASTM) Standard C39,³ "Compressive Strength of Cylindrical Concrete Specimens." In addition, the "Technical Position on Waste Form," Revision 1, stipulates that the small-scale waste-form specimen should have a leachability index greater than 6 when leach-tested for a minimum of 5 days in accordance with the American National Standards Institute/American Nuclear Society (ANSI/ANS) Standard 16.1, "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure."⁴

The purpose of this research study is to evaluate the stability of actual low-level radioactive ion-exchange wastes solidified in cement using the tests from the "Technical Position on Waste Form," Revision 1, for compression testing and leachability, and to determine the release rates of radionuclides, chelating agents, and transition metals from solidified ion-exchange resin wastes that have been immersed in deionized water. This work is identified in the NRC's Low-Level Radioactive Waste Research Program Plan,⁵ which defines a strategy for conducting research on issues of concern to the NRC in its efforts to ensure stability of solidified low-level radioactive waste, leading to safe disposal.

The leachability of decontamination ion-exchange resin waste forms that contain chelating agents is being evaluated because the leachability of these waste forms is a function of a number of factors, including the chemical characteristics of the radionuclide being leached, resin waste chemistry, solidification agent, chelating agent, and the final solidified matrix. Burial site hydrology and groundwater chemistry also influence leaching rates, as do cyclic wet and dry conditions. The complexity of the interactions that occur among radionuclides, chelating agents, groundwater, and soil introduces uncertainties into the models used to predict the impact of the decontamination resin wastes on the performance of shallow-land burial sites. Therefore, it is important to establish a data base on the stability and leachability of decontamination resin waste forms representative of waste forms commonly generated at operating commercial power stations in leachants that are expected to be representative or more aggressive than the actual disposal site groundwaters. Deionized water is considered to be an acceptable aggressive leachant as discussed in Appendix A of the "Technical Position on Waste Form," Revision 1.

The results of numerous investigations reported in the literature indicate that the rate of radionuclide release from a decontamination ion-exchange resin waste form is influenced by poorly understood interactions related to the properties of the solid and the leaching system. Further, waste forms of this type have been found to not solidify as expected or to produce poor waste forms. Among the factors that are known to influence the leachability of cement-solidified waste forms are the chemical composition of the cement used, the waste-to-binder ratio, the amount of water used to set the cement, and the presence of additives that are used to accelerate or retard cement hydration. Other factors such as temperature, leachant composition, pH, volume, and residence time may also influence leachability. Solidification, which did occur as expected, and the factors affecting solidification were discussed in depth at the Workshop on Cement Stabilization of Low-Level Radioactive Waste.⁶ Specific process details and problem wastes were

reviewed. One of the conclusions of this conference was to revise the solidification formulation to improve the stability of the waste form. As will be discussed, the Peach Bottom Atomic Power Station Unit 3 (Peach Bottom-3) waste forms leached as part of this study were some of the first wastes stabilized using improved formulations, and this study was conducted to assess the effectiveness of this improved formulation.

A number of studies,⁷⁻¹⁷ as shown in Table 1, have been performed, which address solidified resins or decontamination solutions. Results of these studies are used for comparison purposes or to assist in the interpretation of the results of this study. Common to these studies is an emphasis on more realistic leaching situations involving local groundwater,^{7,10,12,17} seawater,^{12,17} and actual nuclear power plant resin wastes.^{8,9,10,12,17} In these studies, solidified ion-exchange resin specimens were subjected to leach tests following either the ISO 6961 leach-test procedure¹⁸ or the ANSI/ANS 16.1 leach-test procedure. Both of these methods are nonequilibrium tests in the respect that the solidified waste-form specimen is completely immersed over an extended period of time in a large volume of leachate that is periodically replaced with new leachant. Other more recent studies¹⁹⁻²³ performed using various waste types have suggested that leachant effects, limited solubility, and breakdown of organo-radionuclide complexes may limit the effects of chelating agents on releases from waste forms.

Differences in leaching behavior have been observed in the studies that appear to be a result of changes in leachant composition. A study of the

effects of seawater on leach rates of non-resin wastes by Fuhrman and Colombo²⁴ indicates that release rates were a factor of 10 less than those observed for deionized water. The slower release rates are thought to occur because of the much higher ionic strength of the seawater leachant and cement/waste-leachant reactions. However, studies at the Idaho National Engineering Laboratory (INEL) (Reference 17) indicate that the effect of seawater on radionuclide releases from solidified decontamination ion-exchange resins varies and may be similar to groundwaters.

The present study is a continuation of studies^{12,17,25} previously performed at the INEL for the NRC that measured the compressive strength and leachability of cement-solidified evaporator concentrates and decontamination ion-exchange bead resin waste forms leached in deionized water, groundwaters, and seawater. The studies at the INEL are the only continuing studies in which actual commercial nuclear power plant decontamination radioactive waste is being evaluated.

In the current study, untreated ion-exchange resin wastes and small-scale Portland Type I-P (pozzolanic) waste-form specimens were collected from solidification vessels during a solidification at Peach Bottom-3. LOMI-NP-LOMI decontaminations were performed on the primary coolant recirculation system and the reactor water cleanup system at Peach Bottom-3 during late December 1987 and early January 1988. A description of these decontaminations is presented in References 26 and 27, and a summary of the waste solidification and sampling process is presented in Appendix B.

Introduction

Table 1. Summary of recent cement-solidified resin waste leach studies.

Author	Reference number	Sample size	Leachant	Solidification	
				Agent(s)	Resin type
Criscienti, L. J. Serne, R. J.	7	2.5 cm × 5.0 cm (diameter × length)	Hanford groundwater	Portland cement type III	BWR evaporator concentrates and ion-exchange resins
Howard, C. G. Jolliffe, C. B. Lee, D. J.	8,9	28 mm × 28 mm (diameter × length)	Demineralized water	Portland cement, blast furnace slag, microsilica	LOMI ion-exchange resin DOWEX 50X8 ^{60}Co , ^{137}Cs
Ipatti, A. Harkonen, H.	10	Small lab-scale specimens	Groundwater	Cement	Spent ion-exchange resins from Loviisa Power Plant
Torstenfelt, B. Hedin, G.	11	Not specified	Not specified	Sulfate resistant Portland cement	Powdered ion-exchange resins with 5 wt.% zeolite
McIsaac C. J. W. Mandler	12	5 cm × 10 cm (diameter × length)	Groundwater and seawater	Portland cement Type I-P	Decontamination ion-exchange resins IRN-77, IRN-78, C-100-H, and A-600
Bishop, J. V.	13	2 in. × 4 in. (diameter × length)	Compression test only	Portland cement Lime and NaOH	Ion-exchange resins Ionac A-365 and C-267 with picolinic acid only
Davis, M. S. Picuilo, P. L. et al.	14	2 m × 4 m (diameter × length)	Demineralized water	Portland cement Type I and VES	Ion-exchange resins IRN-77, IRN-78, Ionac-365 with chelates only
Lee, J. O. Han, K. W. Buckley, L. P.	15	Not specified	Demineralized water	Cement, polyester resin and Zeoloin 900-Na	Ion-exchange resins, Li ⁺ -DH form, ^{60}Co , ^{85}Sr , and ^{137}Cs
Soo, P.	16	5 cm × 10 cm 15 cm × 15 cm 30 cm × 30 cm (diameter × length)	Deionized water Formate and pico-linate solutions	Portland cement Type I and VES	Simulated decontaminated resin waste
McIsaac et al.	17	5 × 10 cm	Deionized water groundwater, seawater	Portland cement Type I-P	Power plant decontamination ion-exchange resin wastes

EXPERIMENTAL PROCEDURES

This section summarizes the experimental procedures used in this study. Many are specific to the characterization of commercial reactor waste streams and cement solidified decontamination ion-exchange resin waste forms. Included are the methods used to collect the untreated resin waste and waste-form specimens, procedures used to test the waste-form specimens for compressive strength and leachability, and the analytical methods used to determine the concentrations of radionuclides, metals, and chelating agents in the untreated resin waste and leachate samples.

Sample Collection

Resin wastes and waste-form specimens were collected at Peach Bottom-3, which is operated by the Philadelphia Electric Company, from October 17 through October 25, 1989. The solidification was performed on decontamination ion-exchange resins from a LOMI-NP-LOMI decontamination performed by Pacific Nuclear Services (References 25 and 26). The Peach Bottom-3 primary coolant recirculation system and the reactor water cleanup system were decontaminated during late December 1987 and early January 1988, and the resins were held until October 1989 for solidification.

The ion-exchange resins used to process the spent LOMI decontamination reagents and the corrosion products removed from internal sur-

faces of the primary coolant recirculation system and the reactor water cleanup system were Ionac A-365 weak base anion resin and Ionac C-267 strong acid cation resin, which are manufactured by Sybron Chemicals, Inc., of Birmingham, New Jersey. About 160 ft³ of anion resin and about 136 ft³ of cation resin were used to process the LOMI decontamination slurry. During January and August 1988, the ion-exchange resins were sluiced to three separate liners. A solidification was attempted in December 1988; however, the process control program (PCP) samples failed quality assurance tests, and the solidification was delayed. Information on the decontamination is contained in References 26-28. As will be discussed later, personnel at Chem Nuclear Systems Incorporated (CNSI) believe that an enhanced exotherm during the initial stages of the solidification process may result in an improper solidification and in poor waste form characteristics.

Following an attempted solidification in December 1988, about 7 ft³ of resin was transferred from liner 446828-15 to liner 446692-1 in order to more evenly distribute the resin among the three liners to ensure that the resin loading was more equal for the three liners and thereby reduce the waste-to-cement ratio. All three liners contained mixtures of anion and cation resins. Table 2 presents the types and quantities of ion-exchange resins in the three solidification liners as of October 1989 when CNSI solidified the waste.

Table 2. Compositions of ion-exchange resin wastes in the three liners solidified at Peach Bottom-3.

Liner #	Anion resin		Cation resin		Picolinic acid loading (wt%)
	(ft ³)	(m ³)	(ft ³)	(m ³)	
446692-1	45.6	1.29	45.6	1.29	1.8
446828-10	59.5	1.68	33.3	0.94	4.5
446828-15	56.0	1.58	57.0	1.61	5.5

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By October 1989, personnel at CNSI had developed and tested a revised solidification formula that they felt confident would work in solidifying the LOMI decontamination ion-exchange resin wastes being stored at the Peach Bottom Station. EG&G Idaho personnel requested that liner 446828-15 be solidified first. Since it contained the highest concentration of picolinic acid and activation metals, for leach testing purposes, it was the best candidate of the three liners for sampling. Table 3 lists the contents of liner 446828-15.

The initial PCP tests indicated that samples prepared using smaller quantities of cement than the amount used to prepare PCP samples during initial tests in December 1987 failed to solidify and that in addition, a strong odor of ammonia was noticed near the sample that failed to solidify. As discussed in Appendix B, additional PCP samples were prepared with more trisodium phosphate than those prepared previously. The samples with the revised formulation were hard when examined the following day and, based on this information, it was determined that the solidification could proceed.

During the solidification of liner 446828-15, after the fill-head was removed from the top of the liner, the resin/cement mixture inside the liner

was sampled under EG&G Idaho supervision. The sampling tool, which was simply a plastic tube equipped with a plunger, was inserted into the resin/cement mixture five or six times to a depth of about 3 ft below the top surface of the mixture. Following each insertion, the material inside the tube was transferred to a plastic-lined bucket. About 2 gal of resin/cement mixture was collected. The resin/cement mixture was quite fluid, which made it possible to pour the material into the molds that were used to prepare waste-form specimens. Between 2:35 and 3:05 p.m. on October 24, 19 waste-form samples 2 in. (5 cm) in diameter and 4 in. (10 cm) long were prepared. Individual samples had contact exposure rates of about 100 mR/hour. The sample molds were sealed immediately after they were filled, and at 6:30 p.m. on October 24, they were all placed in an oven that was maintained at 145°F to simulate the hydration exotherm.

Three of the waste-form specimens were removed from the oven the following morning and were examined. All of the samples that were removed had some free-standing water on their top surfaces. They felt firm but were certainly not yet hard. These samples were then returned to the oven, and all 19 specimens were baked at 145°F for a total time of about 72 hours.

Table 3. Quantities of materials in Peach Bottom-3 liner 446828-15.

Type of material	Volume		Mass	
	(ft ³)	(m ³)	(lb)	(kg)
Ionac A-365 anion resin	56.0	1.58	3,749	1,701
Ionac C-267 cation resin	57.0	1.61	3,818	1,732
Trisodium phosphate	4.8	0.14	550	249
Flyash	19.8	0.56	2,730	1,238
Portland Type I-P cement	28.78	0.81	4,982	2,260
Total:	166.4	4.7	15,829	7,180

All three liners were solidified during the solidification campaign; however, the week after the solidification, the liners were visually inspected and a broom handle test was performed to assess the strength of the waste form. (A broom handle or shaft of a similar diameter is pressed onto the surface of the waste form to assess whether it has hardened sufficiently to prevent penetration into the cement.) Both 446692-1 and 446828-10 were solid; however, 446828-15 was only solidified on the surface. A thin crust had formed over the top surface of the resin/cement mixture, but below the crust, the material had not yet solidified. CNSI personnel suggested that ammonia in the resin/cement mixture was retarding the setting of the cement. They recommended that the liner be vented to help remove the ammonia. Plant personnel followed the recommendation and began venting the liner the week of October 30. The liner was not examined again until December 21, 1989. On that date, the upper surface of the monolith was again probed with a broom handle, and this time, the broom handle did not penetrate the surface.

Potential problems with the LOMI decontamination process were evaluated by CNSI personnel,²⁸ and they concluded that several factors may be contributing to problems with the solidification of LOMI wastes. They believed that the presence of calcium ions from the lime used to adjust the pH of the waste may have caused the exothermic hydration process²⁹ to begin earlier than expected and could have resulted in higher temperatures in the liner when the cement was added, thereby causing a poor solidification. To control the exothermic process, CNSI proposed that the temperature of the liner be monitored during the pH adjustment phase and that the liner be allowed to cool before cement was added because the cement would further increase the temperature (Reference 28).

In addition, CNSI suggested that a possible cause of the problems with LOMI solidifications may also be due to the switch to Ionac A-365 anion resin (Reference 28), which is reported to have a greater capacity for picolinic acid and consequently, more calcium, if the picolinic acid

degrades during the pH adjustment process, thereby leaving additional reaction sites for calcium ions. Hydration of the calcium would generate heat that would be localized at the ion-exchange resin bead and possibly cause generation of ammonia due to the degradation of the resin (a tertiary amine). An industry survey³⁰ was performed to assess the degradation mechanism. Correspondence with Sybron Chemicals,³¹ the manufacturer of Ionac A-365, and a review of the ion-exchange resin chemistry by this laboratory³² indicates that these resins are sensitive to elevated temperatures and that they do degrade over their service life.

Whether the presence of ammonia would be expected to retard solidification has been evaluated,^{30,31} and this evaluation indicates that low concentrations of various amine compounds will not retard solidification. However, other reports^{6,32} indicate that ammonia compounds such as ammonium chlorides are strong retarding agents and will inhibit solidification. In any event, solidification of the vessel containing 5.5 wt% picolinic acid did not occur immediately, and the probable reasons suggested by CNSI are the hydration of calcium released during the pH adjustment period or possibly the presence of ammonia caused by degradation of the resins.

Leach-Test Method

The test procedure used to measure the release of radionuclides, transition metals, and chelating agents from decontamination resin wastes solidified in Portland Type I-P cement was ANSI/ANS 16.1. This standard is intended to provide a means of quantifying the release of radionuclides from waste forms using the results of relatively short-term tests performed in a laboratory. It is not intended to serve as a representative test for long-term leaching behavior of waste forms under conditions representing actual burial conditions. The method specified by the standard for analyzing leach test data is based on the assumption that diffusion is the only release mechanism. Other mechanisms such as dissolution, ion exchange, corrosion, cracking, etc., are not incorporated into the models used to describe releases. Although the adequacy of this assumption has been ques-

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tioned,³³ the test procedure is, however, believed suitable^{2,34} to establish a data base on the release of radionuclides in the presence of relatively high concentrations of chelating agents and transition metals.

Test Procedure

The three waste-form specimens obtained from Peach Bottom-3 that were leach tested were right-circular cylindrical solids with nominal dimensions of 1.75 in. (4.4 cm) in diameter by 3.5 in. (8.9 cm) long. The actual dimensions of each sample and the curing times are shown in Table 4. A photograph of a specimen typical of the mixed-bed resin waste forms collected from Peach Bottom-3 is shown in Figure 1. Figure 1 shows that the mixed-bed resin waste-form specimens prior to being leached were solid right-circular cylinders having smooth surfaces with few surface imperfections and no cracks. All samples

were leached in cylindrical polyethylene containers having capacities of about 3 L. Figure 2 shows a representative waste-form specimen suspended in a leach-test vessel. In all cases, the waste-form specimen was supported in the leach-test vessel by coarse mesh plastic netting that was suspended by a wire from the container lid. The leachant used for this study was deionized water having a conductivity of less than 3 $\mu\text{mho}/\text{cm}$ at 298 K (25°C).

The percentage of dry resin shown in Table 4 was calculated as the ratio of the weight of free interstitial water to the weight of cement, trisodium phosphate, and flyash used during the solidification of the full-scale waste form from which the waste-form specimen was collected. This percentage was calculated using the method developed by Neilson,³⁵ which divides the water content of the resins between the water content of

Table 4. Physical parameters of ion-exchange resin waste-form specimens collected from Peach Bottom-3.^a

Sample characteristic	Sample identification		
	#8	#4	#12
Diameter by height (cm)	4.7 × 9.3	4.4 × 8.9	4.4 × 8.9
Weight (g)	246.3	248.1	244.8
Surface area (cm ²)	137.2	123.0	123.0
Volume (cm ³)	161.3	135.2	135.2
Volume-to-surface ratio	1.18	1.10	1.10
Resin loading ^b (vol%)	67.9	67.9	67.9
Resin loading ^b (wt%)	47.8	47.8	47.8
Dry resin loading (wt%)	14.8	14.8	14.8

a. Sample #8 was cured for 950 days prior to leach testing, and samples #4 and #12 were cured for 1,304 days prior to leach testing. Compression tests were performed near the earlier time.

b. Loadings determined based on liner composition.



Figure 1. Peach Bottom leach test specimen.

the "as-received" and dewatered ion-exchange resins to determine the amount of available interstitial water. In addition, there is typically a nominal 1 to 2 in. (2.5 to 5 cm) of free standing water above the resin in the liner bed. This quantity was not included in the calculation because the actual level is not known.

As required by ANSI/ANS 16.1, the ratio of leachant volume to the geometric surface area of the specimen was kept constant at a ratio of at least 10:1. The actual volumes of leach solution used were about 1.7 L. This volume exceeds the 10:1 ratio because ANSI/ANS 16.1 requires that the sample must be surrounded by a leachant layer exceeding 10 cm in thickness or the minimum specimen dimension. In this case, the minimum specimen dimension was used to determine the leachant volume. The pH and conductivity of the leachates were measured at the end of each leaching period.

During leach-testing, the requirements established in ANSI/ANS 16.1 were followed. This

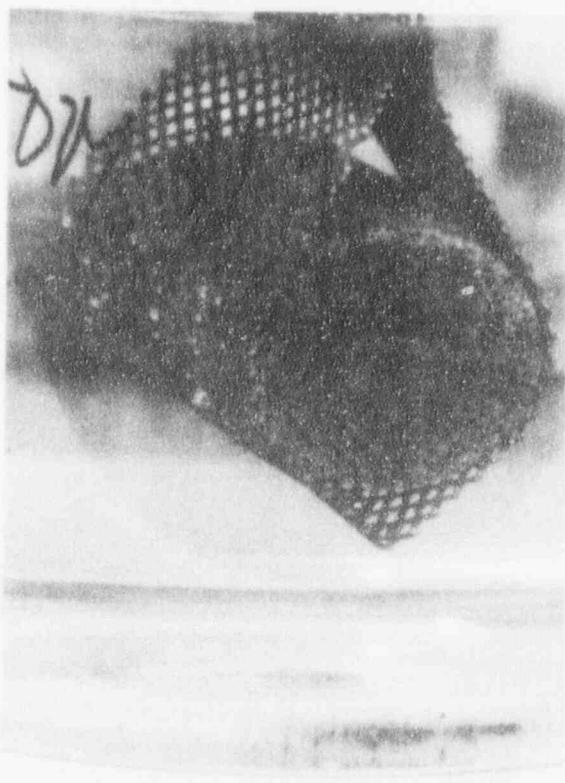


Figure 2. Peach Bottom leach test specimen suspended in leachant.

standard requires a minimum leach period of 5 days, which corresponds with the current requirement in the "Technical Position on Waste Form," Revision 1, for a 5-day leach test. During the period of the 5-day leach testing, the required ANSI/ANS 16.1 changeout schedule was maintained within several minutes, and the leachant temperature was maintained at $77 \pm 5^{\circ}\text{F}$ ($25 \pm 3^{\circ}\text{C}$).

Data Analysis

Leaching occurs as the result of mass transport of species both inside and from the surface of a waste form. Mass transport processes that have been identified as occurring in solidified waste forms during leaching include diffusion, dissolution, ion exchange, corrosion, and surface effects. A considerable amount of data that were obtained from samples that maintained their integrity during leaching indicate that internal bulk diffusion is the most likely rate-determining mechanism during the later phases of the leaching process (after 1–2 days).^{36–38} The methods used to assess

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leachability (i.e., fractional release, cumulative fractional release, and leachability index) are based on the premise that internal bulk diffusion is the rate-determining process and are predicated on an intact waste form.

For each waste-form specimen that was leach-tested, the initial inventory of each species in the waste form was determined by two methods. The first method was to multiply the measured concentration of the species in the untreated wet resin waste, expressed as $\mu\text{Ci/g}$ or $\mu\text{g/g}$, by the mass of water-saturated resin estimated to be in the waste-form specimen. The quantity of resin estimated to be in the waste form is based on the weight fraction of water and resin present in the resin/cement mixture in the solidification liner from which the specimen was obtained. This method is considered relatively accurate because the waste-form samples were obtained from the interior of liners that had been well mixed before sample collection. In addition, to confirm the analysis results for the resin samples, samples of the solidified waste form were analyzed to determine the actual radionuclide, metals, and picolinic acid content per gram of solidified waste. These measurements indicated that the resin loading was approximately 53 wt% based on the radionuclide content of the resin and waste form. This is similar to the resin loading (48 wt%) determined from Table 2. The difference is probably due to small irregularities in the loading of the cement. The 53 wt% value is probably more accurate.

Upon completion of the leach testing, the inventory information and the leach test data are used to determine the average absolute and fractional release rates of radionuclides, metals, and chelating agents from each waste-form specimen. The average absolute release rate is defined as the quantity released per unit surface area per second ($\mu\text{Ci/cm}^2 \cdot \text{s}$ or $\mu\text{g/cm}^2 \cdot \text{s}$), and the fractional release rate is defined as the fraction of the initial inventory released per unit surface area per second ($\text{cm}^2 \cdot \text{s}^{-1}$). The surface area used in this calculation is the external geometrical surface area of the intact waste form.

The cumulative fraction release (CFR) of radionuclides, metals, and chelating agents from the waste-form specimens is the sum of the individual fractional releases calculated as

$$\text{CFR} = \frac{\sum_{i=1}^n [(C_L)_i V_i]}{C_R M_R} \quad (1)$$

where

$(C_L)_i$ = the concentration of the species in the leachate collected following leaching interval i ($\mu\text{Ci/mL}$ or $\mu\text{g/mL}$)

V_i = the volume of leachate collected following leaching interval i (mL)

C_R = the concentration of the species in the resin waste ($\mu\text{Ci/g}$ or $\mu\text{g/g}$)

M_R = the mass of resin waste estimated to be in the waste form (g).

Uncertainties in the CFRs of selected radionuclides; metals; and citric, oxalic, and picolinic acids were calculated at the one-sigma confidence level using representative data for resin waste and leachate samples. Uncertainties in the absolute counting efficiencies of the hyperpure germanium (HPGe) spectrometers used to determine concentrations of gamma-emitting radionuclides were assumed to be $\pm 5\%$. The uncertainty in the volume of any given leachate sample was assumed to be $\pm 1\%$, and the uncertainty in the mass of resin waste in any given waste-form specimen was assumed to be $\pm 5\%$.

The ANSI/ANS 16.1 standard provides for the calculation of a leachability index, which is one of the measures used in the NRC's "Technical Position on Waste Form," Revision 1, to determine the acceptability of a waste form for disposal at a waste site. The leachability index, L , is based on the effective diffusivity of the species being leached from the waste form. When depletion of the diffusant is less than 20%, the effective diffusivity (D_n) is defined as

$$D_n = \pi [(a_n/A_0)/\Delta_n]^2 \times (V/S)^2 \times T \quad (2)$$

where

D_n = the effective diffusivity of a species during leaching interval n (cm^2/s)

a_n = the quantity of the species leached during leaching interval n (μCi or μg)

A_0 = the total amount of the species originally present in the waste-form specimen (μCi or μg)

Δ_n = the duration of leaching interval n (s)

V = the volume of the waste-form specimen (cm^3)

S = the external geometric surface area of the waste-form specimen (cm^2)

T = the leaching time representing the "mean time" of the n^{th} leaching interval (s).

As shown in Equation (2), effective diffusivity is proportional to the square of the ratio of specimen volume to surface area.

The leachability index is based on the effective diffusivity of the species of interest, and is also dependant on the estimated surface area of the debris. The leachability index, L, is defined as

$$L = \frac{1}{k} \sum_{n=1}^k [\log(b/D)]_n \quad (3)$$

where

b = defined constant ($1.0 \text{ cm}^2/\text{s}$)

D = effective diffusivity of the species (cm^2/s)

k = number of leaching intervals.

As shown in Equation (3), the leachability index of a particular diffusing species is the summation of the incremental leachability indexes for all leach intervals.

Compressive Strength Test Method

The test procedure used to measure the compressive strength of waste-form specimens was ASTM C39, "Compressive Strength of Cylindrical Concrete Specimens,"³ as required by the NRC in the "Technical Position on Waste Form."¹ The requirement is for a minimum compressive strength for waste forms of 50 psi ($3.4 \times 10^2 \text{ kPa}$). This requirement has been modified in Appendix A of the "Technical Position on Waste Form," Revision 1, to 500 psi ($3.4 \times 10^3 \text{ kPa}$) for cement-solidified waste forms. The revised higher value is deemed necessary to ensure that cement-solidified waste forms maintain integrity and exhibit long-term stability as required by 10 CFR 61.³⁴

The vice and load gauge used to measure compressive strength are shown in Figure 3. Each specimen was placed in the vice, and the load on the specimen was gradually increased until the load gauge registered a decrease in load, which occurred when the specimen began to fail. The maximum load that each waste-form specimen withstood was recorded and was used to compute the compressive strength of the specimen based on its cross-sectional area.

Analytical Methods

The data analysis methods of ANSI/ANS 16.1 require a knowledge of the initial inventories of diffusing species in the waste-form sample being tested and a knowledge of their concentrations in the leachates generated during leach testing. In order to provide a basis for estimating the initial inventories of species of interest, samples of unsolidified resin waste, solidified waste-form samples, and leachate samples were analyzed using several different analytical techniques.

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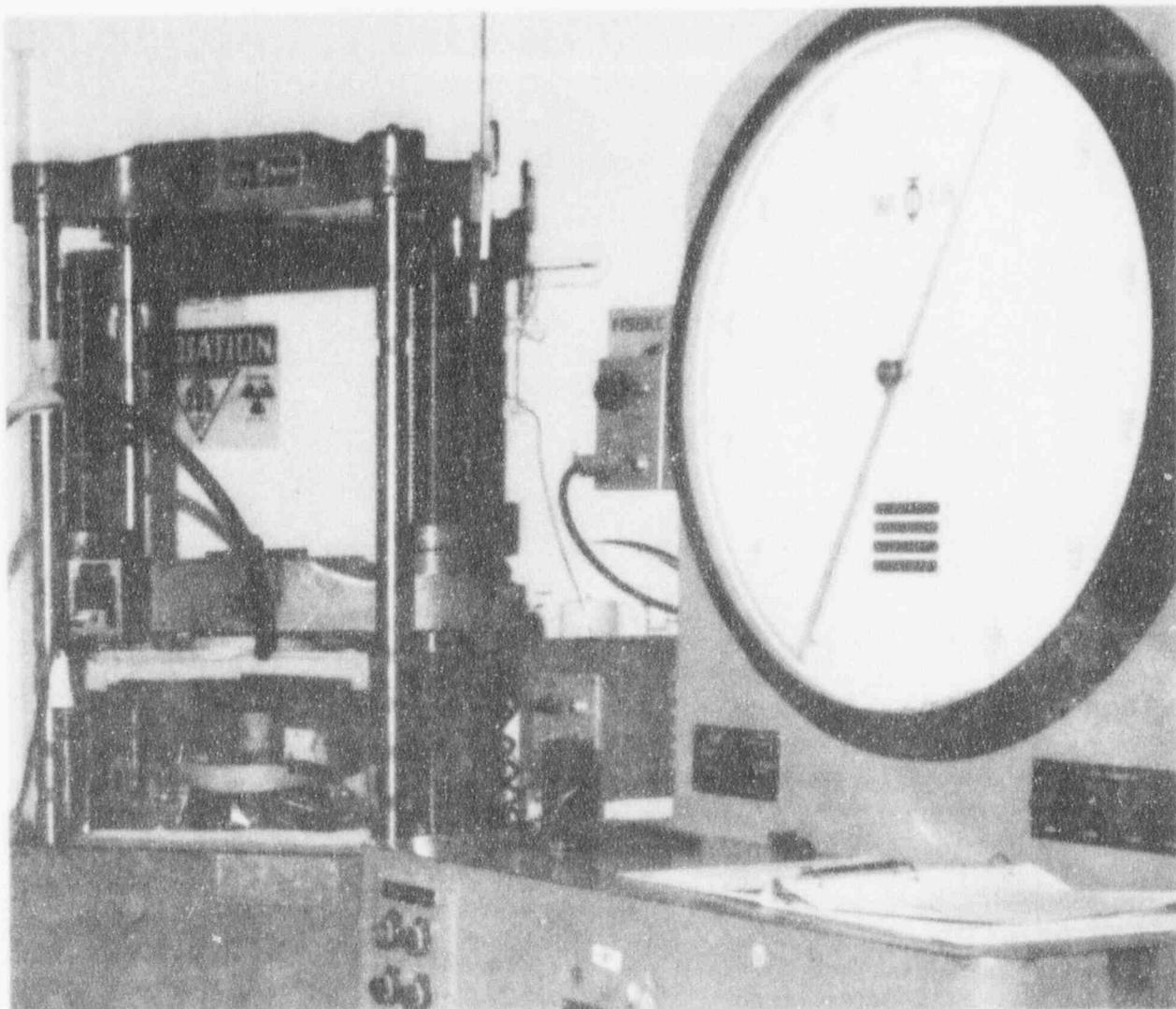


Figure 3. Compressive strength testing system.

Because of the differences in their physical characteristics and because the concentrations of radionuclides and metals in the leachates were expected to be as much as several orders of magnitude lower in the leachates compared to the resins, the methods required to prepare the resin and leachate samples for analysis were generally different.

Prior to analysis, leachate solutions were partitioned into volumetric samples according to the analysis scheme shown in Figure 4. The analytical methods used to determine concentrations of radionuclides, transition metals, and chelating agents in leachate samples included high-resolution gamma-ray spectrometry, liquid scintillation and gas proportional counting, inductively

coupled plasma-atomic emission spectroscopy, and ion chromatography. The detection of chelating agents in some leachate solutions required that the chelating agent be chemically separated from other components of the sample prior to ion chromatography analysis.

The following sections provide descriptions of the sample preparation and analysis methods used to analyze the resin waste and leachate samples obtained during the course of this study.

Radionuclide Analysis

Resin waste samples were analyzed for radionuclides specified in 10 CFR 61 (i.e., ^{14}C , ^{60}Co , ^{63}Ni , ^{90}Sr , ^{99}Tc , ^{129}I , ^{137}Cs , ^{238}Pu , ^{239}Pu ,

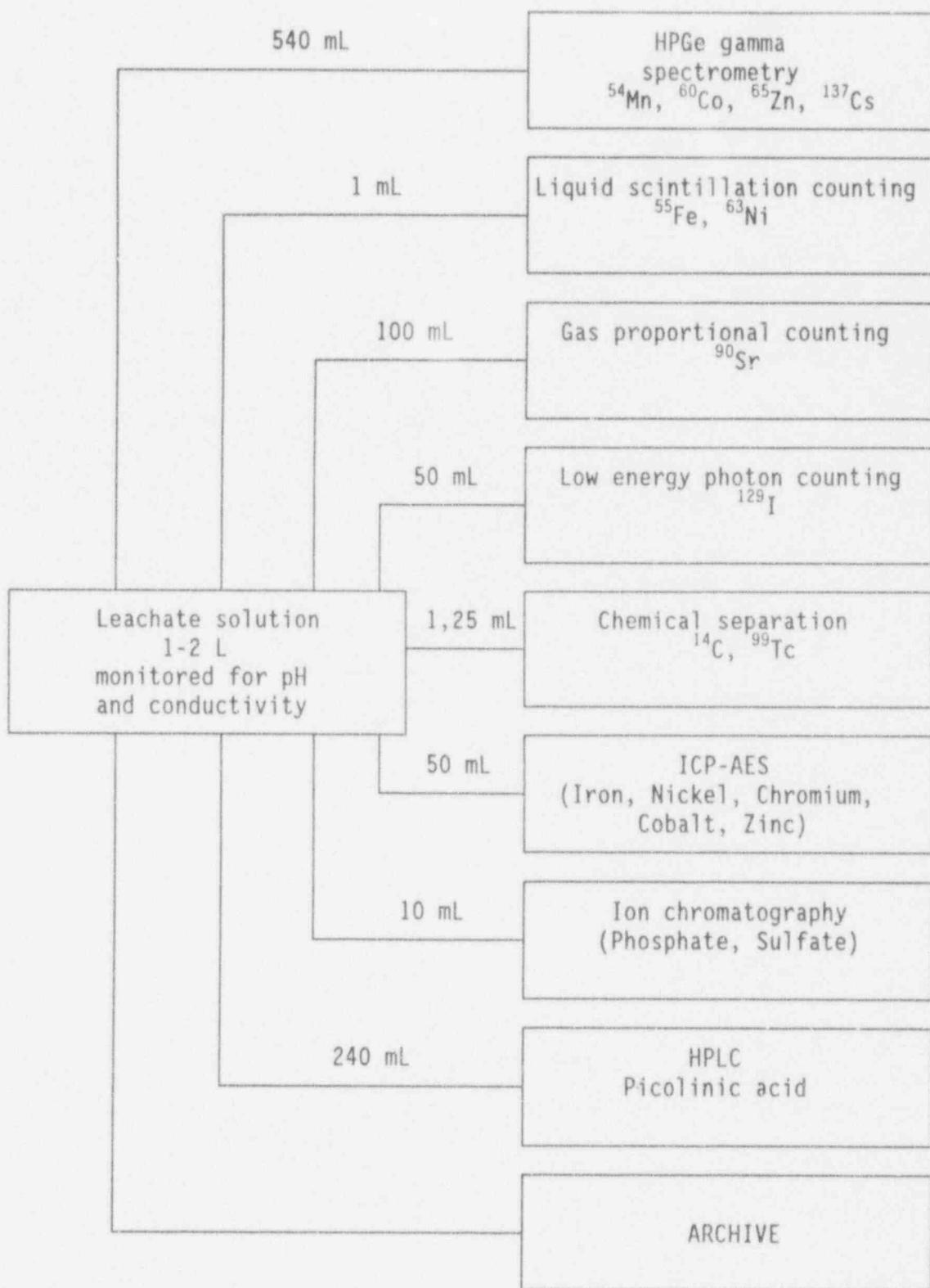


Figure 4. Leachate analysis methods.

Experimental Procedures

^{241}Am and ^{244}Cm) and for other radionuclides using standard environmental analysis procedures. Concentrations of gamma-emitting radionuclides (e.g., ^{60}Co , ^{137}Cs , and other measurable gamma emitters) in each resin sample were measured by diluting an aliquot of the dissolved resin with water and analyzing this volumetric sample using gamma-spectrometric techniques. When resin waste-form samples were analyzed, a small sample of the solidified waste was analyzed directly at a distance of 10 cm from the detector. The HPGe spectrometers used to analyze these samples were calibrated using reference sources traceable to the National Institute of Standards and Technology.

Concentrations of beta emitters (e.g., ^{55}Fe , ^{63}Ni , ^{90}Sr , ^{99}Tc , and ^{241}Pu) in the resin wastes were determined using radiochemical separation techniques followed by liquid scintillation or gas proportional counting. In the case of ^{14}C , the solid sample is carefully dissolved in an alkaline environment, and the dissolved sample aliquot undergoes oxidation, separation, collection as carbon dioxide, and analysis via liquid scintillation counting.³⁹ In the case of ^{129}I , the sample is fused with sodium hydroxide and extracted using carbon tetrachloride.⁴⁰ The ^{129}I is quantified using low-energy photon spectrometry. Specific procedures for these analyses are discussed in Appendix C.

Stable iron, nickel, iodine, carbon, and strontium carriers and ^{85}Sr tracer were added to an aliquot of the dissolved resin waste, and the solution was then passed through a chloride-form anion exchange column. Iron in the solution was left on the column while nickel and strontium passed through it. Unwanted radionuclides were washed from the column using HCl and HF acids, and the iron was then eluted using 0.5 M HCl. Ammonia and iron were added to the eluent of the column, and this alkaline solution was treated with dimethylglyoxime to selectively extract nickel, and sulfate was added to another aliquot to precipitate strontium as strontium sulfate. Concentrations of ^{55}Fe and ^{63}Ni were determined by analyzing the separated activities using a liquid scintillation

counter, and the concentration of ^{90}Sr was determined using a gas proportional counter.

The resin and waste-form samples analyzed for concentrations of transuranic isotopes were wet-ashed using nitric, sulfuric, and perchloric acids. This procedure was followed by a pyrosulfate fusion to dissolve any remaining undissolved compounds. The fusion was then dissolved in 2 M HCl, and the actinides were precipitated using barium sulfate. The barium sulfate precipitate was then dissolved in alkaline ethylene-diamine-tetra-acetic (EDTA) to precipitate the actinides as hydroxides. The hydroxide precipitate was then dissolved in acid, and then the solution was oxidized to adjust the oxidation state of plutonium. Americium and curium were precipitated as fluorides and, following filtration, this precipitate was mounted for analysis using a high-resolution alpha spectrometer. The filtrate was reduced, and plutonium was precipitated as plutonium fluoride. The plutonium fraction was also analyzed using a high-resolution alpha spectrometer.

Gamma-ray-emitting radionuclides in the leachate samples (e.g., ^{54}Mn , ^{60}Co , and ^{137}Cs) were analyzed using gamma-ray spectrometry as shown in Figure 4.

Concentrations of ^{14}C , ^{55}Fe , ^{63}Ni , ^{99}Tc , ^{129}I , and ^{90}Sr in leachate samples were determined by first using radiochemical techniques to selectively extract and concentrate these radionuclides. Carriers and tracers were added to volumetric samples of the leachates, and the samples were then evaporated to dryness. The samples were then dissolved in an HCl solution. These samples were then processed and analyzed using the same procedures used for the resin-waste samples.

The procedure used to analyze the leachates for ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm was the same as that used for the resins in that a pyrosulfate fusion and the following analysis program were the same for both the waste form samples and the leachates.

Elemental Analysis

Samples of dissolved resin wastes and leachate solutions were analyzed using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). All samples were analyzed for concentrations of chromium, iron, and nickel. These elements were selected for analysis because they are the primary constituents of stainless steel and Inconel, the materials used to line the internal surfaces of LWR primary coolant systems. In addition, the samples were analyzed for zinc, which is present in the reactor coolant system due to the presence of Admiralty brass. Potential analytical background interferences introduced by the presence of high concentrations of Ca^{+2} and other ions were evaluated using prepared standards.

Only lot-analyzed, trace-metal, analysis-grade reagents were used to prepare standards.

Chelating Agent Analysis

The waste-form specimens and leachates were analyzed for picolinic acid using ion chromatography. A Dionex ion chromatograph equipped with an ion-exchange column (AS-4A) was employed using 5 mM sodium hydroxide as an eluent. The detector was set at 254 nm. This method has a sub-parts-per-million detection limit for picolinic acid. For the Peach Bottom-3 cemented waste form samples, dilutions of the dissolved samples were analyzed because of the high concentration of other waste-form constituents present.

EXPERIMENTAL RESULTS

Experimental results from the Peach Bottom-3 waste-form samples can be divided into the results of two tests that were performed to evaluate waste form integrity/stability and leachability using the ASTM C39 compression testing procedure³ and the ANSI/ANS 16.1 leach test procedure,⁴ respectively. Compression tests were performed both before and after immersion tests as specified and described in Appendix A of the "Technical Position on Waste Form," Revision 1,² and discussed in Reference 34. The objective of the compression tests is to assess the structural stability of the waste form both before and after the 7- or 90-day immersion tests specified in the regulatory requirement.

During the 5-day leach test that was performed according to the ANSI 16.1 procedure, measurements were performed for pH and conductivity of the leachate; the concentrations of radionuclides, stable metals, and picolinic acid in the waste form; and the releases of these constituents from the waste form into the leachant. The releases to the leachant are quantified in terms of the absolute and fractional release rates, cumulative fractional release, effective diffusivity, and leachability index.

Waste Form Structural Stability

According to 10 CFR 61.56 (b)1, "a structurally stable waste form will generally maintain its physical dimensions and its form under the expected disposal conditions such as weight of overburden and construction equipment." The initial criterion for structural stability as identified in the NRC "Technical Position on Waste Form"¹ is that the cement-solidified waste-form specimen must exhibit a compressive strength of 50 psi (3.4×10^2 kPa). This was later raised to 60 psi (4.1×10^2 kPa) to reflect an increase in burial depth to 55 ft at the Hanford site.³⁴ However, as cement waste forms are nominally capable of achieving compressive strengths of 5,000 to 6,000 psi (3.4×10^4 to 4.1×10^4 kPa), Appendix A of the "Technical Position on Waste

Form," Revision 1, recommends a mean compressive strength of 500 psi for waste-form specimens cured for a minimum of 28 days. It has been determined in previous studies,⁴¹ that solidified ion exchange resins should be able to meet this criterion.

In addition to the initial compression tests, Appendix A of the "Technical Position on Waste Form," Revision 1, requires that a compressive strength test be performed after the waste form has been cured and immersed in water for varying periods of time depending on the waste type. In the case of decontamination ion-exchange resins with chelating agents, the waste form specimen must be cured for at least 180 days, immersed for a minimum of 7 days, followed by a drying period of 7 days. After the drying period, the waste form compressive strength should exceed 500 psi or 75% of the pre-immersion compressive strength to meet the regulatory requirement. The reason for the special requirements for ion-exchange resins is that several studies have shown that cure time and immersion resistance are related and that longer cure times improve the compressive strength of the waste form.^{41,42} In addition, in the case of pozzolanic cements (e.g., Brunswick, FitzPatrick, and Peach Bottom-3), hydration of the cement is slower; therefore, ultimate strength may not be obtained for up to a year.^{43,44} The Peach Bottom-3 samples were cured for 900 days prior to compression testing and consequently had reached their maximum strength.

In this study, compressive strength measurements were performed on specimens both before and after immersion testing. The results of the structural stability tests can be divided up into the results of the compression testing that was performed on the specimens before immersion testing and those performed on ones that retained their structural stability during immersion testing. Table 5 summarizes the compressive strength test results for both before and after immersion testing. Some compression tests were also performed on the plant PCP samples that are used for the

Table 5. Compressive strengths of Peach Bottom-3 waste form samples.^{a,b}

Sample	Area (in. ² /cm ²)	Yield load (lb/kg)	Yield strength (psi/kPa)
<u>90-day leach test</u>			
1. Unleached	2.7/17.4	3,370/1,530	1,240/8,570
2. Unleached	2.7/17.4	2,620/1,190	970/6,710
3. Unleached	2.7/17.4	3,250/1,480	1,210/8,340
Average $\pm \sigma$		3,000 \pm 400/(1,360 \pm 180)	1,140 \pm 150/(7,870 \pm 1,010)
1. Deionized water	2.72/17.5	3,240/1,470	1,190/8,210
2. Deionized water	2.54/16.4	3,390/1,540	1,330/9,200
Average $\pm \sigma$		3,320 \pm 110/(1,500 \pm 50)	1,260 \pm 100/(8,710 \pm 700)
1. Sea water	2.7/17.4	2,720/1,230	990/6,840
2. Sea water	2.7/17.4	3,810/1,730	1,370/9,630
Average $\pm \sigma$	—	3,270 \pm 770/(1,480 \pm 350)	1,190 \pm 280/(8,240 \pm 1,970)
<u>7-day immersion test</u>			
Control ^c	—	2,760 \pm 30/(1,250 \pm 10)	1,010 \pm 10/(6,980 \pm 70)
Deionized water ^{c,d}	—	2,590 \pm 30/(1,180 \pm 10)	956 \pm 10/(6,600 \pm 70)
Deionized water ^{c,d}	—	2,750 \pm 30/(1,250 \pm 10)	1,010 \pm 10/(6,980 \pm 70)
Simulated Barnwell ^{c,d}	—	2,750 \pm 30/(1,250 \pm 10)	1,020 \pm 10/(7,060 \pm 70)
Simulated Barnwell ^{c,d}	—	2,660 \pm 30/(1,210 \pm 10)	970 \pm 10/(6,720 \pm 70)
<u>PCP samples</u>			
Control (PCP #2)	2.54/16.8	1,780 \pm 280/(810 \pm 130)	360 \pm 50/(2,490 \pm 350)
Deionized water (PCP #1)	2.69/17.4	5,770 \pm 270/(2,620 \pm 122)	1,140 \pm 50/(7,900 \pm 350)
Simulated Barnwell (PCP #3)	2.	2,250 \pm 750/(1,020 \pm 340)	385 \pm 275/(2,670 \pm 1,900)

a. The total cure time varied but was approximately 900 days from when samples were taken. The water-to-cement ratio is approximately 0.33 based on the interstitial water content of the samples, and the cross-sectional area for all is nominally 16 cm².

b. The PCP compression tests identified in the table were performed on conical samples. The compression surface area was based on the smaller of the two ends as discussed in Reference 48. Nominally, the smaller surface area was 17 cm². These samples were leached for 90 days.

c. Additional measurements were performed to compare compression tests for deionized and simulated Barnwell groundwater samples. This compression test study was performed after the samples had been leached in the listed leachants for a period of 7 days. The data indicate no significant differences between the effect of the deionized water and simulated Barnwell water on the compressive strength. Unleached "control" samples were compression tested for quality control purposes and for comparison with the leached samples.

d. Compression testing performed after 7 days immersion and 7 days drying. All other samples were immersed for 90 days prior to compression testing.

Experimental Results

plant tests and were prepared by the Peach Bottom staff. They were cast into paper cups and were therefore not true cylinders. After a review of the compression test procedure,⁴⁸ it was determined that the yield strength should be calculated using the smaller surface area of the two ends.

Table 5 lists the compression test results for the unleached samples. These samples all easily meet the regulatory requirement with the exception of one PCP sample. However, their compressive strengths are well below the nominal compressive strength values of 5,000 to 6,000 psi for cement waste forms that don't contain decontamination ion-exchange resins. The PCP control sample has the lowest compressive strength at 360 psi. The water-to-cement (w/c) ratio noted in Footnote a of Table 5 was calculated as the ratio of the weight of free interstitial water to the weight of cement and other constituents used during the solidification of the full-scale waste form from which the waste-form specimen was collected. The w/c ratio was calculated using the method developed by Neilson,³⁵ which divides the water content of the resins between the water content of the "as-received" and dewatered ion-exchange resins to determine the amount of available interstitial water. It should be noted that the w/c ratio has been identified as the critical parameter affecting the strength and chemical resistance of a hardened cement mix⁴⁵ and in determining the permeability of a waste form. Studies (Reference 45) indicate that permeability of the waste form increases significantly at w/c ratios greater than 0.5. In this study, the w/c ratio is about 0.3; consequently, the increased permeability due to high w/c ratios would not be expected to influence the results of this study as permeability is similar in the range of w/c ratios from 0.3 to 0.5 (Reference 5).

The leachants used for the immersion tests were deionized water, simulated seawater, and simulated groundwater. As indicated in Table 5, all immersion tests were performed for 90 days with the exception of those identified by Footnote d. For the samples that were immersion-tested for 90 days, after 30 days of immersion, small amounts of resin and cement debris (<1g) were

observed in the bottoms of the leaching containers. However, all samples maintained their physical integrity and remained without cracks during the immersion experiment. Figure 5 shows a Peach Bottom-3 sample after immersion in deionized water and compression testing, and Figure 6 shows a leached PCP sample after the compression test. The figures show the difference in the configuration of the INEL samples as compared to the PCP samples prepared by the plant. The cracking in the waste forms was not present prior to the beginning of the compression test.

An assessment of the Peach Bottom-3 post-immersion compression testing results shown in Table 5 indicates that the compressive strengths ranged from 956 to 1,370 psi for all samples except the PCP sample leached in simulated Barnwell groundwater. These compressive strengths are lower than the compressive strengths of laboratory specimens of solidified decontamination ion-exchange resin.⁴⁶ Compression test data from that laboratory study indicated nominal compressive strengths for Citrox waste forms of approximately 3,000 psi and of 1,800 psi for LOMI waste forms. The lower value for the LOMI waste form was not explained in Reference 46. The reduced compressive strength of the actual waste form samples may be due to the additional constituents found in the waste forms as compared to the laboratory formulations. The low compressive strength of the sample immersion tested in simulated Barnwell groundwater may be due to poor sample preparation as the PCP control sample also had a low compressive strength.

The compressive strength data from the deionized water and simulated Barnwell groundwater tests also show no apparent difference in the effects of the leachants in which the specimens were immersion-tested. Consequently, leachants have no apparent effect on compressive strength. Further, there were no apparent differences in the compressive strengths of the Peach Bottom-3 specimens that were immersion-tested for either 7 or 90 days. In all cases, the waste forms met the requirement in the "Technical Position on Waste Form," Revision 1, for a 500-psi load, and there



Figure 5. Peach Bottom waste form specimen after immersion and compression testing

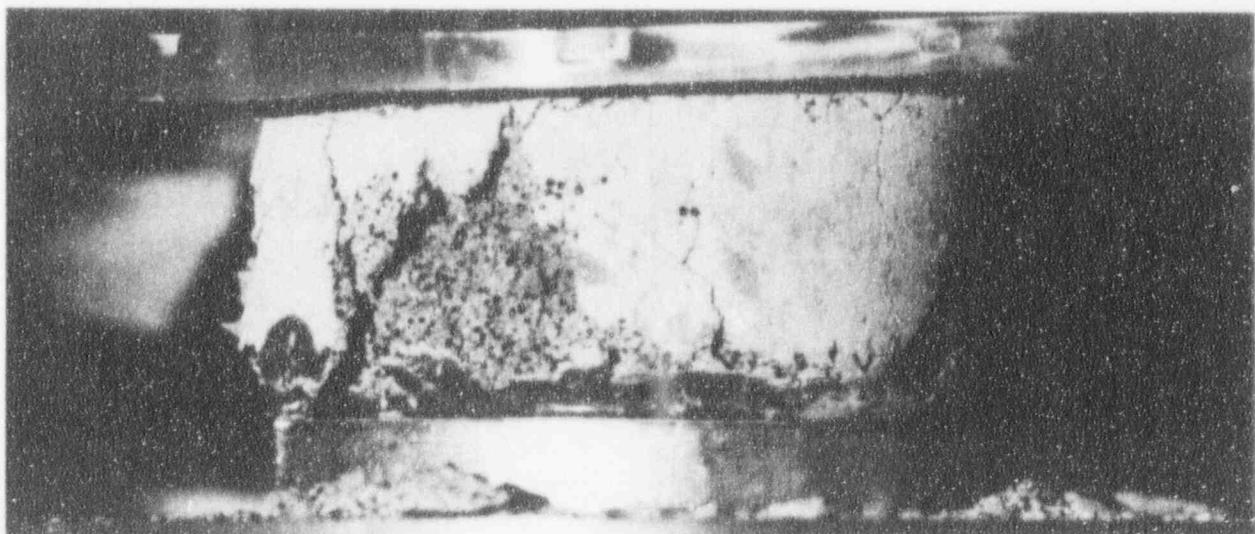


Figure 6. Process control program specimen after immersion and compression testing.

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were no significant differences between the samples immersed in deionized water and those immersed in seawater.

Leach Test Results

Appendix A of the "Technical Position on Waste Form," Revision 1, has made two changes to the requirements of the "Technical Position on Waste Form" in the area of leach testing. First, the leach test period has been reduced from 90 days to 5 days. Second, the bulk of the leach testing may be conducted with only one of two leachants, either deionized water or seawater, whichever is most aggressive (i.e., whichever has the higher initial release rate). The objective of the 90-day test was to determine whether the leaching process was time dependant as has been assumed in the ANSI/ANS 16.1 leach test procedure. The changes were reflected in Revision 1 because it was assumed that if mechanisms existed other than diffusion-controlled leaching (e.g., erosion, corrosion, or dissolution), they would be identified by visual inspection during the immersion test and that the primary diffusion-controlled release occurs during the initial 5 days.³⁴ The primary objective of the leach test is to ensure that the leachability index is greater than 6.0, which is required to meet the regulatory requirement.

During the Peach Bottom-3 leach test, leachates were analyzed for the radionuclides, chelating agents, and stable metals found in the decontamination resin waste forms. In addition, measurements were made on the leachates for pH and conductivity, and to determine the leachability of the species released from the decontamination ion-exchange resin waste form. The following sections present results of the pH and conductivity measurements; the concentration measurements performed for radionuclides, stable metals, and picolinic acid in the waste forms; and the leach test results. Leaching data are shown graphically as plots of cumulative fractional release versus the square root of elapsed leaching time. This presentation was chosen to facilitate evaluation of the data since the plots will be linear if diffusion is the controlling release

mechanism and will have slopes that are proportional to the corresponding effective diffusivities.

Leachate pH and Conductivity

To assess the effects of pH and conductivity on the leachability of the waste-form specimens, pH and conductivity measurements were made on the Peach Bottom-3 leach solutions at the end of each leaching period. The pH and conductivities for each leaching interval are plotted in Figures 7 and 8, respectively, and the numerical results are listed in Appendix D. During the first 5 days of leaching, leachates were changed out after elapsed times of 30 seconds, 2 hours, 7 hours, 24 hours, 48 hours, 72 hours, 96 hours, and 120 hours. The final pH of the leachates for all three waste-form samples reached maxima that ranged between 10.5 and 11.9 for all leachates except the 30-second prerinse. These data suggest that the pH is affected within a few hours and probably within a few minutes by the chemistry of the waste form.

Vejmelka¹⁹ indicates that the cement chemistry will control the leachate pH unless magnesium, a constituent of seawater, is present in the leachate in significant concentrations. He indicates that the solubility of the magnesium hydroxide formed in solution will set the pH for seawater. The effects of pH on releases of radionuclides and stable metals are not clear because the pH (with the exception of seawater) is controlled by the pH of the waste form. It has been suggested⁴⁷ that the ion strength of seawater may be a more important parameter and have a greater effect on radionuclide solubilities in the leachate than the pH.

Conductivity is a measure of the quantity of ions that are present in a solution and that are capable of transferring an electrical charge. As noted in Reference 47, the conductivity, which is a measure of ion strength, can affect leachability. Leachate conductivities shown in Figure 8 (expressed as $\mu\text{mho}/\text{cm}$ at 25°C) were measured at the same time pH measurements were made at the end of each leach period for sample #8. However, in the cases of samples #4 and #12, the conductivity measurements were made at a later date

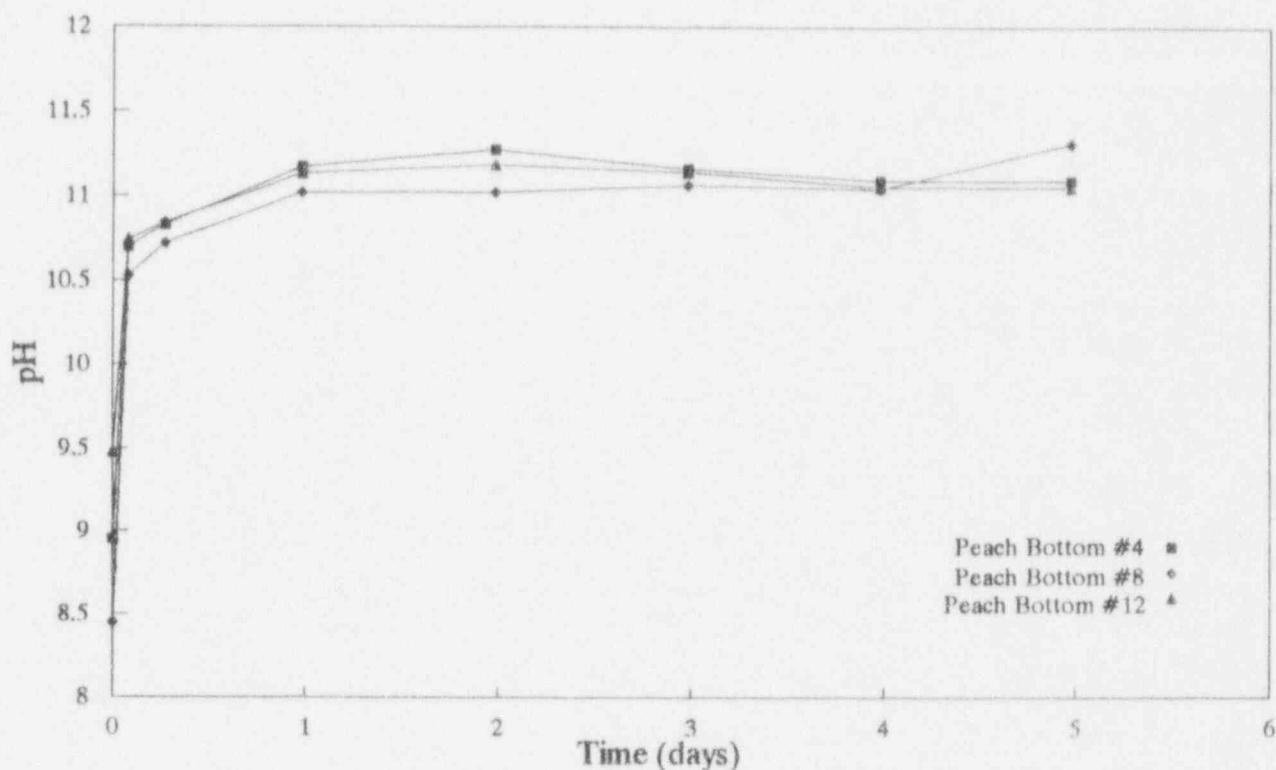


Figure 7. Leachate pH for all specimens.

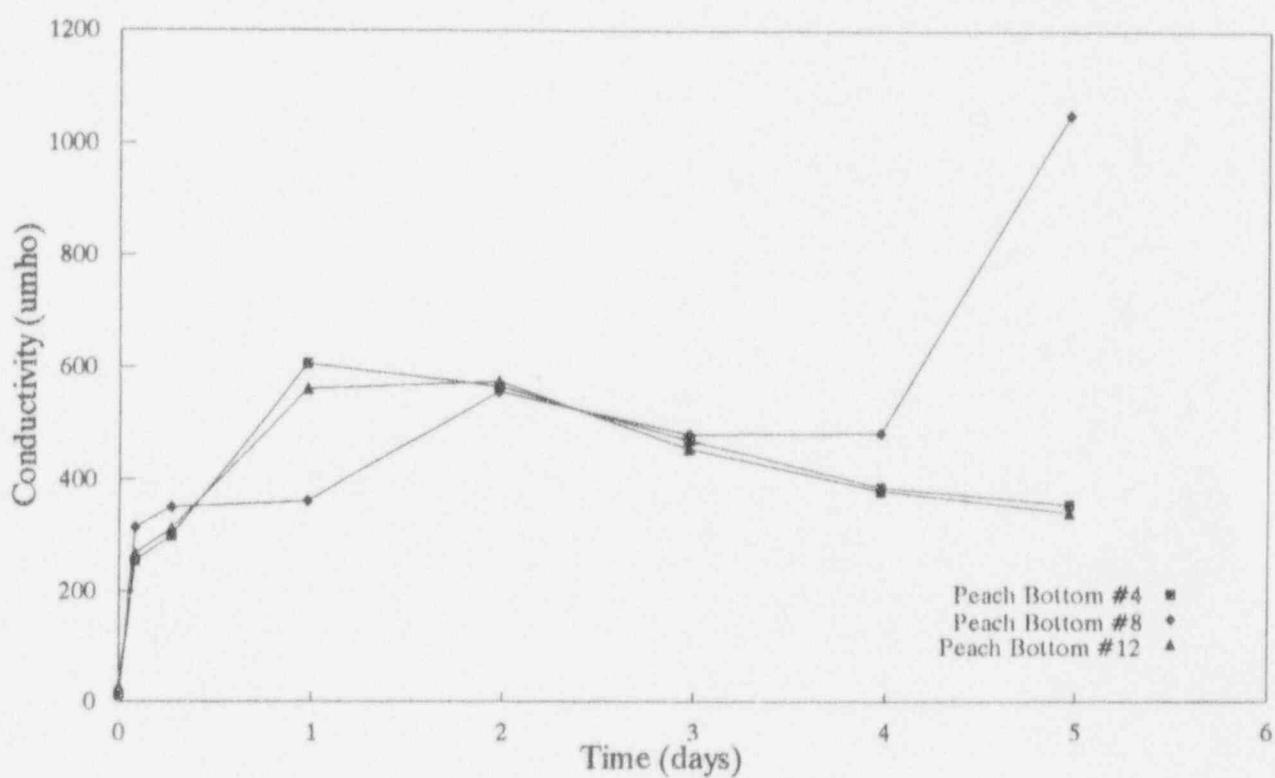


Figure 8. Leachate conductivity for all specimens.

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because of an instrument failure. Comparison of the results between samples indicates that the delay did not effect the conductivities due to the similarity between the results for all samples.

For all waste-form samples, the conductivities ranged from 255 to 350 $\mu\text{mho}/\text{cm}$ after the initial 2 hours of leaching. They then rose to maxima ranging between 560 and 605 $\mu\text{mho}/\text{cm}$ between the first and second day; then they decreased to minima between 341 and 355 $\mu\text{mho}/\text{cm}$ for samples #4 and #12. In contrast, the conductivity for sample #8 went to a maximum of 1,050 $\mu\text{mho}/\text{cm}$ at 5 days. This behavior is inconsistent with the conductivity of the other samples and suggests an instrument or recording error, although no discrepancies could be identified. No effect of this change in conductivity on the pH or leaching results has been identified.

Concentrations of Radionuclides, Stable Metals, and Chelating Agents in Resin Wastes and the Cemented Waste-Form

As noted in Appendix A of the "Technical Position on Waste Form," Revision 1, although some reactor waste streams are relatively well characterized and free of secondary ingredients, some waste streams such as ion-exchange resins may contain chemicals that can retard or accelerate the hydration of cement or otherwise adversely affect cement waste-form performance. Some chemicals commonly found in nuclear power plants that may affect solidification are listed in the proceedings of the Cement Workshop.⁶ One primary component identified in this evaluation was the chelating agent, picolinic acid.

In this study waste stream samples were obtained to determine the inventories of radionuclides, stable metals, and chelating agents that would be leached from the waste forms being leached. Table 6 lists the concentrations of radionuclides, stable metals, and chelating agents present in the Peach Bottom-3 wastes solidified as part of this study. In Table 6, the primary decontamination-produced radionuclides to be discussed are ^{14}C , ^{55}Fe , ^{60}Co , and ^{63}Ni . The

results for fission products ^{137}Cs , ^{99}Tc , and ^{129}I have been added for comparison purposes and because of the importance of possible releases of ^{129}I . Only a brief discussion of the transuranic radionuclides is presented as they were measurable in the resin and waste form samples, but they were not detected in the leachates. Consequently, only detection limit values were available for calculational purposes.

The concentrations of radionuclides, chelating agents, and stable metals in the resin wastes and waste forms are presented in Table 6 as $\mu\text{Ci/g}$ for radionuclides or $\mu\text{g/g}$ of stable metals or chelates in the water-saturated resin. Analyses of both resin and solidified waste-form samples were performed. The summed radionuclide content is 7.8 $\mu\text{Ci/g}$ of waste form based on a combination of the resin and cement results. Resin results for ^{54}Mn , ^{125}Sb , ^{137}Cs , ^{242}Cm , ^{14}C , and ^{99}Tc were multiplied by the resin-to-total-material ratio (0.5) for inclusion in the total. The primary decontamination radionuclides present in the resins based on their measured concentration are ^{54}Mn , ^{65}Zn , ^{60}Co , ^{55}Fe , ^{63}Ni , and ^{14}C . The summed activity of these radionuclides is 7.7 $\mu\text{Ci/g}$ waste form or about 98% of the total activity. Carbon-14 makes up about 58% of the total activity. The dominant decontamination radionuclides, ^{60}Co and ^{55}Fe , make up about 31% and 1.9% of the total activity in the resin waste, respectively. In contrast, the fission products ^{90}Sr , ^{99}Tc , ^{129}I , and ^{137}Cs collectively constitute about 0.3% of the total activity. The concentrations of the transuranic isotopes are also low and sum to a total of $1.6 \times 10^{-3} \mu\text{Ci/g}$ (0.02% of the total activity). Greater than 87% of the transuranic activity was ^{241}Pu .

A review of Table 6 indicates that there are significant differences between the measured resin and waste form concentrations for some radionuclides. In order to compare the resin and waste-form data, the radionuclide concentrations in the resin are multiplied by the ratio of the weight of resin in the sample to the weight of all components (0.52) (includes resin, cement, trisodium phosphate, and pozzolana) for comparison purposes. This ratio is approximately 0.5, in contrast

Table 6. Summary of Peach Bottom resin and cement waste form characterizations (decay date 10/25/89).

Radionuclide	Peach Bottom resin ($\mu\text{Ci/g}$ or $\mu\text{g/g}$)	Peach Bottom cemented waste forms	
		Sample #1	Sample #2
^{54}Mn	4.3E-1 \pm 2.7E-2		
^{55}Fe	1.3E 0 \pm 5.5E-2	1.5E-1 \pm 1.6E-2	1.5E-1 \pm 1.6E-2
^{60}Co	4.6E 0 \pm 1.2E-2	2.4E0 \pm 1.5E-2	2.4E0 \pm 1.5E-2
^{63}Ni	7.5E-2 \pm 6.8E-3	1.6E-2 \pm 1.4E-3	3.7E-2 \pm 3.1E-3
^{65}Zn	6.0E-1 \pm 1.1E-2	3.9E-1 \pm 7.8E-2	5.6E-1 \pm 1.6E-1
^{125}Sb	1.7E-2 \pm 9.8E-4		
^{137}Cs	3.6E-3 \pm 5.1E-4		
^{90}Sr	2.2E-5 \pm 2.3E-6	3.0E-5 \pm 5.5E-6	5.3E-5 \pm 7.6E-6
^{238}Pu	6.6E-5 \pm 1.6E-6	7.2E-5 \pm 4.1E-6	9.5E-5 \pm 5.2E-6
^{239}Pu	2.2E-5 \pm 7.5E-7	2.6E-5 \pm 1.6E-6	2.4E-5 \pm 1.5E-6
^{241}Pu	1.8E-2 \pm 4.3E-4	2.7E-4 \pm 1.5E-5	2.5E-3 \pm 1.4E-4
^{241}Am	5.2E-5 \pm 1.4E-6	1.0E-4 \pm 6.0E-6	1.3E-4 \pm 7.0E-6
^{242}Cm	9.2E-6 \pm 9.2E-7		
^{244}Cm	1.3E-4 \pm 1.6E-4	1.5E-4 \pm 8.0E-6	1.3E-4 \pm 6.9E-6
^{14}C	8.7E+0 \pm 8.7E-2	3.1E-4 \pm 2.7E-5	2.5E-4 \pm 2.1E-5
^{99}Tc	3.7E-2 \pm 8.6E-4	2.7E-4 \pm 1.5E-5	1.25E-3 \pm 6.9E-5
^{129}I	< 5E-6	< 5.8E-6	2.4E-5 \pm 3.5E-6
Chromium ^a	650 \pm 20	103 \pm 6	103 \pm 6
Iron ^a	3,000 \pm 160	1.73E+4 \pm 500	1.78E+4 \pm 600
Zinc ^a	158 \pm 12	66 \pm 3	83 \pm 3
Nickel ^a	730 \pm 30	66 \pm 12	54 \pm 8
Cobalt		15	12
Boron ^b		16 \pm 2	21 \pm 3
Phosphate ^b			
Sulfate ^b			
Picolinic acid ^c	< 5E-6 M	1.68 wt% \pm 0.21	2.02 wt% \pm 0.08

a. Analyses performed using inductively coupled plasma spectroscopy elemental analysis methods.

b. Analyses performed using ion chromatography.

c. Analysis performed using picolinic acid titration. Due to uncertainties in the solidified waste analysis, the loading in the liner will be used.

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to the volume ratio (0.68). If this is done, the following radionuclides still have significant differences between the measured resin and waste form concentrations (the factor of the resin divided by the waste form concentration is shown in parenthesis): ^{55}Fe (4.5), ^{241}Pu (13), ^{14}C (1.6×10^4), and ^{99}Tc (25). Nominally, these results indicate significant differences in the radionuclide concentrations in the resin and the waste-form samples. However, differences in the concentrations in the two waste-form samples that were analyzed are also indicated in Table 6. Ratios of the radionuclide concentrations for the two cemented waste forms analyzed were calculated to assess significant differences in the waste-form concentrations. Significant differences are indicated for ^{63}Ni (2.3), ^{241}Pu (9.2), ^{241}Am (1.3), ^{244}Cm (1.2), ^{14}C (1.2), and ^{99}Tc (4.6). These data indicate that the radionuclide concentrations in the resin and the waste form are not uniform; therefore, the inventories are not known within a factor of between about two and 10 for the identified radionuclides. However, uniformity does not explain the differences observed for ^{14}C and ^{99}Tc . In both cases, the differences between the resin and waste-form concentrations are large. Discussions with laboratory personnel indicates that the waste-form analyses were difficult and that interferences or losses due to the presence of the waste-form constituents may have affected the results. Consequently, for calculational purposes, the higher resin concentrations have been used. These data suggest that differences in leaching results that are less than a factor of two are insignificant and that differences of between 5 and 10 are required to make significant conclusions.

Among the stable metals whose concentrations were measured in the resin wastes, the iron concentration was highest at 3,000 $\mu\text{g}/\text{gram}$ resin. This was followed by nickel and chromium. In addition, analyses were performed for sulfate and phosphate. Neither type of ion was detectable in the waste form or in the resin samples. Differences between the resin and waste-form concentrations have also been identified. The ratio of the resin to waste-form sample concentrations differ for chromium (3.2) and nickel (5.5). These

data again suggest that the waste forms are not homogeneous.

The concentration of picolinic acid was measured for several waste-form samples. These results ranged from 1.68 to 2.02 wt%, which is less than the 5.5 wt% added to the liner. However, because of concerns about degradation of the picolinic acid, the acid leaching technique used for the waste-form measurements may not have provided a quantitative yield. Consequently, the liner inventory value will be used for the leaching calculations.

Chelating Agent, Radionuclide, and Stable Metal Releases

Piciulo⁴⁶, Soo,¹⁶ and others^{19,39,48} have addressed the release of chelating agents, radionuclides, and/or stable metals in the laboratory. These include LOMI decontamination processes with picolinic acid as the chelating agent and other waste forms with other chelating agents. However, these studies used simulated wastes that did not contain all of the constituents found in commercial nuclear power plant waste streams; also, as discussed in Reference 34, other waste stream constituents found at commercial nuclear reactors may affect the leachability characteristics of the waste form.

The physical factors that influence mass transfer of chelating agents, radionuclides, and stable metals into the leachant can be divided into three mechanisms: (a) convection, (b) diffusion through pores within the waste form, and (c) diffusion at the interface of the waste form and the surrounding coolant.¹⁵ Convection refers to bulk movement through the waste form and is expected to be insignificant for intact waste forms. In this study, where a number of the waste forms decomposed during leaching, it may be expected to be significant, but as will be discussed, there does not appear to be a significant difference in the releases from degraded and intact waste forms. Mechanism 2, pore diffusion, is induced by concentration gradients within a waste matrix and results in diffusion of radionuclides and other constituents to the surface of the waste form. This mechanism has been described

as a porous solid with water-filled pores.^{19,45} The final mechanism is the diffusion at the interface between the surface of the waste form and the surrounding leachant. This can be considered to be the formation of a diffusion gradient between the surface of the waste form and the surrounding leachant. If this diffusion gradient is formed and there is little movement of leachant, an equilibrium would be established that would be expected to reduce releases from the waste form. Each of these mechanisms may contribute to releases from the intact and degraded waste forms and would be expected to be modified by the effects of other constituents of the waste form, with chelating agents possibly having the greatest effect due to their high concentration in the waste form (up to 6 wt%).

An assessment of the leachability characteristics of decontamination ion-exchange resin waste forms is performed through comparisons of fractional release rates, cumulative fractional releases, effective diffusivities, and leachability indexes for the species of interest to assess the effects of the different chelating agents on releases from the waste forms. Appendix D contains detailed tables showing the leach test results. Raw results with uncertainties are shown in Appendix Table D-5. Appendix Table D-6 has been included for comparison purposes and contains the same information for the FitzPatrick sample (Reference 17), which degraded during leaching. Various forms of data presentation have been used, including cumulative fractional release/cm² surface area/year, Ci/year, Ci/m³/year, cumulative fraction released/year, and cumulative fraction released/m³/year. These data are specific to the Peach Bottom-3 waste forms and should not be extrapolated to other waste-form dimensions or compared with releases from other waste forms as the data are decay dependent.

Tables 7 to 9 contain summaries of the CFRs, the average absolute release rates, the average fractional release rates, the average effective diffusivities, and the leachability indexes for the Peach Bottom-3 samples. These averages are for nonlinear rate phenomena and, therefore, include

high-rate data from early in the leaching process and low-rate data from later in the process. Averages are not typically calculated for phenomena of this type; however, these averages and their associated uncertainties have been calculated because they are the common method used for comparisons with other leach test studies. Consequently, uncertainties associated with the average rates are expected to be large (typically from 50–100% at one standard deviation). Uncertainties listed for the average absolute release rates, the average fractional release rates, and the average effective diffusivities are the internal uncertainties associated with differences in the rate or diffusivity results and do not contain uncertainties associated with counting statistics, waste form inventories, or other uncertainties associated with the leaching process. These uncertainties have been excluded because they are either small relative to the quoted uncertainty or are not quantifiable based on the tests performed. Exceptions (e.g., ⁹⁰Sr) are discussed in the following sections. An uncertainty of one standard deviation is quoted in the tables.

Table 10 contains error-weighted averages with a one-standard-deviation uncertainty for the results from Tables 7 through 9 for samples #4 and #12 and for samples #4, #8, and #12. The uncertainties listed in Table 10 are based on the uncertainties listed in Tables 7 through 9.

Chelating Agent

In this study, the release of the chelating agent picolinic acid was evaluated first because it provides a basis for interpreting the radionuclide and stable-metal results. If the radionuclide or stable-metal release is enhanced by chelating effects, the release rates of the chelant and the species of interest may be comparable and should provide an indication of which radionuclides or stable metals have enhanced release rates. Figures 9 and 10 show the fractional release rates and cumulative fractional releases for picolinic acid. The chelating agent was measured in all the leachate samples for waste-form samples #4 and #12. The remaining Peach Bottom-3 sample, #8, was analyzed by a different laboratory, which was

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Table 7. Leach test results for Peach Bottom solidified resin waste form #4, leached in deionized water.

Nuclide	CFR	Release rate		Average effective diffusivity (cm ² · s ⁻¹) ^a	Leachability index
		Absolute ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) ^a	Fractional (cm ² · s ⁻¹) ^a		
¹⁴ C	1.2E-4	6.9 ± 13E-9	6.2 ± 12E-12	4.3 ± 5.0E-14	14.1
⁵⁵ Fe	8.6E-4	1.2 ± 2.4E-9	7.6 ± 16E-11	3.8 ± 8.2E-12	12.9
⁶⁰ Co	1.7E-3	2.4 ± 3.1E-8	6.4 ± 8.2E-11	4.4 ± 1.9E-12	11.4
⁶³ Ni	5.6E-3	1.4 ± 1.8E-9	2.1 ± 2.7E-10	5.2 ± 5.3E-11	10.4
⁹⁰ Srb	2.4E-3	4.8 ± 6.4E-12	5.0 ± 6.7E-10	8.2 ± 10E-11	10.6
⁹⁹ Tc	2.1E-3	2.0 ± 1.7E-10	4.3 ± 3.6E-11	9.2 ± 17E-12	11.5
¹²⁹ I	9.6E-2	3.6 ± 5.6E-11	5.9 ± 9.3E-9	3.6 ± 3.1E-8	7.9
Chelating agent or metal		($\mu\text{g} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)			
Chromium	1.4E-2	1.0 ± 1.1E-5	4.1 ± 4.2E-10	3.0 ± 0.7E-10	9.5
Iron ^b	1.8E-5	2.2 ± 0.5E-6	5.1 ± 1.1E-13	2.3 ± 0.7E-15	14.7
Cobalt	4.3E-2	5.4 ± 6.5E-6	1.7 ± 2.0E-9	3.6 ± 3.3E-9	8.6
Nickel	2.3E-2	1.1 ± 1.4E-5	7.7 ± 9.2E-10	8.5 ± 2.7E-10	9.1
Zinc ^b	4.4E-3	1.1 ± 1.3E-5	6.1 ± 6.9E-10	1.4 ± 1.0E-10	10.0
Boron ^b	3.0E-2	9.1 ± 11E-6	2.0 ± 2.3E-9	3.1 ± 0.9E-9	8.5
Picolinic Acid	2.0E-2	8.2 ± 8.8E-3	6.0 ± 6.5E-10	5.4 ± 1.4E-10	9.3

a. Internal uncertainty associated with calculated results. Does not include counting statistics or other uncertainties associated with the leaching process.

b. Average values were calculated excluding zeros.

unable to detect picolinic acid in the leachate samples. The fractional release rates for the Peach Bottom-3 samples indicate similar behavior for both samples with the highest rate for the 2-hour leachate. In addition to the fractional release rates for the Peach Bottom-3 sample, the fractional release rates for the FitzPatrick mixed-bed resin waste-form specimen leached in deionized water during a previous part of this study¹⁷ have been included in Figure 9. As discussed in Reference 17, this

sample decomposed early in the leaching process and provides a basis for comparison with the intact Peach Bottom-3 samples.

A comparison of the fractional release rates for the Peach Bottom and FitzPatrick samples indicates a varying release rate for picolinic acid from the FitzPatrick sample probably due to convection release during the breakup of the waste form (significant cracking within 2 hours). However, a comparison of the average fractional release rates

Table 8. Leach test results for Peach Bottom solidified resin waste form #12, leached in deionized water.

Nuclide	CFR	Release rate		Average effective diffusivity (cm ² · s ⁻¹) ^a	Leachability index
		Absolute ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) ^a	Fractional (cm ⁻² · s ⁻¹) ^a		
¹⁴ C	5.5E-5	2.7 ± 4.3E-9	2.5 ± 3.9E-12	5.5 ± 4.8E-15	14.4
⁵⁵ Fe	2.0E-3	6.6 ± 6.4E-10	4.4 ± 4.2E-11	1.1 ± 1.7E-11	11.5
⁶⁰ Co	1.7E-3	2.2 ± 2.8E-8	6.1 ± 7.7E-11	4.4 ± 1.5E-12	11.4
⁶³ Ni	6.2E-3	1.5 ± 2.0E-9	2.4 ± 3.2E-10	5.8 ± 2.9E-11	10.3
⁹⁰ Srb	1.4E-3	1.5 ± 1.8E-12	1.6 ± 1.9E-10	1.4 ± 0.9E-11	11.2
⁹⁹ Tc	3.1E-3	2.0 ± 1.3E-10	4.4 ± 2.8E-11	1.7 ± 1.7E-11	11.5
¹²⁹ I	5.7E-2	1.6 ± 2.0E-11	2.6 ± 3.4E-9	1.9 ± 2.8E-8	8.3
Chelating agent or metal		($\mu\text{g} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)			
Chromium	1.9E-2	1.5 ± 1.8E-5	6.1 ± 7.3E-10	5.4 ± 1.3E-10	9.3
Iron ^b	7.4E-5	1.4 ± 1.5E-5	3.2 ± 3.5E-12	1.1 ± 0.6E-14	14.0
Cobalt	4.1E-2	3.7 ± 3.9E-6	1.2 ± 1.2E-9	3.4 ± 2.3E-9	8.7
Nickel	3.5E-2	1.8 ± 2.2E-5	1.2 ± 1.5E-9	2.0 ± 0.6E-9	8.7
Zinc	1.2E-2	8.6 ± 11E-6	4.7 ± 6.1E-10	2.6 ± 1.5E-10	9.6
Boron ^b	2.4E-2	6.3 ± 4.2E-6	1.4 ± 0.9E-9	4.0 ± 1.9E-9	8.4
Picolinic acid	2.0E-2	7.6 ± 7.8E-3	5.6 ± 5.8E-10	5.4 ± 1.3E-10	9.3

a. Internal uncertainty associated with calculated results. Does not include counting statistics or other uncertainties associated with the leaching process.

b. Average values were calculated excluding zeros.

for the Peach Bottom-3 samples with the FitzPatrick sample (Tables 7, 8, and 9 with the average in Table 10) indicates that the average fractional release rate of picolinic acid for the FitzPatrick sample ($3 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$) is about a factor of five higher than that from Peach Bottom-3 ($5.8 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$). Examination of the FitzPatrick data indicates that the average

fractional release rate ($2.4 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$) after the initial high rates due to convection is statistically the same as that from the Peach Bottom-3 sample ($5.8 \pm 4.3 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$). These data suggest that after an initial high release rate from the decomposed waste form due to convection, diffusion-driven release is similar for both intact and decomposed waste forms.

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Table 9. Leach test results for Peach Bottom solidified resin waste form #8, leached in deionized water.

Nuclide	CFR	Release rate		Average effective diffusivity (cm ² · s ⁻¹) ^b	Leachability index
		Absolute ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) ^a	Fractional (cm ² · s ⁻¹) ^a		
¹⁴ C	6.9E-4	2.5 ± 3.0E-8	2.2 ± 2.8E-11	9.9 ± 7.2E-13	12.1
⁵⁵ Fe	1.9E-2	1.8 ± 3.2E-8	1.2 ± 2.1E-9	1.4 ± 2.1E-9	9.7
⁶⁰ Co	3.0E-3	3.3 ± 4.1E-8	9.0 ± 11E-11	1.8 ± 1.6E-11	10.9
⁶³ Nib	5.0E-5	2.6 ± 3.3E-11	4.0 ± 5.1E-12	1.2 ± 1.1E-14	14.3
⁹⁰ Srb	1.7E-2	1.5 ± 2.4E-11	1.6 ± 2.5E-9	1.7 ± 2.4E-9	9.3
⁹⁹ Tc	2.6E-2	1.9 ± 3.3E-9	4.1 ± 7.0E-10	2.4 ± 5.8E-9	10.1
¹²⁵ Sbb	3.7E-3	6.6 ± 1.8E-11	5.5 ± 1.5E-11	4.5 ± 2.7E-11	10.4
¹²⁹ Pb	2.7E-2	3.4 ± 1.6E-12	5.6 ± 2.6E-10	2.4 ± 2.1E-9	8.9
¹³⁷ Cs	1.8E-2	2.3 ± 1.8E-10	4.1 ± 3.3E-10	8.9 ± 9.9E-10	9.3
Chelating agent or metal		($\mu\text{g} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)			
Iron	4.4E-4	1.5 ± 3.0E-4	3.5 ± 7.0E-11	1.1 ± 2.1E-12	13.0

a. Internal uncertainty associated with calculated results. Does not include counting statistics or other uncertainties associated with the leaching process.

b. Average values were calculated excluding zeros.

A comparison of the average absolute release rates for FitzPatrick ($8 \times 10^{-3} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$) and Peach Bottom ($7.9 \times 10^{-3} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$) indicates that the release rates are statistically the same for both waste forms. The absolute and fractional release rate data indicate that the structural stability of the waste form does not affect the release rates of picolinic acid from the waste form other than an early release due to convection and the initial increase in the surface area of the decomposed waste form. The fractional and absolute release rate data suggests that chemical mechanisms either in the waste form or in the resin itself control the release rate from the waste form. In a study by D'Angelis²³ and in other studies,^{49,50} it

was shown that waste-form chemistry and leachant chemistry (ion strength, leachant chemical composition, and leachant replacement frequency) may control leachability. These data tend to support that conclusion.

Figure 10 shows the plot of the CFRs of picolinic acid from the Peach Bottom-3 and FitzPatrick waste forms. These data show that the CFRs for the Peach Bottom-3 waste forms are similar and indicate that the release is diffusion-based on the slope of the CFR curve.^{37,38} The average CFR of picolinic acid from the Peach Bottom-3 waste forms (Table 10) is 2.0×10^{-2} , whereas for FitzPatrick, the CFR is 0.5 (90-day leach test). A comparison of the Peach Bottom-3 and

Table 10. Weighted average leach test results for Peach Bottom-3 cement-solidified waste forms.

Nuclide	CFR ^a	Release rate		Average effective diffusivity (cm ² · s ⁻¹) ^b	Leachability index ^a
		Absolute (μ Ci · cm ⁻² · s ⁻¹) ^b	Fractional (cm ⁻² · s ⁻¹) ^b		
¹⁴ C	8.8E-5	3.7 ± 4.6E-9	3.4 ± 4.1E-12	8.8 ± 6.2E-15	14.2
Includes #8 ^c	2.9E-4	5.7 ± 5.1E-9	5.3 ± 4.6E-12	1.5 ± 0.7E-14	13.5
⁵⁵ Fe	1.4E-3	7.7 ± 7.1E-10	5.1 ± 4.7E-11	6.1 ± 7.8E-12	12.2
⁵⁵ Cr	7.3E-3	1.0 ± 0.9E-9	6.8 ± 5.7E-11	9.8 ± 9.6E-12	11.4
⁶⁰ Co	1.7E-3	2.3 ± 2.1E-8	6.2 ± 5.6E-11	4.4 ± 1.2E-12	11.4
⁶⁰ Mn	2.1E-3	2.6 ± 1.9E-8	7.0 ± 5.0E-11	5.2 ± 1.6E-12	11.2
⁶³ Ni	5.9E-3	1.5 ± 1.3E-9	2.2 ± 2.1E-10	5.5 ± 2.2E-11	10.4
⁶⁵ Zn	4.0E-3	7.4 ± 5.5E-11	1.5 ± 1.0E-11	5.1 ± 1.9E-14	11.7
⁹⁰ Srd	1.9E-3	2.2 ± 2.0E-12	2.4 ± 2.1E-10	2.0 ± 1.1E-11	10.9
⁹⁰ Rb	6.9E-3	2.9 ± 2.2E-12	3.1 ± 2.4E-10	2.8 ± 1.4E-11	10.4
⁹⁹ Tc	2.6E-3	2.0 ± 1.0E-10	4.4 ± 2.2E-11	1.3 ± 1.2E-11	11.5
¹⁰³ Ru	1.0E-2	2.4 ± 1.2E-10	5.2 ± 2.7E-11	1.6 ± 1.5E-11	11.0
¹²⁹ I	7.7E-2	2.1 ± 2.1E-11	3.5 ± 3.5E-9	2.7 ± 2.1E-8	8.1
¹³⁷ Cs	6.0E-2	5.1 ± 2.1E-12	8.4 ± 4.1E-10	5.5 ± 3.2E-9	8.4
Chelating agent or metal		(μ g · cm ⁻² · s ⁻¹)			
Chromium	1.6E-2	1.2 ± 1.0E-5	4.8 ± 3.8E-10	3.8 ± 0.6E-10	9.4
Iron ^d	4.6E-5	2.6 ± 0.7E-6	5.9 ± 1.5E-13	3.2 ± 0.9E-15	14.4
⁵⁵ Cr	1.8E-4	2.8 ± 0.8E-6	6.4 ± 1.8E-13	3.5 ± 1.1E-15	13.9
Cobalt	4.2E-2	4.3 ± 3.4E-6	1.3 ± 1.0E-9	3.5 ± 1.9E-9	8.6
Nickel	2.9E-2	1.4 ± 1.2E-6	9.3 ± 8.1E-10	1.2 ± 0.3E-9	8.9
Zinc ^d	8.2E-3	9.7 ± 8.5E-6	2.1 ± 4.6E-10	1.9 ± 0.8E-10	9.8
Boron ^d	2.7E-2	7.1 ± 4.3E-6	1.6 ± 0.9E-9	3.3 ± 0.9E-9	8.4
Picolinic acid	2.0E-2	7.9 ± 5.8E-3	5.8 ± 4.3E-10	5.4 ± 0.9E-10	9.3

a. Simple average based on data in Tables 7 through 9.

b. Error weighted average based on data in Tables 7 through 9.

c. Includes the results from sample #8.

d. Average values were calculated excluding zeros.

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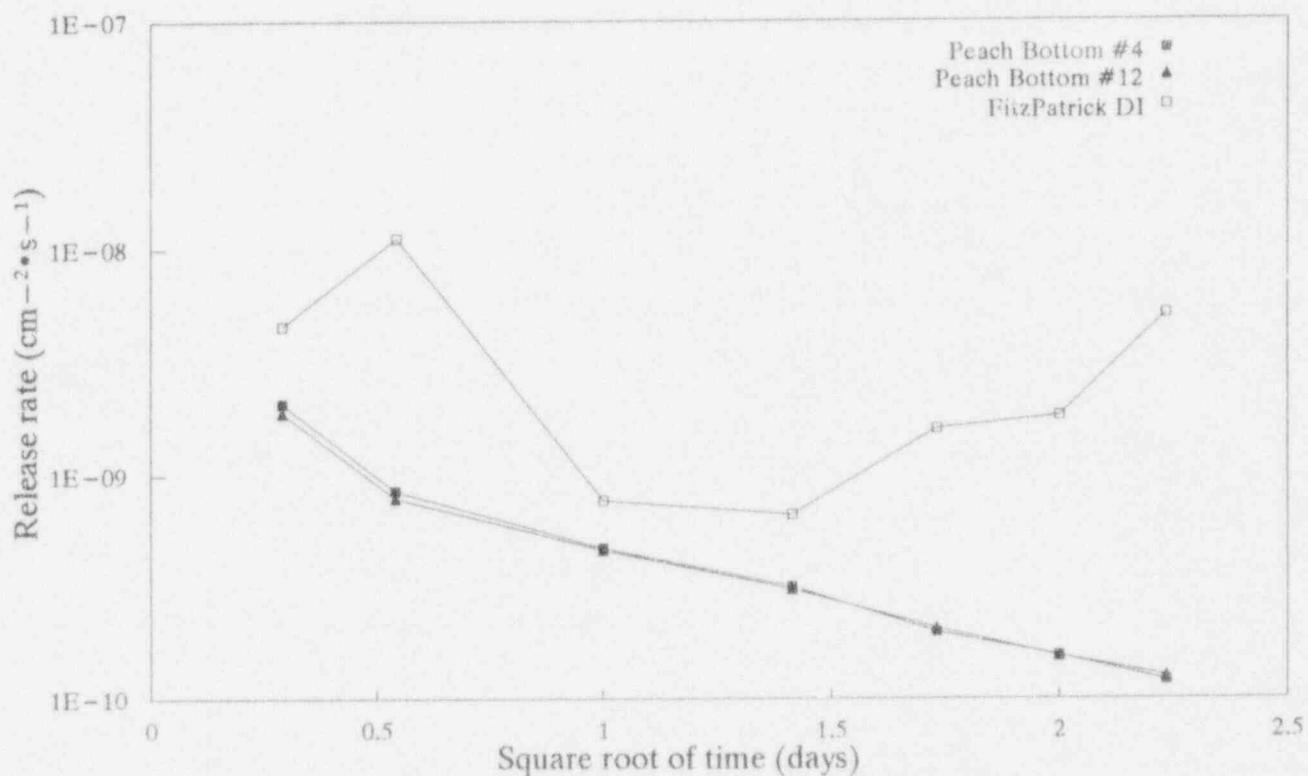


Figure 9. Fractional release rates of picolinic acid.

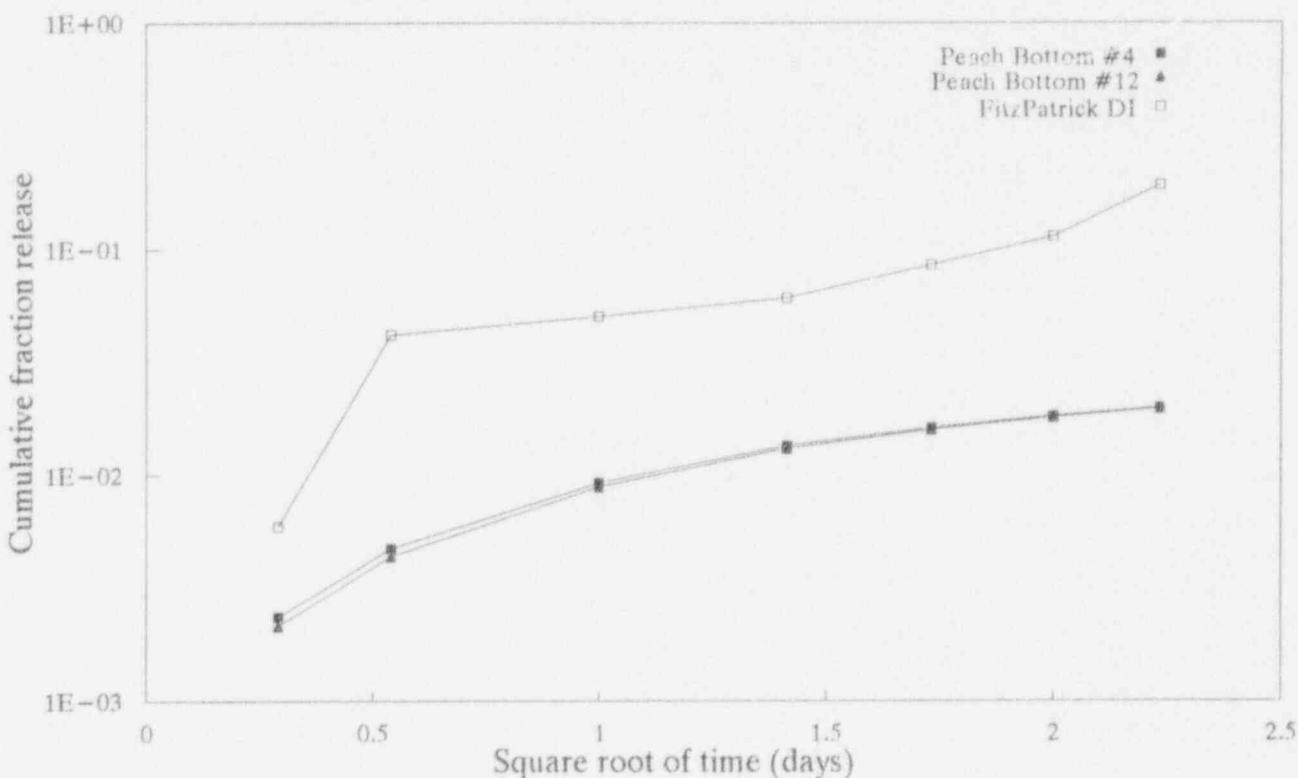


Figure 10. Cumulative fraction release of picolinic acid.

FitzPatrick data indicates that the slope of the diffusional release curves are similar for both waste forms. These data confirm the conclusion developed from the fractional release rate data that the diffusional release (following the high-rate convective release) is similar for both intact and decomposed waste forms.

The average effective diffusivities and leachability indexes for picolinic acid are shown in Tables 7 to 10. The leachability indexes for picolinic acid from the Peach Bottom samples as shown in Appendix D range from 9.1 to 9.5, and are above the regulatory requirement of 6.0. The average leachability index is 9.3, as shown in Table 10.

Decontamination Radionuclide Releases

The releases of radionuclides from the Peach Bottom-3 cement solidified waste forms is dependent not only on the physical and chemical characteristics of the waste form but on the chemistry of the radionuclide. The primary radionuclides from the decontamination process are ^{55}Fe , Co^{60} ,

and ^{63}Ni . They are transition metals and form coordination complexes (i.e., complex hybrid bonds between metals and anions, cations, or molecules) with chelating agents that may keep these metals in solution and therefore potentially increase their mobility from a waste form. Other radionuclides found in the waste forms, ^{90}Sr , ^{14}C , ^{99}Tc , ^{129}I , and ^{241}Pu , have varying chemical characteristics that are discussed below.

The fractional release rates for ^{55}Fe , Co^{60} , and ^{63}Ni are shown in Figures 11 through 13. Examination of ^{55}Fe data indicates that there are variations in the fractional release rates for sample #4 and that the fractional release rates from sample #8 are generally higher than those from the other two samples. The fractional release rates from sample #4 at leach times of 2 and 3 days are lower than those for samples #8 and #12. Two days into the leach test, the release rate from sample #4 drops to about $10^{-13} \text{ cm}^2 \cdot \text{s}^{-1}$. However, examination of the average fractional release rates for samples #4 and #12, as shown in Tables 7 and 8, indicates that they are statistically

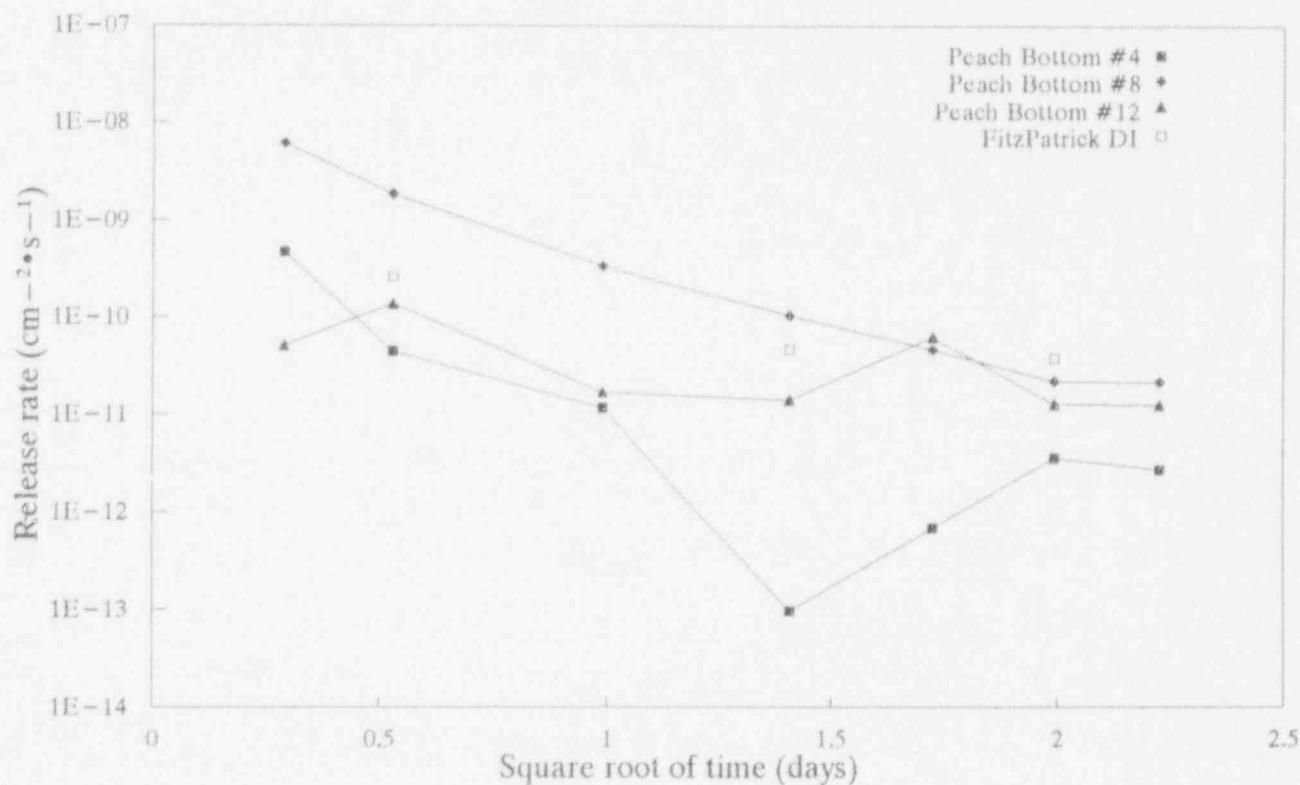


Figure 11. Fractional release rates of ^{55}Fe .

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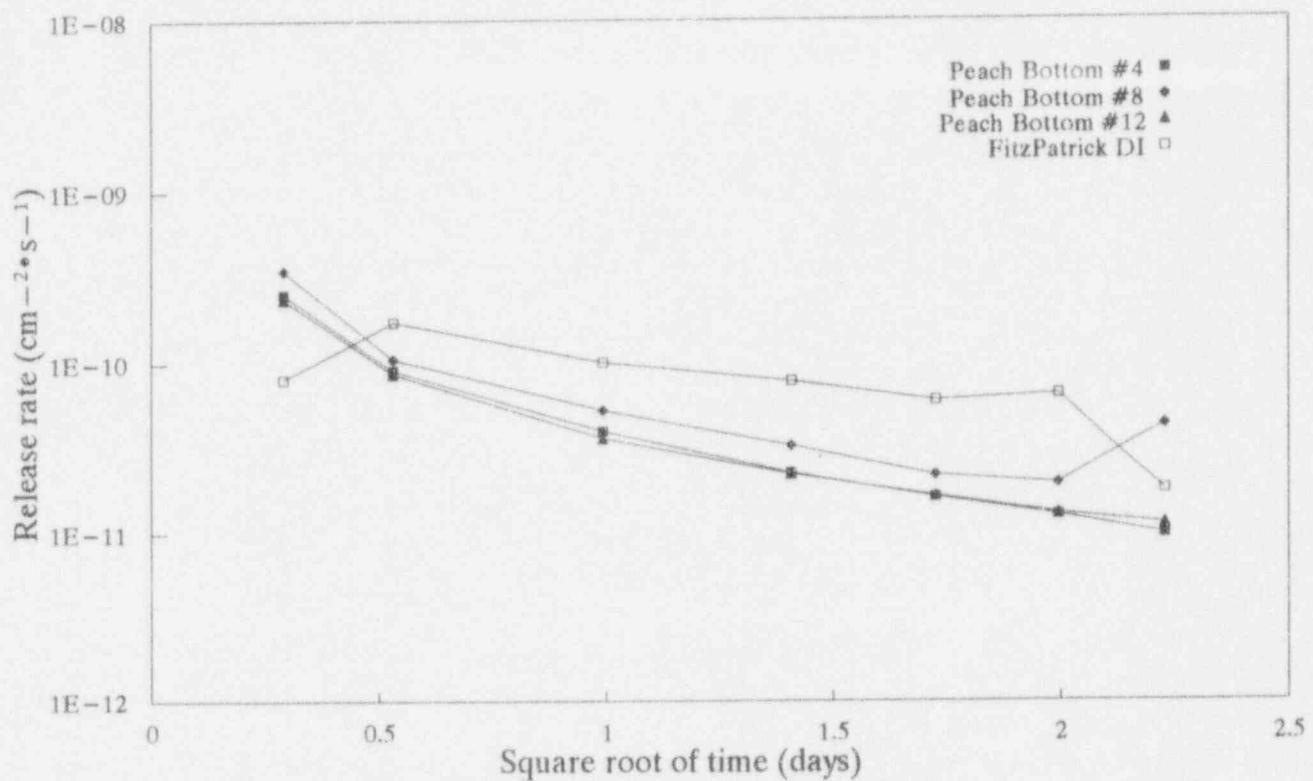


Figure 12. Fractional release rates of ^{60}Co .

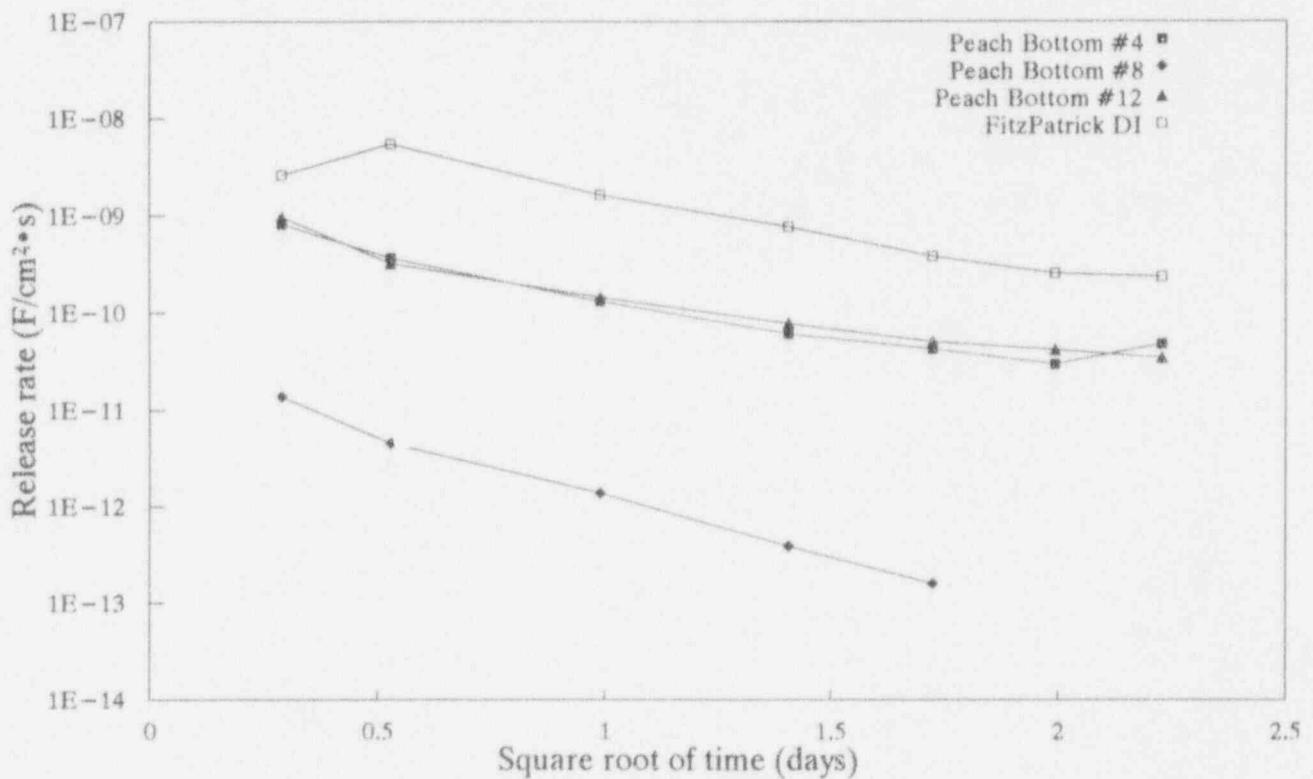


Figure 13. Fractional release rates of ^{63}Ni .

the same and have a weighted average value of $5.1 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$ as shown in Table 10. When the weighted average fractional release rate for all samples is calculated (including the value of $1.2 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$ for sample #8), it is $6.8 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$, as shown in Table 10. This weighted average fractional release rate for all samples is statistically the same as that for samples #4 and #8 because of the relatively small uncertainties associated with the sample #4 and #8 results and because of the large uncertainty ($2.1 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$) associated with the average fractional release rate of ^{55}Fe from sample #8. This large uncertainty results in the weighted average being dominated by the smaller uncertainties associated with samples #4 and #12. Consequently, when large uncertainties are present in the data, the weighted average will be dominated by the smaller uncertainty. Comparisons in which only the larger uncertainty overlaps the less-certain value are not considered to be statistically significant in this assessment. No reason for the higher fractional release rates from sample #8 has been identified.

In comparison, the average fractional release rate of ^{55}Fe from the FitzPatrick sample is $7 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$. Consequently, the weighted average fractional release rate from the Peach Bottom-3 samples is statistically the same as that of the FitzPatrick sample. The similarity of these data suggests that the fractional release rate of ^{55}Fe is not affected by the structural stability of the waste form (remember that the FitzPatrick samples fell apart during leaching). A comparison of the Peach Bottom-3 data with the weighted average fractional release rate of picolinic acid ($5.8 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$) indicates that the chelating agent is being released faster than ^{55}Fe and that retention mechanisms may hold ^{55}Fe in the waste form.

The weighted average of the absolute release rates of ^{55}Fe for the three Peach Bottom-3 waste forms ($1.0 \times 10^{-9} \mu\text{Ci/cm}^2 \cdot \text{s}$), as shown in Table 10, is statistically the same as that measured for the FitzPatrick waste form ($7 \times 10^{-10} \mu\text{Ci/cm}^2 \cdot \text{s}$) if a similar uncertainty is applied to the FitzPatrick data. This suggests that the

concentration of the radionuclide in the waste form does not affect the absolute release rate because the concentration of ^{55}Fe in the FitzPatrick waste form is about a factor of four less than that in the Peach Bottom-3 waste form. These absolute release rates indicate that the release rate of ^{55}Fe from LOMI waste forms may be independent of the inventory in the waste form and that other factors, besides diffusion, may control the release rate from the waste form.

Figure 12 shows the fractional release rates of ^{60}Co from the Peach Bottom-3 samples. These data are similar for all three Peach Bottom-3 waste-form samples and suggest that the observed variations in the leach rates for ^{55}Fe are not problems associated with the leaching process but are indicative of the behavior of ^{55}Fe . A comparison of the ^{60}Co fractional release rates for Peach Bottom-3 with the FitzPatrick data as shown in Figure 12 indicates that the ^{60}Co fractional release rate for FitzPatrick is initially lower (first leach period) and then goes above the Peach Bottom-3 rates for the next 4 days. The weighted average of the fractional release rates of ^{60}Co for all Peach Bottom-3 samples as shown in Table 10 is $7 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$, which is again statistically the same as that for the FitzPatrick sample ($7 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$). It should be noted that the weighted average of the fractional release rates for ^{55}Fe (Table 10) are the same as those for ^{60}Co and indicates similar release behavior for both radionuclides.

Inspection of Figure 12 suggests that the average fractional release rate of ^{60}Co for FitzPatrick should be higher than that for Peach Bottom-3. However, the high fractional release rates shown in Figure 12 for FitzPatrick occur only during the first 5 days of leaching (excluding the first leach period) and after this initial release, the rate drops to about $1.8 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$ for the duration of the 90-day leach test. As noted above, the average fractional release rate of ^{60}Co from the FitzPatrick sample over the 90 days is statistically the same as the weighted average fractional release rate from the Peach Bottom-3 samples as shown in Table 10. These data again indicate that after an initial convective release from the degraded waste form, the fractional release rate from the

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intact waste form is the same as that from the degraded waste form. This again suggests that mechanisms other than structural stability and diffusion may be controlling the release process. A comparison of the weighted fractional release rates of ^{60}Co as shown in Table 10 with those of picolinic acid again indicates, similar to the ^{55}Fe data, that the weighted average fractional release rates of ^{60}Co are less than those observed for picolinic acid and, similarly, that ^{60}Co is being released at a lower rate.

The average absolute release rates for ^{60}Co from the Peach Bottom-3 samples, as shown in Table 7 to 9, are statistically the same and have a weighted average value of $(2.6 \times 10^{-8} \mu\text{Ci}/\text{cm}^2 \cdot \text{s})$ as shown in Table 10. This weighted average absolute release rate is statistically higher than those for ^{55}Fe and ^{63}Ni and is possibly due to the higher concentration of this radionuclide in the Peach Bottom-3 samples. The similarity of the weighted average fractional release rates for the transition metal radionuclides and the difference in the absolute release rates suggests that the release rate of ^{60}Co is dependent on the concentration in the waste form.

Figure 13 shows the fractional release rates for ^{63}Ni and indicates a difference between the samples analyzed at one laboratory (#4 and #12) and a sample analyzed at another laboratory (#8). This difference is about two orders of magnitude, which is well above the statistical differences that might be expected for this type of analysis. These data indicate that the sample #8 ^{63}Ni data, which are much lower than that for the other two samples, are suspect. Discussions with the laboratory provide no indications as to why this discrepancy should exist. Consequently, only the results for samples #4 and #12 will be discussed for this radionuclide.

The weighted average fractional release rate of ^{63}Ni for samples #4 and #12 is about $2.2 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$. A comparison of the sample #4 and #12 data with the average fractional release rate of ^{63}Ni from the FitzPatrick sample ($6 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$) indicates that the weighted average fractional release rate from the Peach Bottom-3

samples is about a factor of three less than that for the FitzPatrick sample. This difference is not within the uncertainty listed in Table 10 for the weighted average fractional release rate and indicates that ^{63}Ni is being released at a faster fractional release rate from the FitzPatrick waste form than from the Peach Bottom-3 waste forms. This is in contrast with the ^{55}Fe and ^{60}Co data, which are statistically the same as the FitzPatrick results. No reason for the enhanced fractional release rate of ^{63}Ni from the FitzPatrick sample has been identified, although it may be due to the different release characteristics of the degraded waste form.

The weighted average absolute release rate of ^{63}Ni for the Peach Bottom-3 samples (#4 and #12) is $1.5 \times 10^{-9} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$, which is approximately an order of magnitude less than that for the FitzPatrick sample. This rate is statistically the same as the weighted average absolute release rate of ^{55}Fe . However, as indicated in Table 10, although the absolute release rates are statistically the same, the fractional release rate of ^{63}Ni is a factor of three higher, which indicates differences in the behavior of the two radionuclides.

A comparison of the absolute and fractional release rate data for the three decontamination radionuclides indicates that the structural stability of the waste form does not appear to significantly affect releases from the waste form. For Peach Bottom-3, the weighted average fractional release rate is $6.8 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$ for ^{55}Fe , $7 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$ for ^{60}Co , and $2.2 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$ for ^{63}Ni (samples #4 and #12). These data indicate that release rates are within a factor of four, but that ^{63}Ni probably has a higher release rate than the other transition metal radionuclides. The fractional release rate of picolinic acid is about $6 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$ and is statistically higher than that of ^{63}Ni and the other decontamination radionuclides. The higher fractional release rates associated with ^{63}Ni may be due to the increased stability of nickel complexes, as the Irving-Williams correlation^{51,52} indicates that the stability of transition metal complexes fall in the order $\text{Ni}^{II} > \text{Co}^{II} > \text{Fe}^{II}$. These data suggest that the stability of the nickel complex with a chelating agent may result in the higher average fractional

release rates of ^{63}Ni relative to the other transition metals.

The CFR plots for the transition metals are shown in Figures 14 through 16. Inspection of the ^{55}Fe data in Figure 14 and Tables 7 to 9 indicates that the cumulative fractional release ranges from 8.6×10^{-4} for sample #4 to 1.9×10^{-2} for sample #8. The CFR for sample #8 is about an order of magnitude higher than that for the other two samples. This higher CFR is due to high release rates during the first three leach periods (see Figure 11). Following this initial release, the rates are similar to those observed for samples #4 and #12. The average CFR for ^{55}Fe from Peach Bottom-3 samples #4 and #12 is 1.4×10^{-3} and 7.3×10^{-3} for all samples (includes #8). No explanation for the initial high release rates associated with sample #8 have been identified. Figure 14 further indicates that the CFRs for the Peach Bottom-3 samples bound the CFR for ^{55}Fe for FitzPatrick. These data further indicate that

the release rates from a waste form are not dependent on the structural stability of the waste form.

Figure 16 shows the CFRs for ^{60}Co . The CFRs for all Peach Bottom-3 samples are similar (i.e., from 1.7×10^{-3} to 3×10^{-3}) and indicate a higher CFR for sample #8. The average CFR of ^{60}Co for the Peach Bottom-3 samples is 2.1×10^{-3} , which is less than the CFR of ^{60}Co from the FitzPatrick sample (5.9×10^{-3} through 5 days). These data indicate that the CFR from the degraded waste form is higher. However, as discussed previously, there was an initial high release probably due to convection during the breakup of the waste form that was followed by lower release rates. Consequently, it would be expected that the CFRs of both the Peach Bottom-3 and FitzPatrick waste forms would probably be similar at 90 days. These data suggest that the 90-day leach test is probably more indicative of the behavior of the waste form than is the 5-day test particularly when degrading waste forms are being tested.

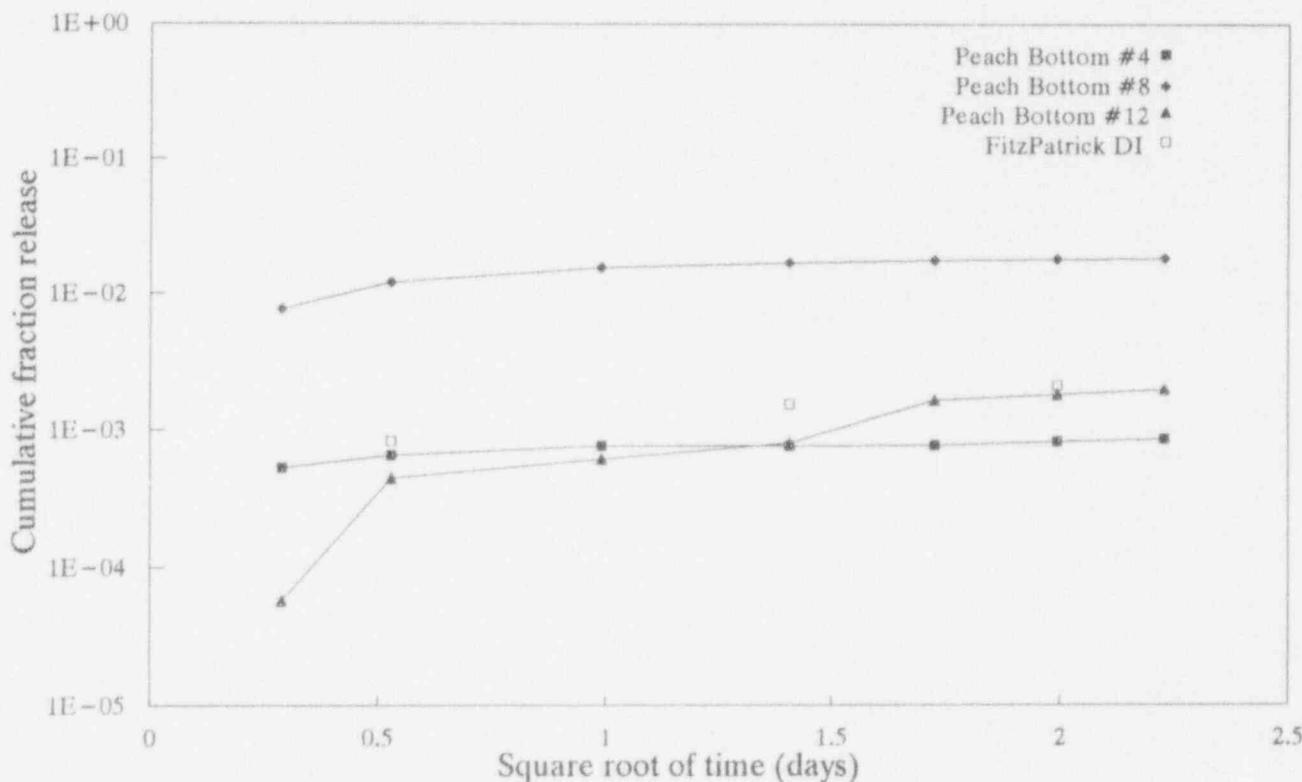


Figure 14. Cumulative fraction release of ^{55}Fe .

Experimental Results

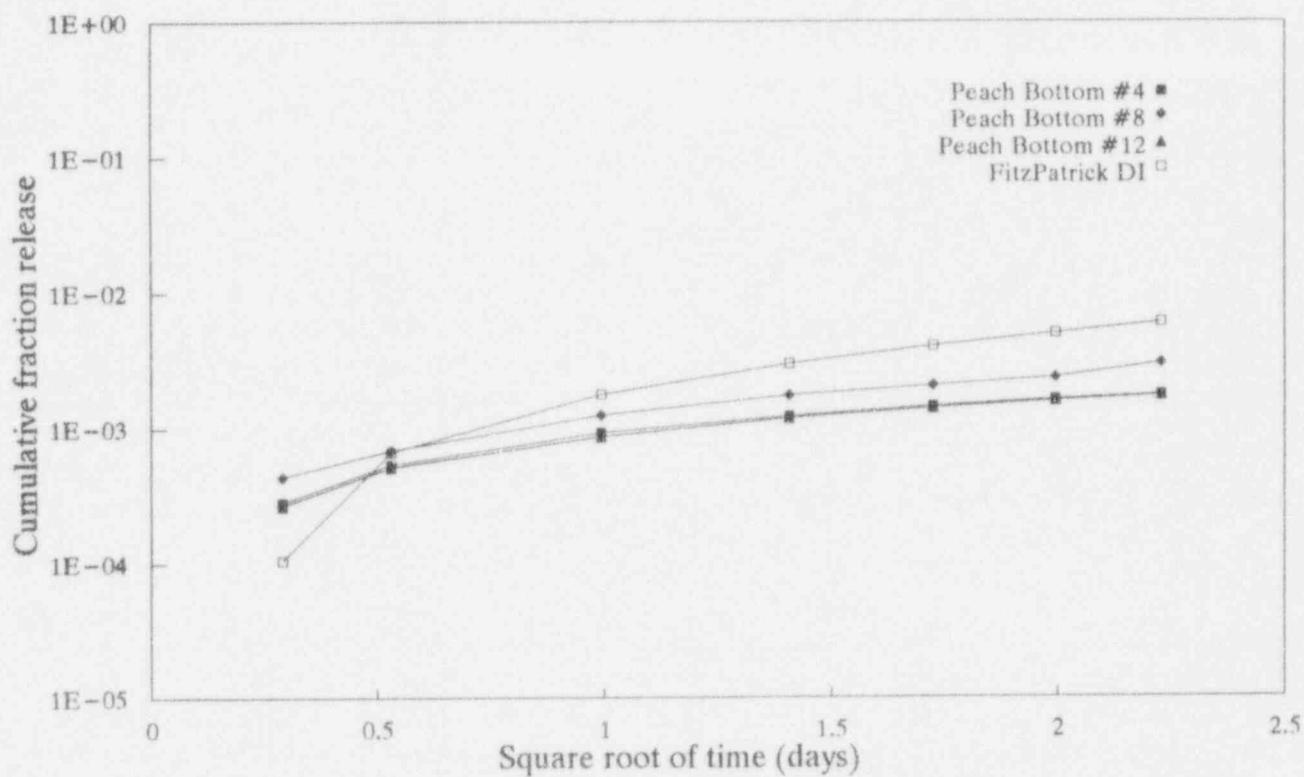


Figure 15. Cumulative fraction release of ^{60}Co .

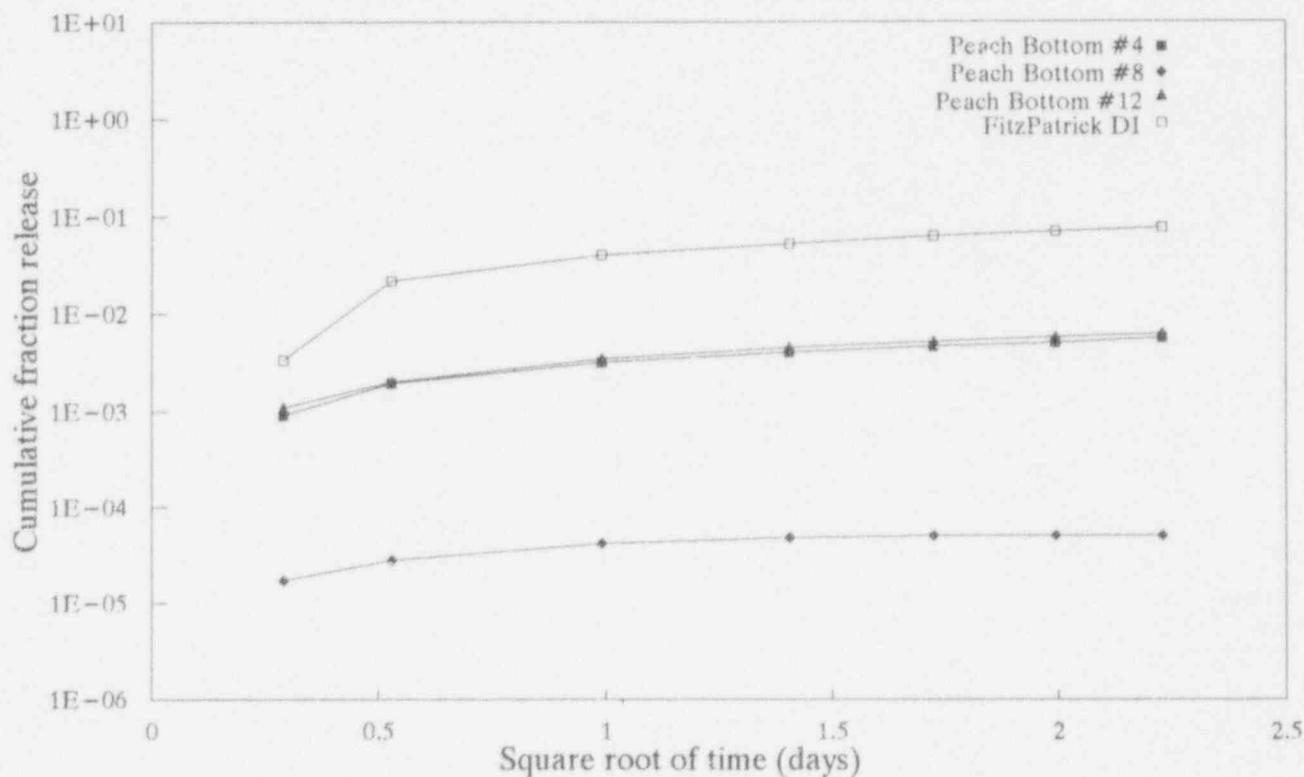


Figure 16. Cumulative fraction release of ^{63}Ni .

Figure 17 shows the CFRs for ^{63}Ni and indicates a low CFR for sample #8 (5×10^{-5}) in contrast to the average of samples #4 and #12 (5.9×10^{-3}). As discussed above, no reason for the low fractional release rates and low CFR from sample #8 have been identified. The average CFR for ^{63}Ni (samples #4 and #8) is 5.9×10^{-3} . This is higher than those for ^{55}Fe and ^{60}Co .

A comparison of the average CFRs for all decontamination radionuclides indicates that ^{55}Fe and ^{60}Co are statistically the same if a 100% uncertainty is applied. However, both are lower than the CFR for ^{63}Ni and again suggests that ^{63}Ni is being released at a higher rate for the reasons discussed previously.

Average effective diffusivities and leachability indexes are listed in Tables 7 through 9 for each sample and are averaged in Table 10. The average effective diffusivities of the transition metals range over an order of magnitude from ^{60}Co ($4.4 \times 10^{-12} \text{ cm}^2 \cdot \text{s}^{-1}$) to ^{63}Ni ($5.5 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$). In addition, the average leachability

indexes range from 10.4 to 12.2 with ^{55}Fe having the highest leachability index and ^{63}Ni having the lowest. These data again confirm the apparent increased mobility of ^{63}Ni .

Other Radionuclide Releases

Other radionuclides for which analyses of the waste form and chelating agents were performed and measurable results were obtained were ^{14}C , ^{99}Tc , ^{129}I , ^{90}Sr , and ^{241}Pu . Carbon-14, ^{99}Tc , and ^{129}I were detectable in most leachate samples, whereas ^{90}Sr was detectable in only a few of the samples, and ^{241}Pu was not detectable in the leachates. Figures 17 through 19 show the fractional release rates for ^{14}C , ^{99}Tc , and ^{129}I . The ^{14}C data shown in Figure 17 indicate that samples #8 and #12 exhibit normal diffusion-driven release characteristics, whereas sample #4 is more variable. The average fractional release rates of ^{14}C for the three samples as shown in Tables 7 to 9 range from 2.5×10^{-12} to $2.3 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$, an order of magnitude difference. Sample #8 has the highest average fractional release rate. From

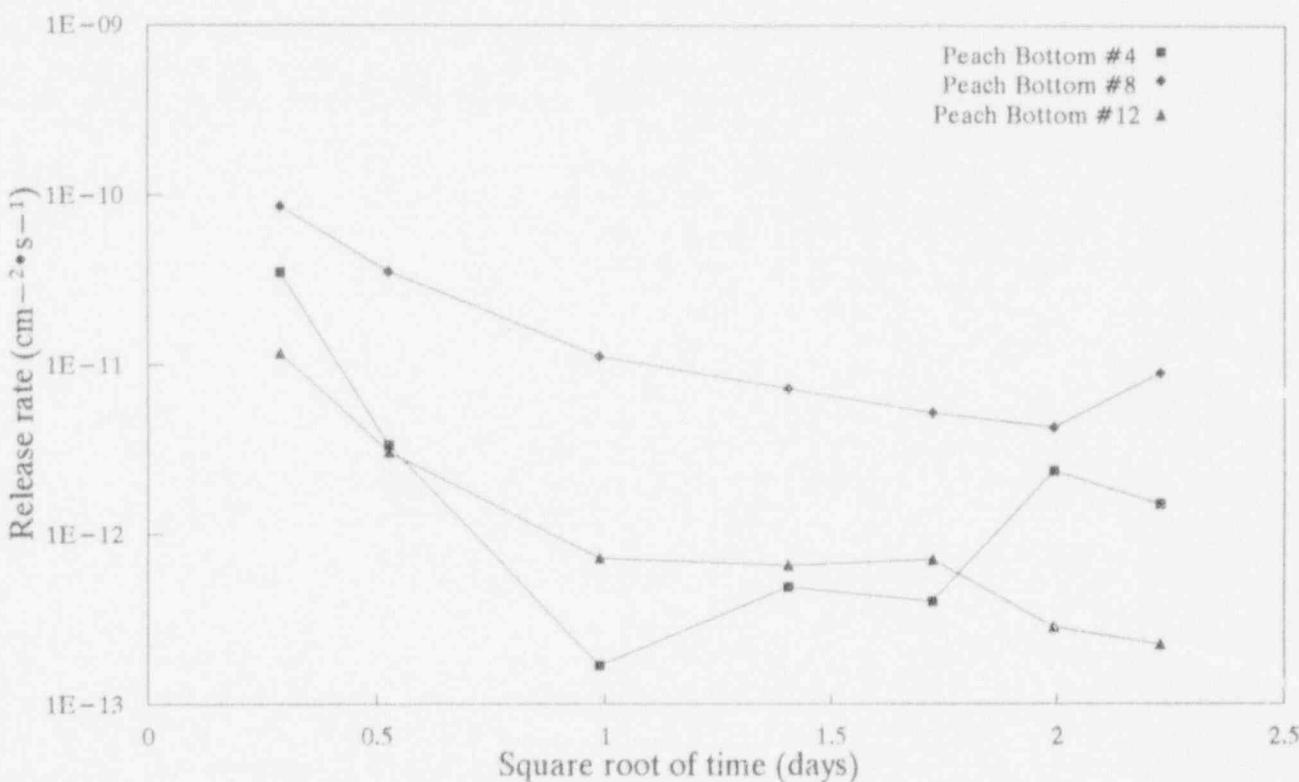


Figure 17. Fractional release rates of ^{14}C .

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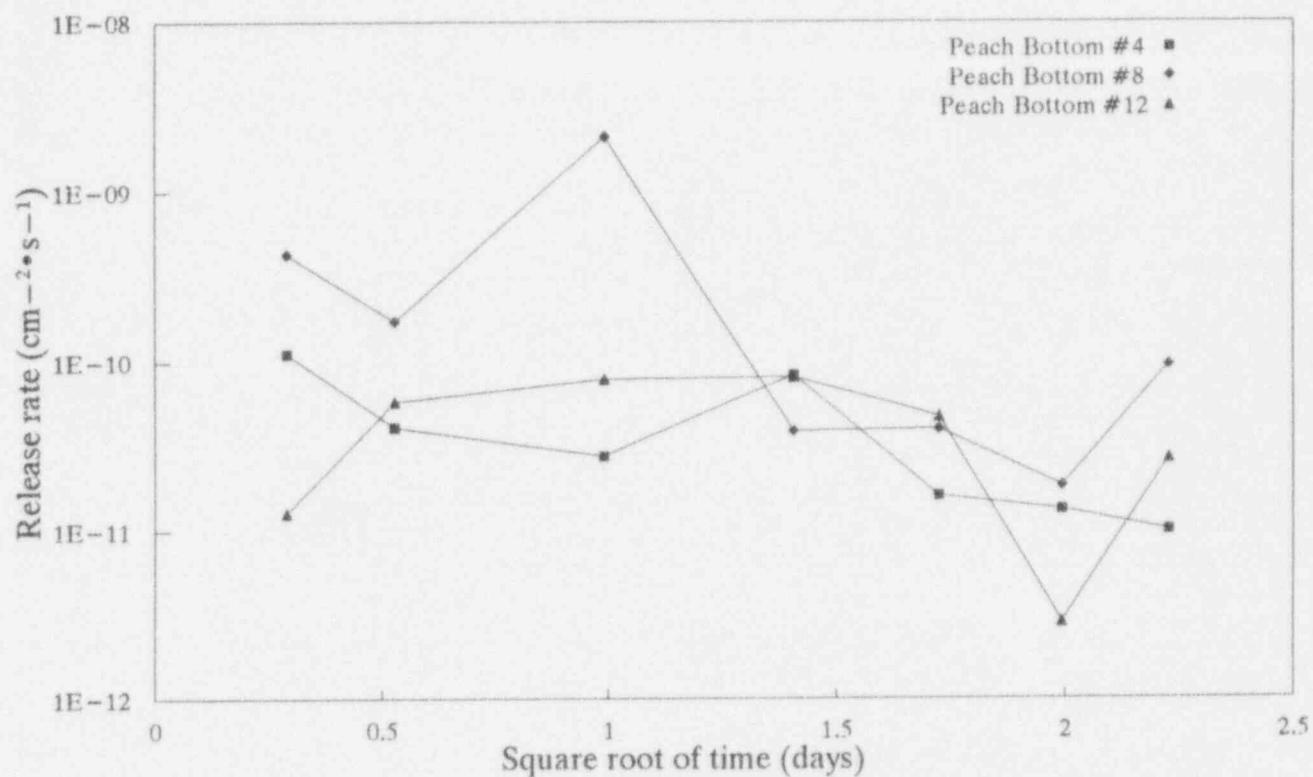


Figure 18. Fractional release rates of ^{99}Tc .

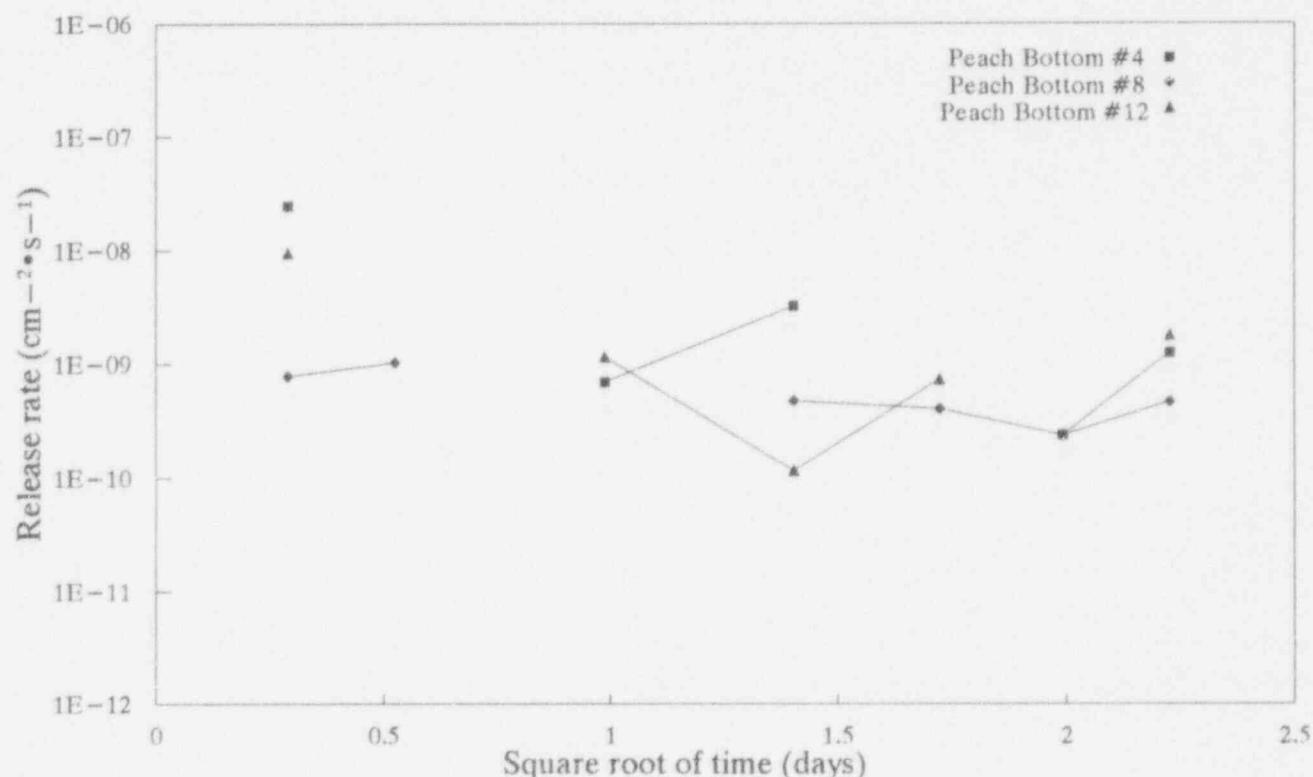


Figure 19. Fractional release rates of ^{129}I .

Table 10, the weighted average fractional release rate of ^{14}C for all samples is $5.3 \times 10^{-12} \text{ cm}^2 \cdot \text{s}^{-1}$ and is statistically the same as that for samples #4 and #12 alone. Consequently, the weighted average fractional release rate for all samples will be used for comparison purposes. The weighted average fractional release rates of ^{14}C are the lowest of any of the radionuclides measured. This is consistent with the results of Krishnamoorthy,⁵³ who attributes the low release rate of ^{14}C measured in his study to the formation of insoluble hydrates and carbonates that slow the release of this radionuclide. Further, he suggests that the fractional release rate of ^{60}Co should be slower than ^{14}C . This is inconsistent with our results in which ^{60}Co is released at a weighted average fractional release rate 20 times faster than ^{14}C . This enhanced release rate may be due to chelating agent effects that are increasing the mobility of ^{60}Co .

In contrast to the fractional release rates, the absolute release rates of ^{14}C from the Peach Bottom-3 samples are near the highest for any radionuclide and are only less than those for ^{60}Co . The weighted average of the ^{14}C absolute release rates as shown in Table 10 is $5.7 \times 10^{-9} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$ for all samples. This is about 20% of the weighted average absolute release rate of ^{60}Co , which is released at a faster absolute rate than any other radionuclide. However, the stable metals are released at faster absolute release rates than any of the radionuclides. The large difference between the fractional and absolute release rates of ^{14}C is due to the relatively large inventory of ^{14}C in the waste form (approximately 60% of the total activity) and suggests that the release rate of this radionuclide is controlled by waste-form chemistry and not diffusion.

The ^{99}Tc fractional release rate data shown in Figure 18 are relatively constant, with the exception of sample #8, which has a high release rate for the first day-long leach period. Further, the release rates at the end of the leach test are similar to those at the beginning. The weighted average fractional release rate of ^{99}Tc is $5.2 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$ for all samples and is statistically the same as that for samples #4 and #12 alone. This

weighted average fractional release rate is also statistically the same as that for ^{55}Fe and ^{60}Co and suggests similar chemical and release rate behavior for ^{99}Tc . This might be expected because technetium is a metal and would be expected to form complexes with organic compounds such as chelating agents.⁵¹

The average absolute release rates for ^{99}Tc range over an order of magnitude from $2.0 \times 10^{-10} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$ for samples #4 and #12 to $1.9 \times 10^{-9} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$ for sample #8. The weighted average for all samples is $2.4 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$. This is within a factor of three of the ^{55}Fe results and suggests some similarity in release rate behavior.

Figure 19 shows the fractional release rates for ^{129}I . As shown in the figure, some leachates did not have measurable concentrations of ^{129}I . The average fractional release rates range from $5.6 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$ for sample #8 to $5.9 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$ for sample #4. The weighted average for all samples is $8.4 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$. This rate is approximately a factor of two greater than the average fractional release rate of ^{137}Cs ($4.1 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$) from sample #8. Iodine, an anion, would be expected to diffuse similarly to other ionic elements such as cesium and its release would not be expected to be affected by chelating agent effects.^{19,54} The fractional release rates of iodine and cesium are the highest of all radionuclides present in the waste form and are statistically the same as that for picolinic acid ($5.8 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$), which suggests that the maximum diffusional fractional release rate from the waste form is between 5×10^{-10} and $1 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$. Krishnamoorthy⁵³ observed similar behavior except that he found that cesium was released at a faster rate than the iodine. The observed behavior, as shown in Figure 19, suggests that anion release rates may not be affected by other mechanisms besides diffusion and that they are released at higher rates than other radionuclides.

The weighted average absolute release rate for ^{129}I is $5.1 \times 10^{-12} \mu\text{Ci}/\text{cm}^2 \cdot \text{s}$. It is near the lowest for all radionuclides and is only slightly higher than that for ^{90}Sr . This low absolute release

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rate is an artifact of the low inventory present in the waste form.

Figures 20 to 22 show the CFRs for ^{14}C , ^{99}Tc , and ^{129}I . Figure 20 indicates relatively standard diffusion behavior for ^{14}C for all samples. The average CFR for ^{14}C for all samples (2.9×10^{-4}) is lower than that for all other radionuclides. This may be due to the fact that, as previously discussed, ^{14}C may be retained better in the waste form due to the formation of insoluble hydrates and carbonates.

Figure 21 shows the CFR of ^{99}Tc from the Peach Bottom-3 waste forms. In this case, the average CFR for all samples is 1.0×10^{-2} and is (2.3×10^{-3}) for samples #4 and #12. These CFRs are representative of the CFRs of the transition metals and suggests similar behavior for this radionuclide as discussed previously.

Figure 22 shows the CFRs of ^{129}I from the Peach Bottom-3 waste forms. Sample #4 has the highest CFR (9.6×10^{-2}) of any of the radionuclides measured, and the average CFR for ^{129}I is

also the highest at 6.0×10^{-2} . As discussed previously, this high CFR may be expected due to the ionic nature of this radionuclide.

A comparison of the CFRs for ^{14}C , ^{99}Tc , and ^{129}I indicates that ^{14}C has the lowest CFR of all radionuclides and that ^{129}I has the highest. Further, the behavior of ^{99}Tc appears to be similar to that of the transition metals such as ^{60}Co and ^{55}Fe . This behavior may be expected because ^{99}Tc is a metal that forms complexes with organic complexing agents similar to the transition metals discussed previously.⁵¹

Average effective diffusivities and leachability indexes for ^{14}C , ^{99}Tc , and ^{129}I are listed in Tables 7 to 9 for each sample and are averaged in Table 10. The weighted average effective diffusivities for ^{14}C , ^{99}Tc , and ^{129}I are 1.5×10^{-14} , 1.6×10^{-11} , and $5.5 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$, respectively. Krishnamoorthy⁵³ also measured effective diffusivities for cement-solidified waste forms, although his did not contain decontamination ion-exchange resins. For ^{14}C , he measured an

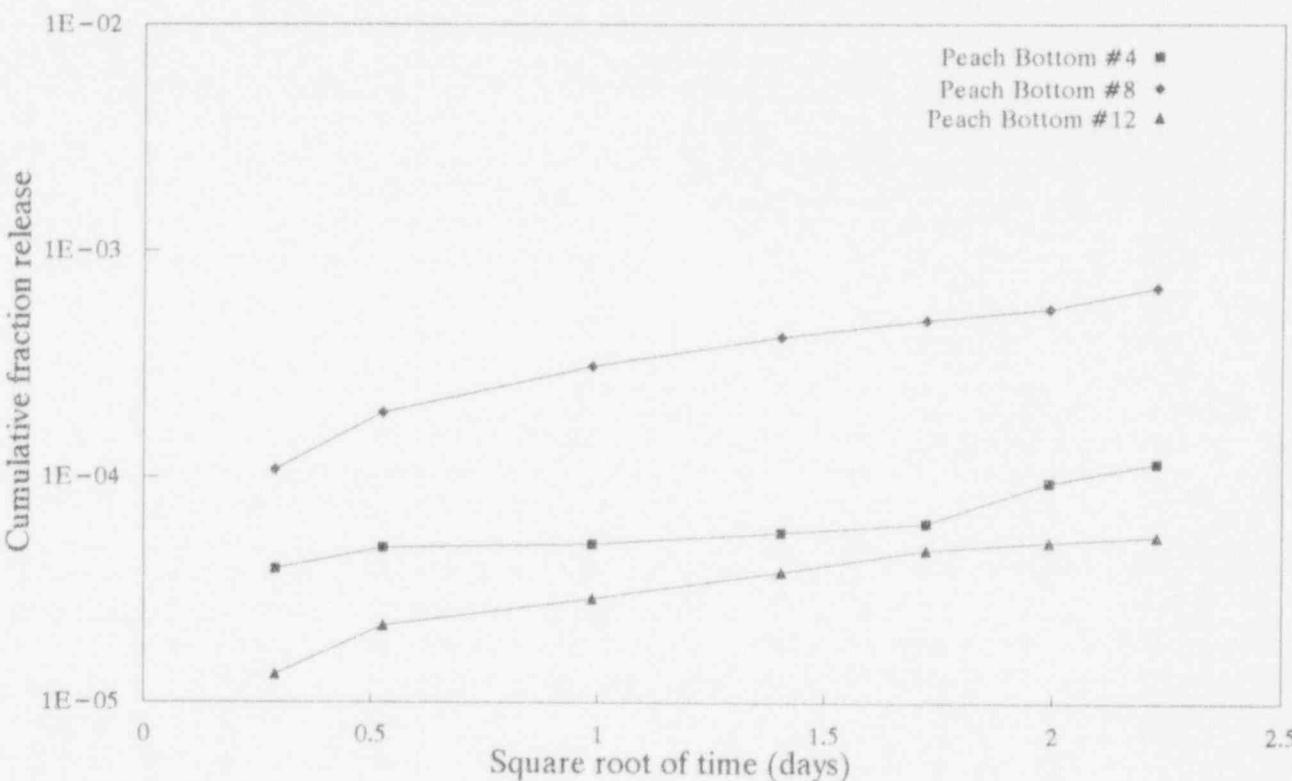


Figure 20. Cumulative fraction release of ^{14}C .

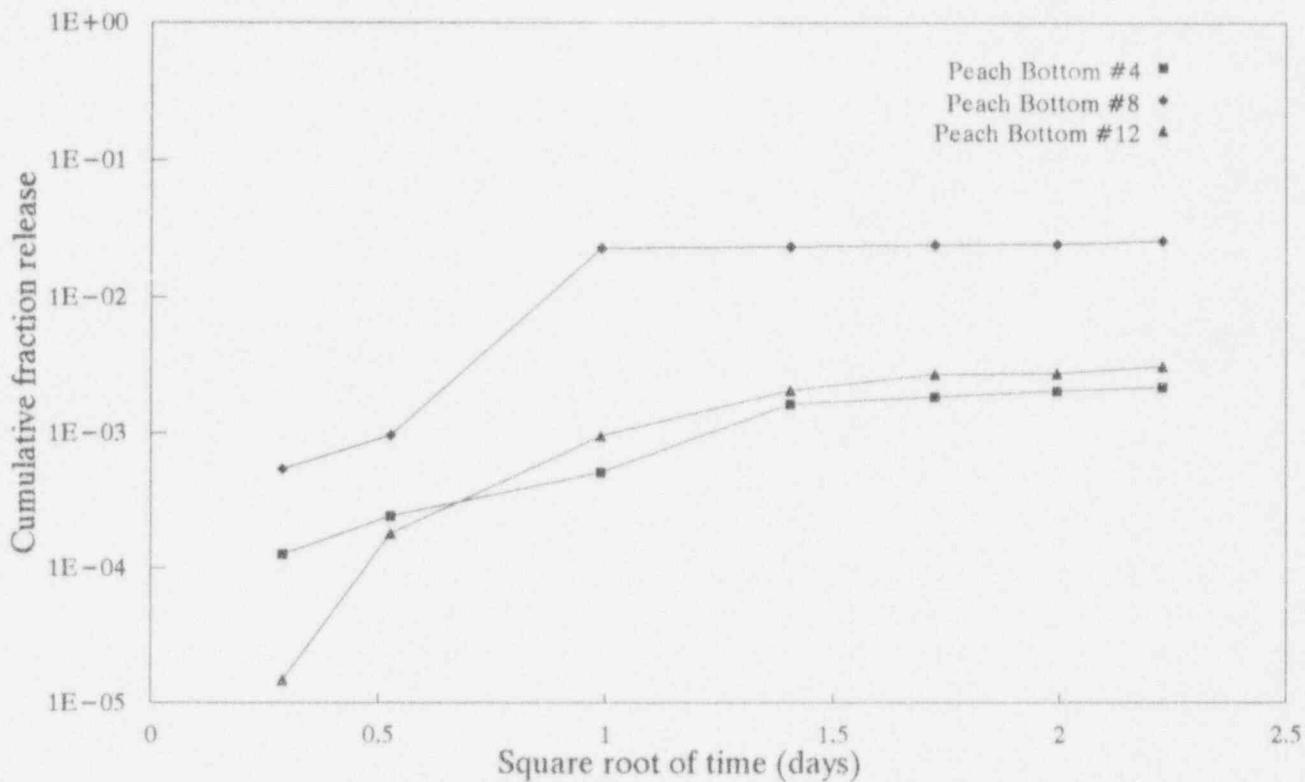


Figure 21. Cumulative fraction release of ^{99}Tc .

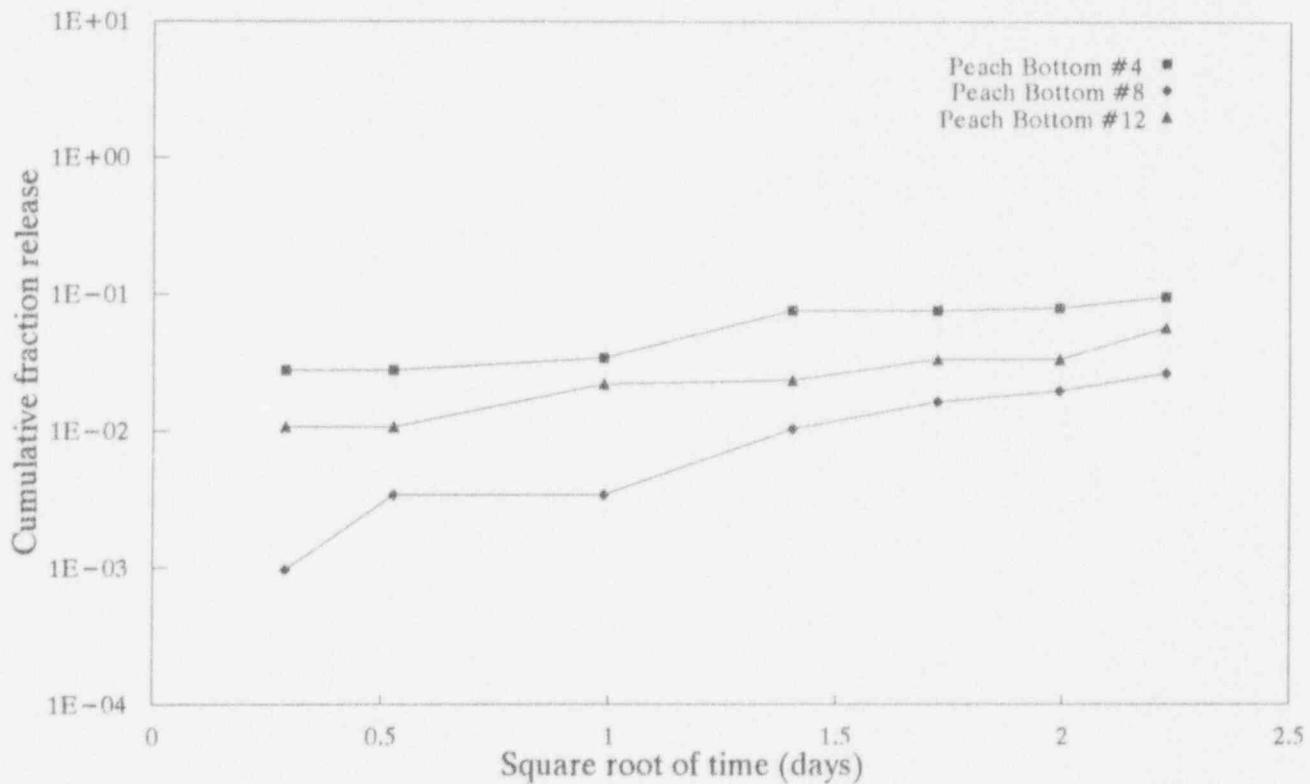


Figure 22. Cumulative fraction release of ^{129}I .

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effective diffusivity of $3 \times 10^{-13} \text{ cm}^2 \cdot \text{s}^{-1}$, which is an order of magnitude higher than the effective diffusivity of ^{14}C measured in this study. For ^{131}I , Krishnamoorthy measured an effective diffusivity of $3.4 \times 10^{-7} \text{ cm}^2 \cdot \text{s}^{-1}$, which is several orders of magnitude higher than the effective diffusivity of ^{129}I measured in this study. Consequently, it can be assumed that there are differences between the behavior of the laboratory-prepared specimens and the actual waste-form specimens used in this study.

The average leachability indexes of ^{14}C , ^{99}Tc , and ^{129}I are 13.5, 11.0, and 8.4, respectively. As expected, ^{14}C would have a high leachability index, and ^{129}I would have a low one. These data again confirm the increased mobility of ^{129}I .

Other radionuclides for which analyses were performed as part of this study were ^{90}Sr , ^{125}Sb , ^{137}Cs , and transuranics including ^{239}Pu , ^{241}Pu , ^{242}Cm , ^{244}Cm , and ^{241}Am . However, the transuranics were not detectable in any of the leachate samples; ^{125}Sb and ^{137}Cs were measurable only in sample #8; and ^{90}Sr was detected in some leachates from each sample. Less-than values were not used for calculating release rates, effective diffusivities, and leachability indexes for the transuranics because, without any actual results, these data would be misleading. In contrast, release rates, effective diffusivities, and leachability indexes were calculated for ^{125}Sb , ^{137}Cs , and ^{90}Sr . The average fractional release rates for ^{125}Sb , ^{137}Cs , and ^{90}Sr are 5.5×10^{-11} , 4.1×10^{-10} , and $3.1 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$ (weighted average). These data indicate that the release rates of ^{137}Cs and ^{90}Sr are the same. This behavior is not expected because ^{90}Sr is not ionic and would not be expected to be released at the same rate as ^{137}Cs . This observed behavior suggests that ^{90}Sr may be enhanced by chelating-agent effects.

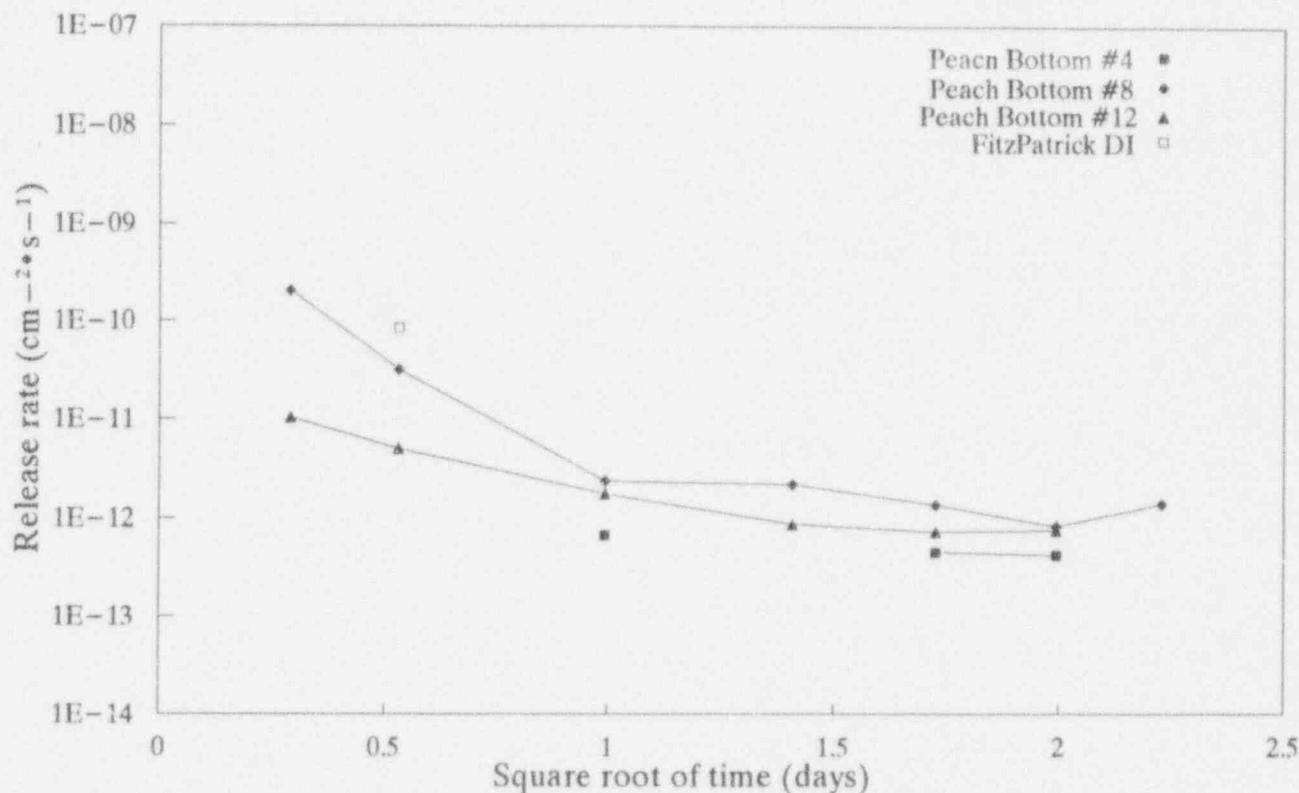
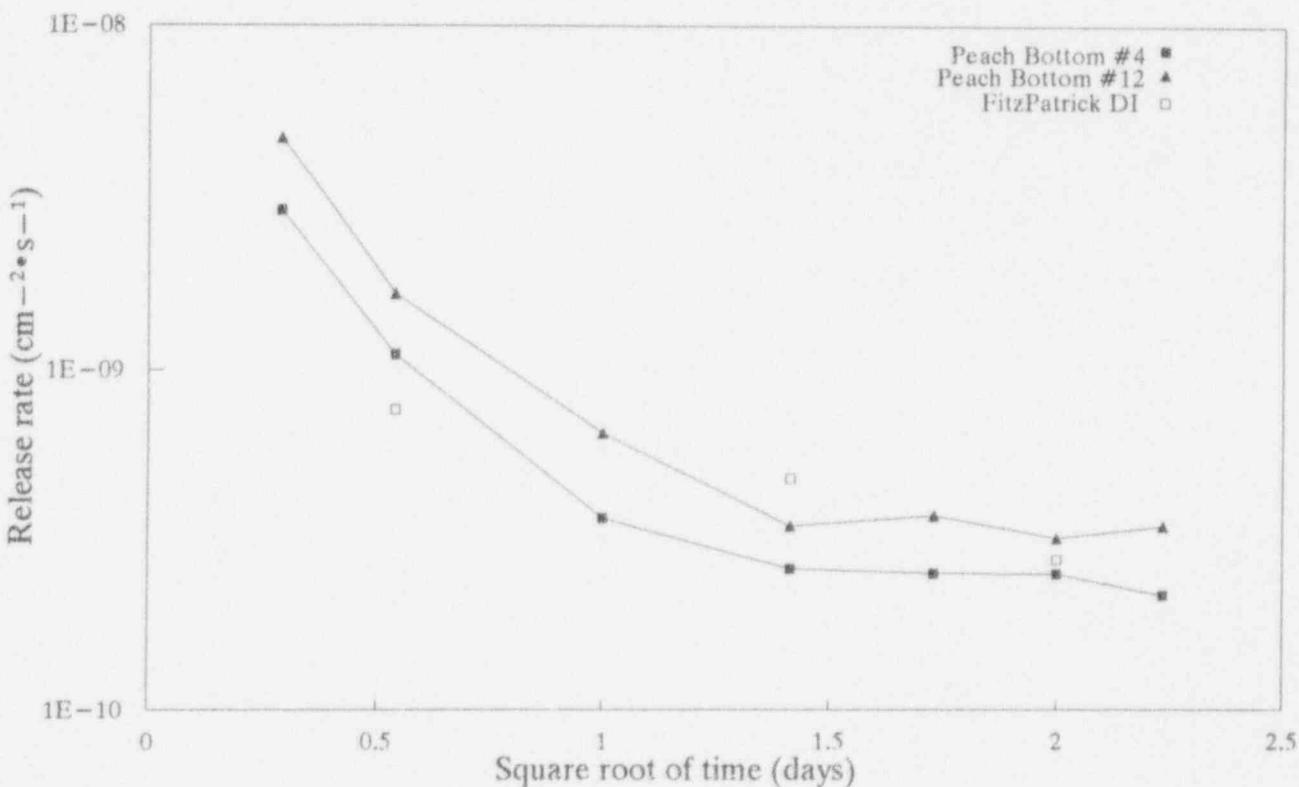
Stable Metals

Stable elements for which analyses were performed were iron, nickel, chromium, cobalt, zinc, and boron. The principal metals of interest are the iron, nickel, and cobalt because they can be used

for comparison with the releases of the decontamination radionuclides ^{55}Fe , ^{63}Ni , and ^{60}Co . Fractional release rates for iron, nickel, chromium, and cobalt are shown in Figures 23 through 26. In addition, the fractional release rates for each element from the FitzPatrick sample leached in deionized water are also shown. Inspection of Figures 23 through 25 indicates that the chromium has similar fractional-release-rate characteristics to the nickel and that the fractional release rates of nickel from the FitzPatrick sample are similar to that for Peach Bottom-3. The average fractional release rates for each of the stable metals are shown in Tables 7 to 9, and the weighted average fractional release rates are shown in Table 10.

The weighted average fractional release rate of nickel ($9.3 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$) and cobalt ($1.3 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$) are the highest at about a factor of two greater than that for chromium. In addition, they are statistically the same as that of picolinic acid. These data suggest that the releases of nickel, cobalt, and picolinic acid are probably diffusion-driven and are also being released at the highest rate possible from the waste form (approximately $1 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$). However, it also suggests that ^{63}Ni and ^{60}Co are in a different chemical form than that of the stable metals because the fractional release rate of ^{63}Ni is about 25% of the stable-nickel-weighted average fractional release rate and the fractional release rate of ^{60}Co is about 5% of the stable-cobalt-weighted average fractional release rate.

In contrast to the nickel data, the data in Figure 23 indicate that the fractional releases of iron from samples #4 and #12 are similar but different than that observed for sample #8. The weighted average fractional release of iron for all samples is $6.4 \times 10^{-13} \text{ cm}^2 \cdot \text{s}^{-1}$. This compares to an average fractional release rate of $6.8 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$ for ^{55}Fe and indicates that ^{55}Fe is released from the waste form faster than the elemental iron by two orders of magnitude. It should be noted that comparisons of the fractional release rates for iron and ^{55}Fe indicate that ^{55}Fe is released faster in the early leach periods, whereas the nickel release rate is relatively constant.

**Figure 23.** Fractional release rates of iron.**Figure 24.** Fractional release rates of nickel.

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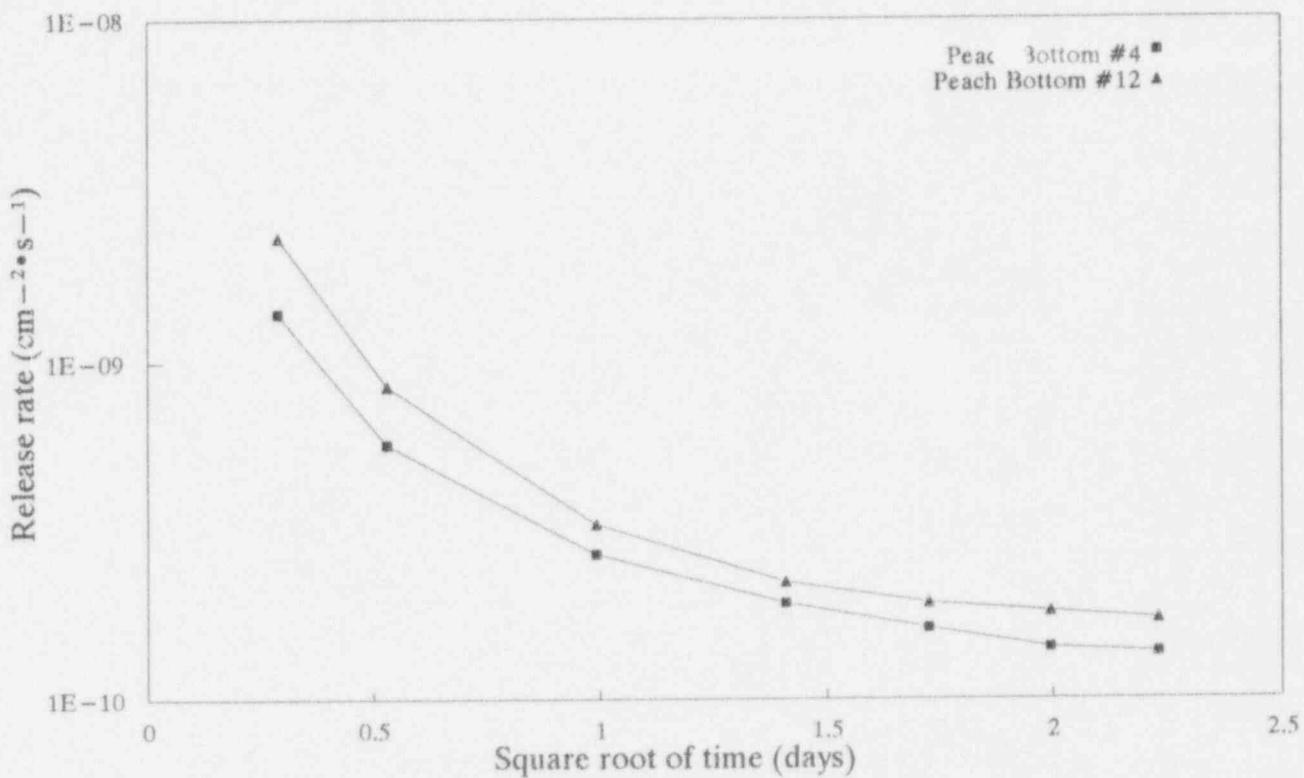


Figure 25. Fractional release rates of chromium.

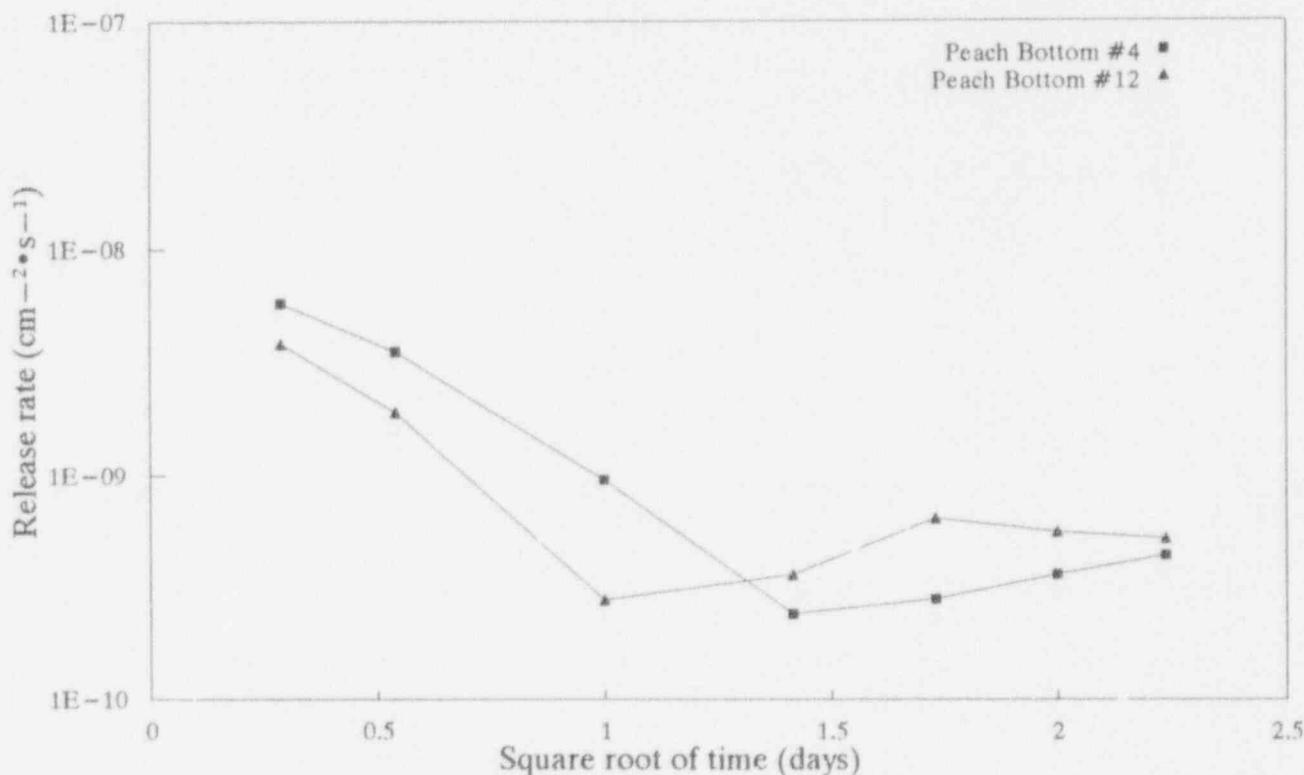


Figure 26. Fractional release rates of cobalt.

No reason for this difference in the behavior of ^{55}Fe and iron is apparent because kinetics suggest that ^{55}Fe should be in equilibrium with whatever chemical form the elemental iron is in.

The average absolute release rates for iron, nickel, and chromium, as shown in Table 10, indicate that the rates are within a factor of four for all elements. These data, when compared with the variable fractional release rate data, suggest that the release of these elements from the waste form is not dependant on the inventory in the waste form.

The CFR curves for iron, nickel, chromium, and cobalt are shown in Figures 27 through 30. In the cases of nickel and cobalt, standard diffusional release is suggested, and in the case of iron, the diffusional release from the intact sample #8 from Peach Bottom-3 is faster than that for the degraded FitzPatrick specimen. CFRs for nickel (2.9×10^{-2}) and chromium (1.6×10^{-2}) are within a factor of two, whereas the CFR for iron is considerably less at 4.6×10^{-5} (samples #4 and

#12). If sample #8 is included, the average CFR for all samples is about 1.8×10^{-4} . Without sample #8, which appears to be anomalous, the CFR for iron is about 0.1% of the CFR for nickel. These data indicate that the iron is retained in the waste form to a much greater extent than the other metal, which may be due to the greater complexing capability of nickel. These data suggest that a complexing effect is present that enhances the release of nickel, cobalt, and chromium. The high iron results for sample #8 have not been explained.

In addition to the results for the stable metals, boron and zinc were measured in some leachates from samples #4 and #12. Boron was measured in only three or four samples as shown in Appendix D. As shown in Table 10, the weighted average fractional release rate is $1.6 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$, with an average CFR of 2.7×10^{-2} and leachability index of 8.4. These data suggest that boron is relatively mobile and is probably in the form of an ion, perhaps a borate.

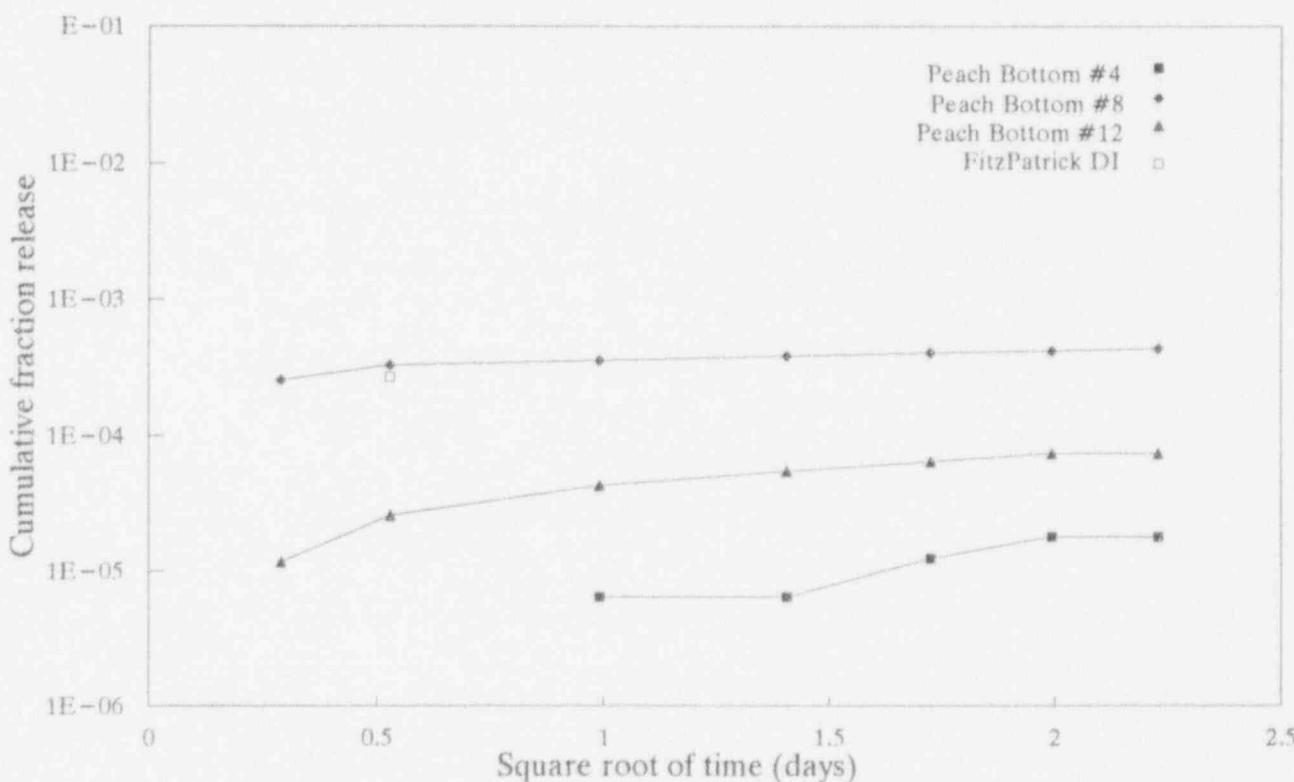


Figure 27. Cumulative fraction release of iron.

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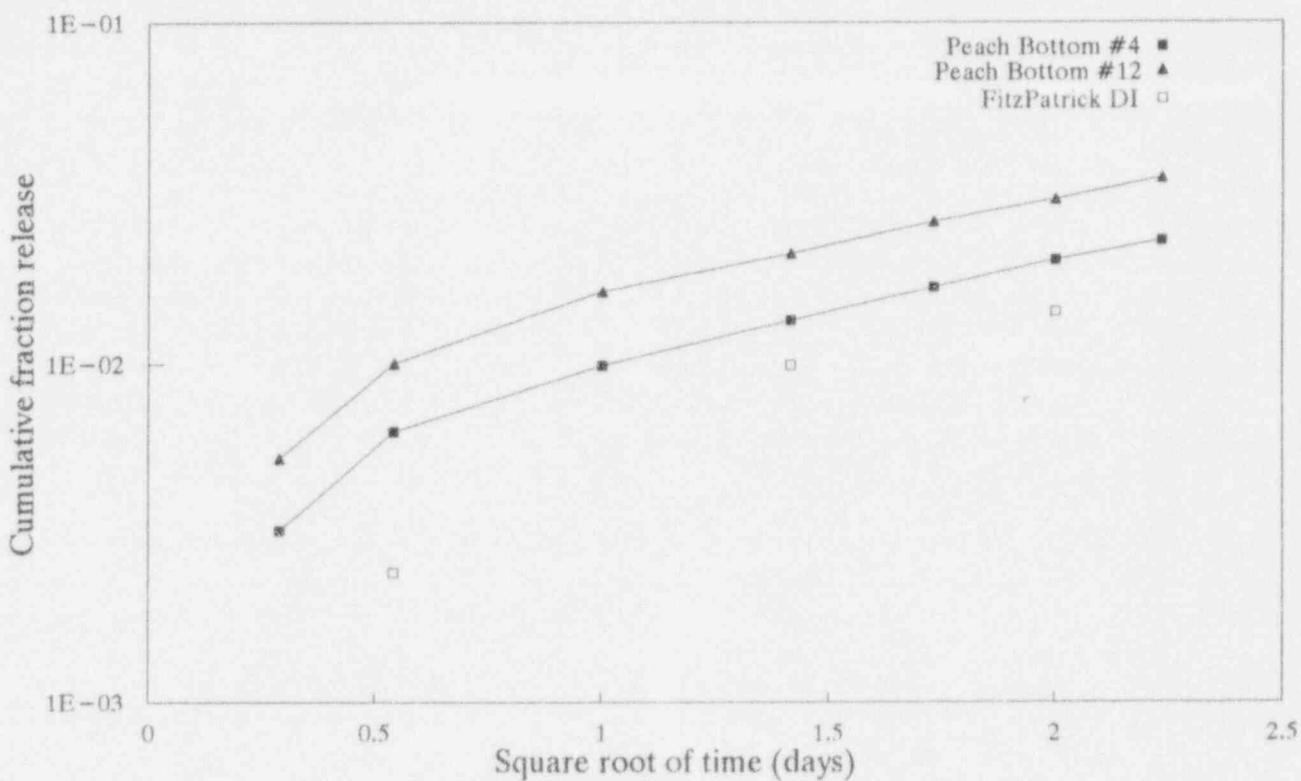


Figure 28. Cumulative fraction release of nickel.

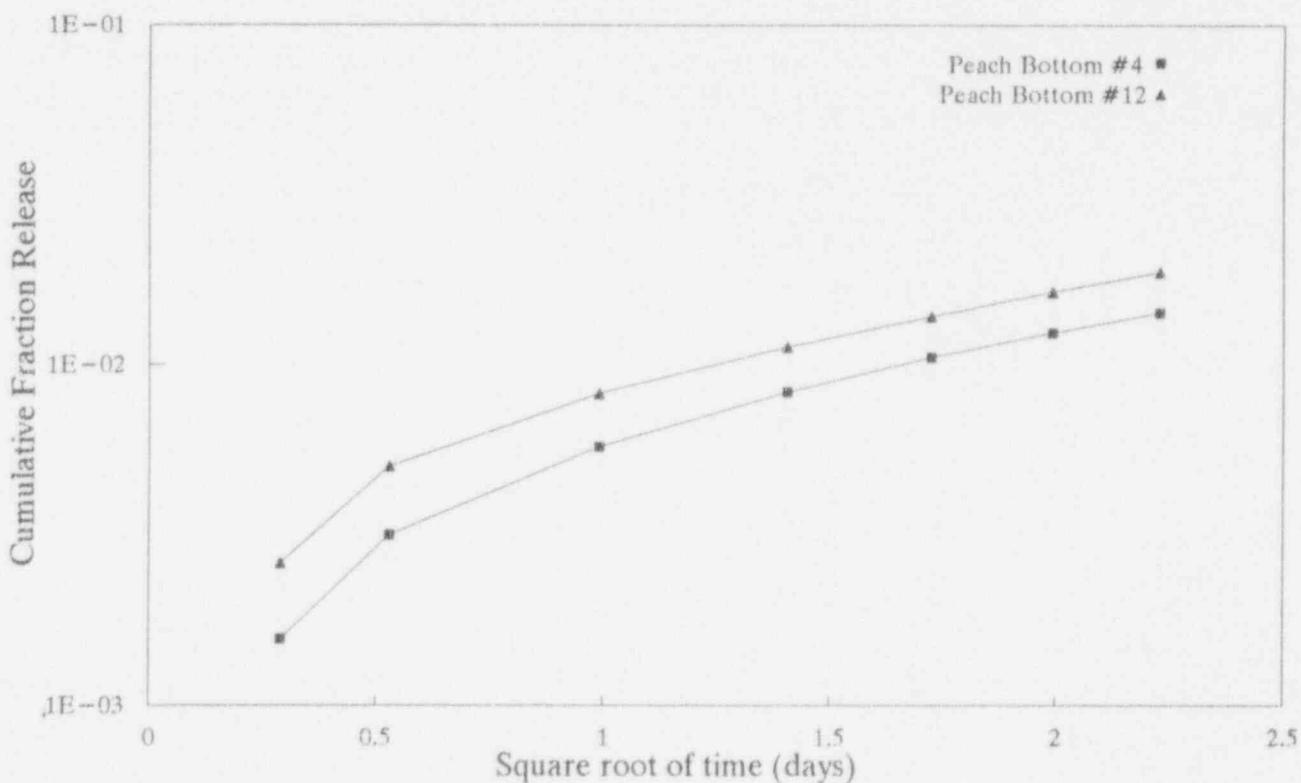


Figure 29. Cumulative fraction release of chromium.

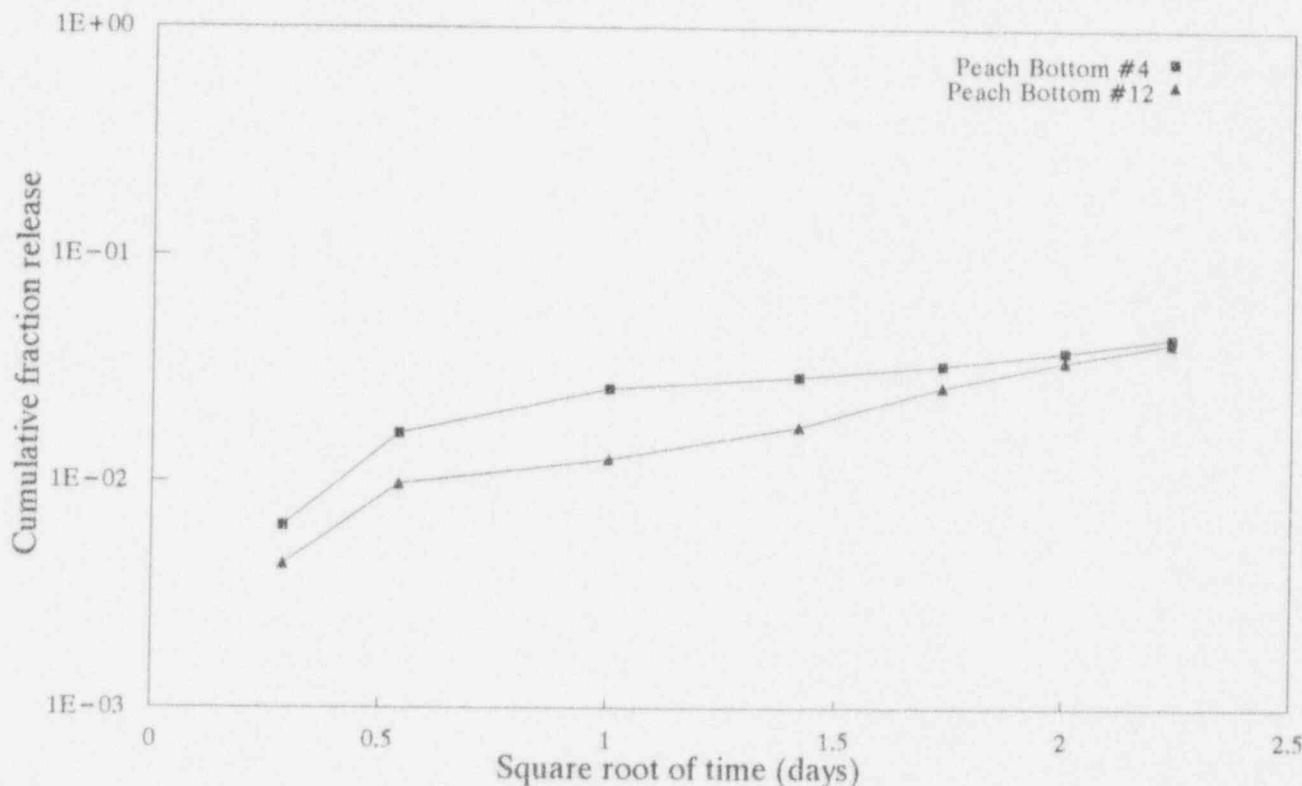


Figure 30. Cumulative fraction release of cobalt.

Zinc was retained to a greater extent in the waste form than boron and had fractional release rates similar to chromium and nickel, which suggests that this metal is diffusion-driven and is released from the waste form relatively quickly.

Average effective diffusivities and leachability indexes for the stable metals are shown in Tables 7 through 10. The leachability indexes are all well above the regulatory requirement of 6. The lowest is for nickel at 8.9, and the highest, as would be expected, is for iron at 14.4.

These results suggest that the average fractional releases of the stable metals may be different than the radionuclides. In addition, the nickel, chromium, and zinc are released at a faster rate than other elements and suggests that complexation of these elements, probably with picolinic acid, may result in enhanced releases of these elements.

The chelating agent, radionuclide, and stable metal data listed above define the release rate

behavior from the Peach Bottom-3 waste forms. The objective of these studies is to assess releases from waste sites or liners. Appendix E contains the inventory of radionuclides in the Peach Bottom-3 liner. The concentrations of radionuclides, chelating agents, and stable metals in the liner is presented in Table E-1 as Ci/liner for radionuclides or kg/liner of stable metals or chelates. The summed radionuclide content is 56 Ci/liner. The primary decontamination radionuclides present in the resins, based on their measured concentrations, are ^{65}Zn , ^{60}Co , ^{55}Fe , ^{63}Ni , and ^{14}C . The summed activity of these radionuclides is 55 Ci or about 98% of the total activity. Carbon-14 makes up about 58% of the total activity.

To assess possible radionuclide releases from the liner, the order of the leachability indexes are summarized below from lowest to highest: $^{129}\text{I} > ^{137}\text{Cs} > ^{90}\text{Sr} > ^{63}\text{Ni} > ^{99}\text{Tl} > ^{60}\text{Co} > ^{55}\text{Fe} > ^{14}\text{C}$. As expected, the cations and anions had the lowest leachability indexes (8–9), and ^{14}C had the highest (13.5).

COMPARISON OF PEACH BOTTOM-3 RESULTS WITH OTHER LOMI WASTE FORMS

This section compares the release rates of key constituents of the Peach Bottom-3 waste form with the releases from other LOMI waste forms including those that degraded during leaching. Inspection of Figure 31 indicates that the LOMI waste forms have similar fractional release rates for picolinic acid that average about $3 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$. This is interesting because the FitzPatrick waste form disintegrated shortly after being placed in the leach solution and yet the average fractional release rate of picolinic acid (3×10^{-9}) from the FitzPatrick specimen is similar to the Indian Point average fractional release rate and only a factor of five greater than the average fractional release rate of picolinic acid for Peach Bottom-3. These results are similar, although different LOMI formulas were used for all three waste forms. These data suggest that the structural stability of the waste form does not affect the release rate from the waste form. Also, the inventory of picolinic acid in the waste form does not

appear to affect the fractional release rate because the inventory of picolinic acid in the Indian Point sample is an order of magnitude less than the other waste forms and yet the fractional release rate is greater than those for FitzPatrick and Peach Bottom-3.

Figure 32 shows the CFR of picolinic acid for the LOMI waste forms and indicates that 50–60% of the picolinic acid was released from the waste forms except for Peach Bottom-3 (3.9%), and that about 80% of the formic acid was released from the Indian Point waste form. These data contrast with the laboratory studies performed by Piciulo⁴⁶ in which the CFR of picolinic acid for LOMI wastes was between 0.12 and 0.20. Further, the CFR for formic acid in Piciulo's study was ~0.3. These data indicate that the laboratory cement-solidified decontamination ion-exchange waste forms may not accurately reproduce the waste compositions found in actual commercial nuclear power plant

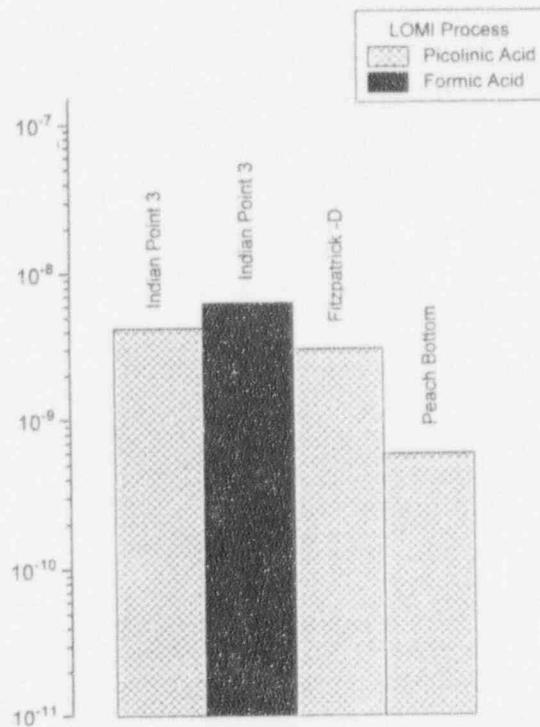


Figure 31. Comparison of average fractional release rates of picolinic acid from LOMI waste forms.

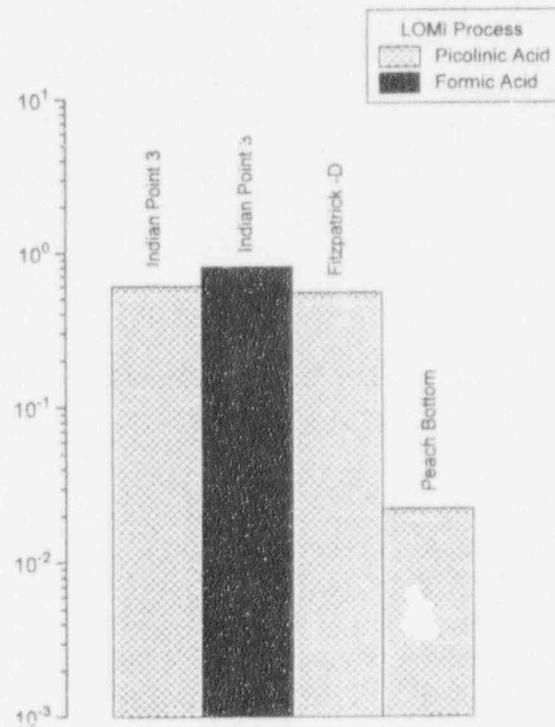


Figure 32. Comparison of cumulative fraction releases of picolinic acid from LOMI waste forms.

wastes and that, indeed, the commercial reactor wastes may contain constituents that enhance the release of picolinic acid from some waste forms (up to a factor of three). The fact that the CFR for Peach Bottom-3 is considerably better (i.e., lower) than that observed for other waste forms suggests that changes in the formulation of the waste form may have improved the leaching properties of the waste form.

Figures 33 and 34 show the average effective diffusivities and the leachability indexes for picolinic acid for the LOMI waste forms. With the exception of Peach Bottom-3, the effective diffusivities average 2.5×10^{-7} and suggest that the rates are similar for the intact and decomposed waste forms. The leachability indexes are similar and range from 6.9 to 8.8. In Piciulo's laboratory study,⁴⁶ his leachability indexes ranged from 8.6 to 9.1, which is reflective of the greater release rates from the actual waste form. In addition, his leachability indexes for formic acid ranged from 8.0 to 8.6, whereas the leachability index for the Indian Point waste form was 6.7.

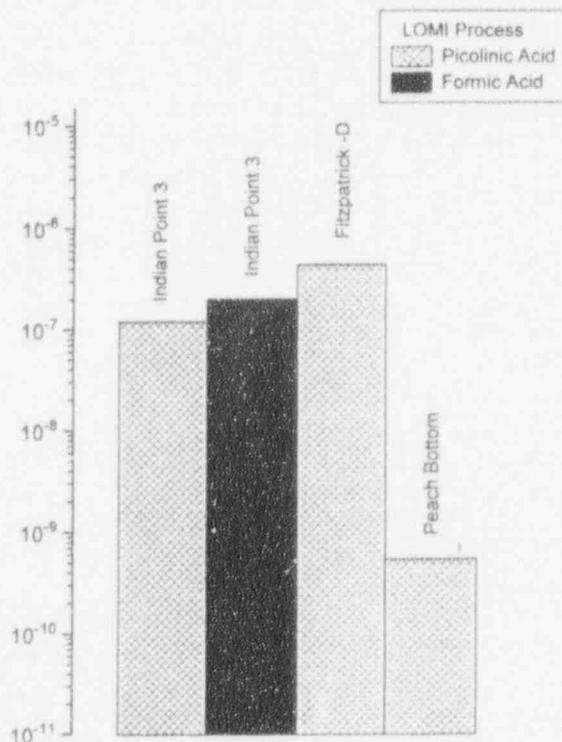


Figure 33. Comparison of average effective diffusivity of picolinic acid from LOMI waste forms.

Assessment of the average fractional release rates of the transition metals for the LOMI waste forms, as shown in Figures 35 through 37, indicates that the ^{60}Co , ^{55}Fe , and ^{63}Ni average fractional releases for Indian Point are similar at about $6 \times 10^{-12} \text{ cm}^2 \cdot \text{s}^{-1}$, whereas for FitzPatrick and Peach Bottom-3, ^{60}Co and ^{55}Fe have average fractional releases of about $6.5 \times 10^{-11} \text{ cm}^2 \cdot \text{s}^{-1}$. In contrast to these data, the ^{63}Ni average fractional release rates are approximately $4 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$ for both FitzPatrick and Peach Bottom-3. These data indicate a 10^3 range of average fractional release rates for the LOMI waste forms. In contrast, the average fractional release for picolinic acid for all three waste forms is approximately $3 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$. There is no evidence why the Indian Point waste form average fractional release rates are similar for all three radionuclides and yet there are differences for FitzPatrick and Peach Bottom-3.

The FitzPatrick and Peach Bottom-3 data suggest that ^{60}Co and ^{55}Fe have similar release rates and therefore may have similar diffusion

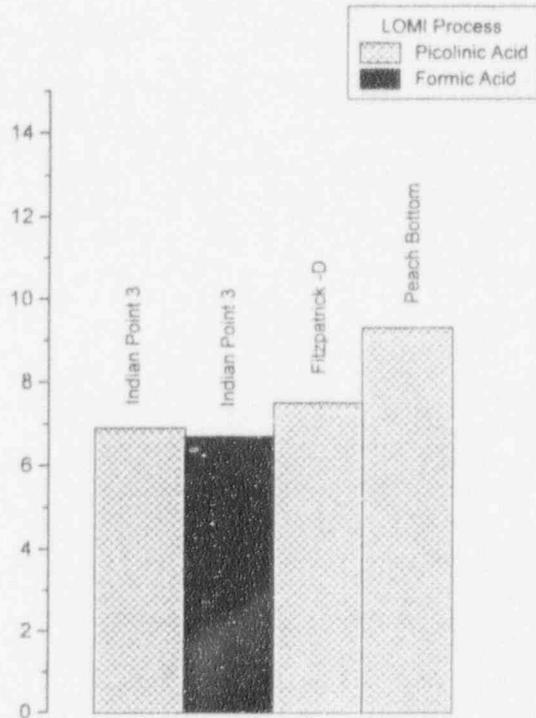


Figure 34. Comparison of leachability indexes of picolinic acid from LOMI waste forms.

Comparison Results

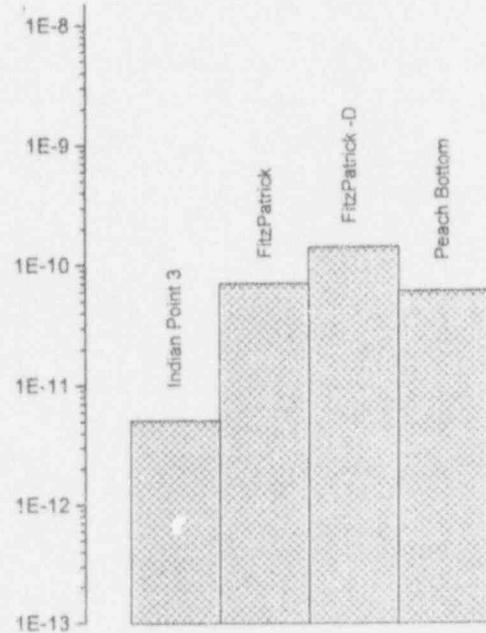


Figure 35. Comparison of average fractional releases of ^{60}Co from LOMI waste forms.

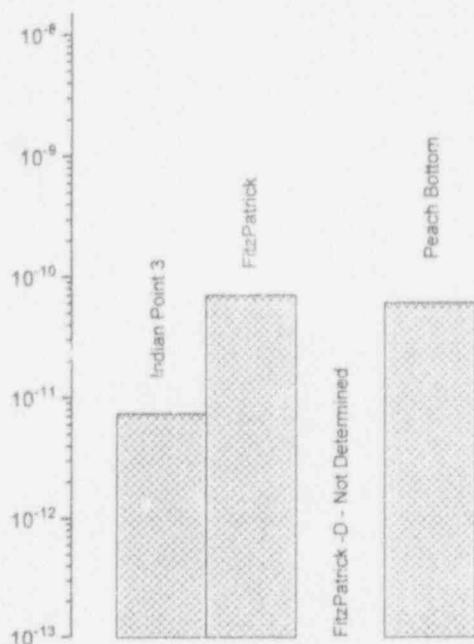


Figure 36. Comparison of average fractional releases of ^{55}Fe from LOMI waste forms.

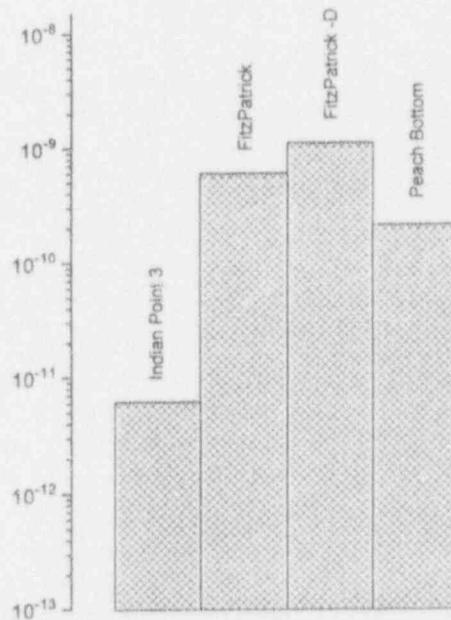


Figure 37. Comparison of average fractional releases of ^{63}Ni from LOMI waste forms.

characteristics from the waste form. Of primary importance is the fact that the average fractional release rates for these radionuclides were similar, although the FitzPatrick waste form decomposed and the Peach Bottom-3 waste form remained intact. The average fractional release rates of ^{63}Ni were also similar for both plants, although the average fractional release rates are approximately an order of magnitude greater than those of the other two transition metals. This difference in release rate may be due to the relative stability of complexes formed by the transition metals. The Irving-Williams correlation^{51,52} indicates that the stability of transition metal complexes fall in the order $\text{Ni}^{\text{II}} > \text{Co}^{\text{II}} > \text{Fe}^{\text{II}}$. These data suggest that the stability of the nickel complex with a chelating agent may result in the higher average fractional release rates of ^{63}Ni relative to the other transition metals. Also, the similarity of the picolinic acid average fractional release rates for all three waste forms suggests that other factors besides complexation with this chelating agent may be affecting releases from the waste form. In fact, it has been suggested that the pH and ion strength associated with the leachant, as affected by the

chemistry of the waste form, may cause the transition metal chelating agent complex to break down.³⁹

A comparison of the cumulative fractional releases for the transition metals is shown in Figures 38 through 40. An assessment of the cumulative fractional releases indicate that the greatest release (9.2%) is for ^{63}Ni from the FitzPatrick waste form. Other cumulative fractional releases range from 0.02 to 1%, with the release of ^{55}Fe and ^{63}Ni from the Indian Point specimen between 0.02 and 0.03%. In contrast, the CFRs for the chelating agents ranged from 50 to 60% with the exception of Peach Bottom-3 (3.9%). These data suggest that the chelating agent concentration is not directly correlatable with the release of radionuclides because the average fractional release rate for the transition metal radionuclides from the Indian Point waste form was much less than for the other plants, and yet the average fractional release rate and the CFR (0.6) of the chelating agent was the highest for this type of waste form.

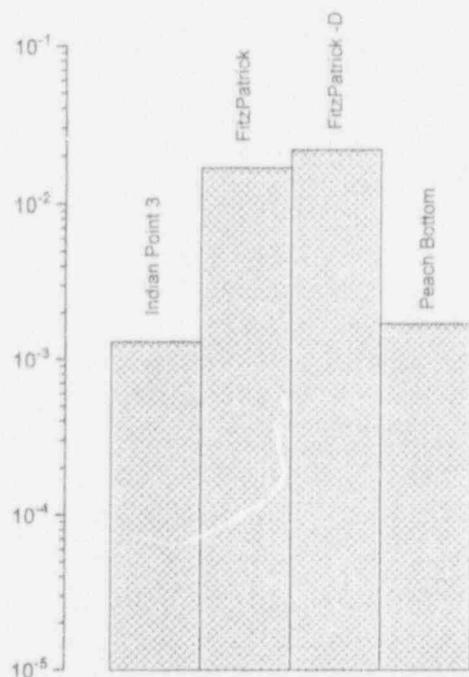


Figure 38. Cumulative fraction releases of ^{60}Co from LOMI waste forms.

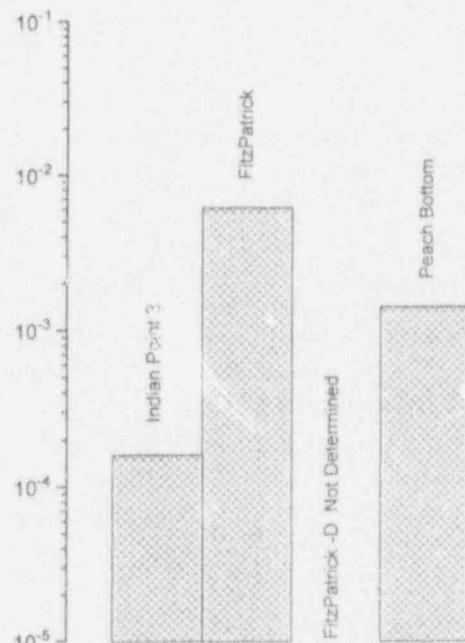


Figure 39. Cumulative fraction releases of ^{55}Fe from LOMI waste forms.

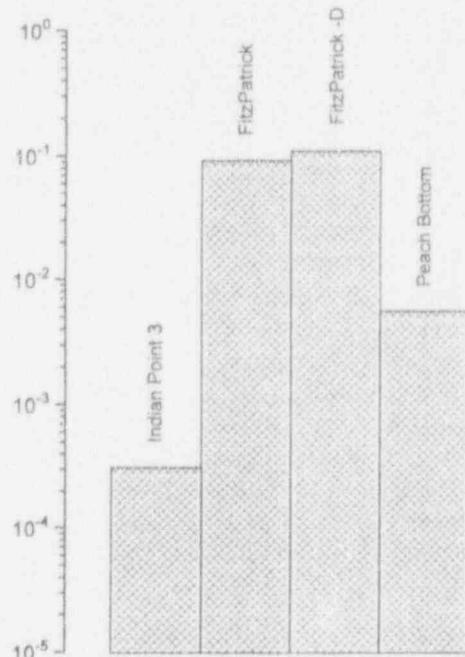


Figure 40. Cumulative fraction releases of ^{63}Ni from LOMI waste forms.

Comparison Results

Figures 41 through 43 show the average effective diffusivities for the transition metals. These results, as previously discussed, typically follow the results of the fractional release rates. For the LOMI waste forms, there is a broad range with FitzPatrick having the highest effective diffusivity. The average effective diffusivities for ^{55}Fe and ^{63}Ni are variable as discussed previously. Typical ranges are difficult to define due to the variations in each waste form.

The leachability indexes for the transition metals are shown in Figures 44 through 46 are more informative. For all transition metals, the leachability indexes are within the requirements of the "Technical Position on Waste Form," Revision 1. This lower leachability index may be due to the apparent mobility of ^{63}Ni , which appears to be generic for all waste forms.

Other radionuclides identified in this study are ^{241}Pu , ^{14}C , ^{99}Tc , and ^{129}I . In the leaching studies

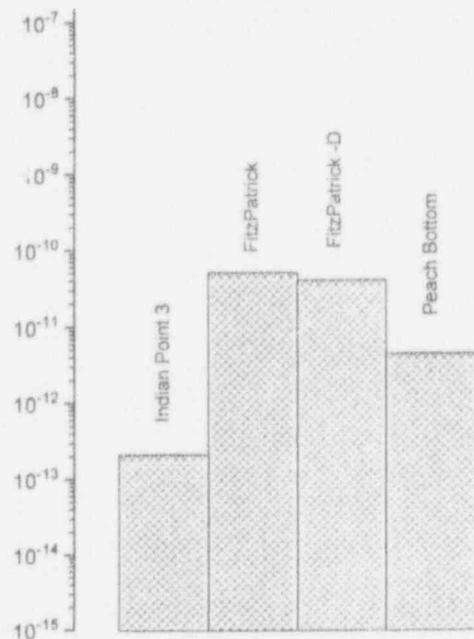


Figure 41. Average effective diffusivity of ^{60}Co from LOMI waste forms.

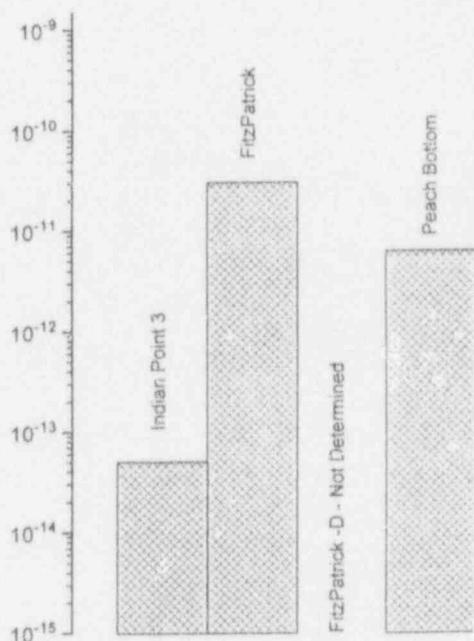


Figure 42. Average effective diffusivity of ^{55}Fe from LOMI waste forms.

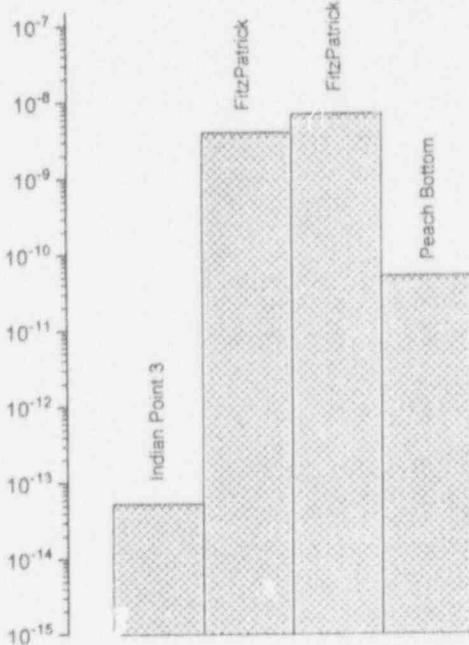


Figure 43. Average effective diffusivity of ^{63}Ni from LOMI waste forms.

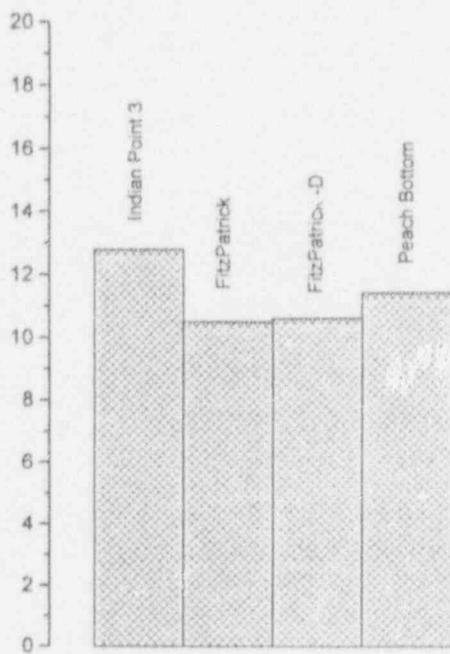


Figure 44. Leachability indexes for ^{60}Co from LOMI waste forms.

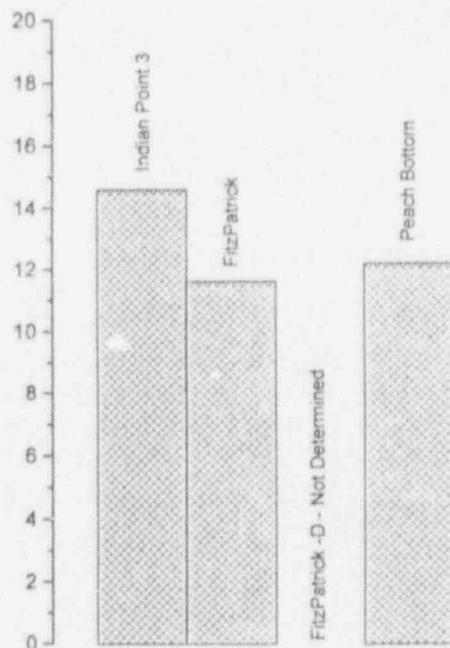


Figure 45. Leachability indexes for ^{55}Fe from LOMI waste forms.

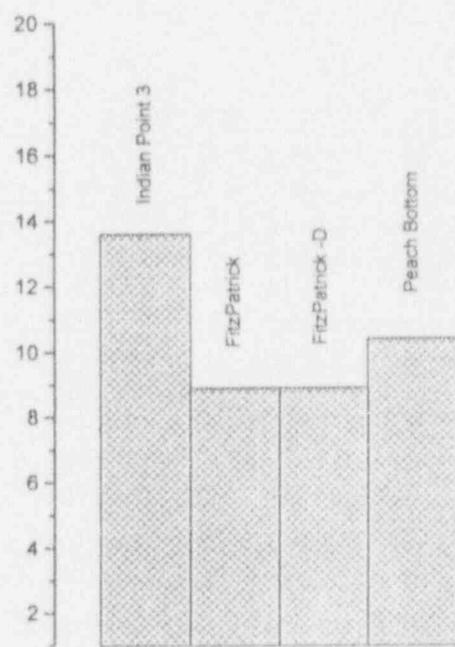


Figure 46. Leachability indexes for ^{63}Ni from LOMI waste forms.

performed for ^{241}Pu , only the inventory in the waste forms was measured. The inventory per gram of resin typically ranged from 10^{-6} to 10^{-4} $\mu\text{Ci}/\text{cm}^3$ of resin. However, no real values were measured. Results for ^{14}C , ^{99}Tc , and ^{129}I were not comparable with other LOMI waste forms because these radionuclides were not measured in previous studies. Consequently, the data developed in this report provide the primary baseline for the release of these radionuclides.

CONCLUSIONS

The primary conclusions of this study relate to the structural stability of the Peach Bottom-3 waste form and the releases of radionuclides, stable metals, and the chelating agent picolinic acid from the waste form. In this study, comparisons have been made with degraded waste forms such as the FitzPatrick waste form and with other LOMI waste forms.

The key conclusions of this study are that the Peach Bottom-3 waste form meets the structural stability requirements (compressive strength >500 psi) and leachability requirements (leachability index >6.0) identified in the NRC's "Technical Position on Waste Form," Revision 1.

To assess possible radionuclide releases from the liner, the order of the leachability indexes are summarized below from lowest to highest: $^{129}\text{I} > ^{137}\text{Cs} > ^{90}\text{Sr} > ^{63}\text{Ni} > ^{99}\text{Tc} > ^{60}\text{Co} > ^{55}\text{Fe} > ^{14}\text{C}$. As expected, the cations and anions had the lowest leachability indexes (8–9), and ^{14}C had the highest.

In the evaluation of the leach test results, primary conclusions were developed in the area of pH effects on leachability and the characteristics of radionuclide and stable metal releases from the waste form. The pH data indicate that the pH of the leachant is affected within a few hours and probably within a few minutes by the chemistry of the waste form.

In the characterization of the waste stream results, concentrations of radionuclides in the resins and concrete varied for several radionuclides. Results of these analyses suggest that there may be irregularities in the concentrations of radionuclides in the waste form. Among the stable metals whose concentrations were measured in the resin wastes, the iron concentration was highest at 3,000 $\mu\text{g}/\text{gram}$ resin. This was followed by nickel and chromium. In addition, analyses were performed for sulfate and phosphate. Neither ion was detected in the waste form or the resin samples.

The summed radionuclide content in the Peach Bottom-3 waste forms is 7.8 $\mu\text{Ci}/\text{g}$ of waste form. The primary decontamination radionuclides present in the resins based on their measured concentration are ^{54}Mn , ^{65}Zn , ^{60}Co , ^{55}Fe , ^{63}Ni , and ^{14}C . The summed activity of these radionuclides is 7.7 $\mu\text{Ci}/\text{g}$ waste form or about 98% of the total activity. Carbon-14 makes up about 58% of the total activity. The dominant decontamination radionuclides ^{60}Co and ^{55}Fe make up about 31% and 1.9% of the total activity in the resin waste, respectively. In contrast, the fission products ^{90}Sr , ^{99}Tc , ^{129}I , and ^{137}Cs collectively constitute about 0.3% of the total activity. The concentrations of the transuranic isotopes are also low and sum to a total of $1.6 \times 10^{-3} \mu\text{Ci}/\text{g}$ (0.02% of the total activity). Greater than 87% of the transuranic activity was ^{241}Pu .

A comparison of the leachability characteristics of picolinic acid from the Peach Bottom-3 and FitzPatrick samples indicates that the average absolute and average fractional release rates of picolinic acid for both waste forms are similar. These data indicate that the structural stability has a limited effect on the release rates of picolinic acid from the waste form and suggest that chemical mechanisms either in the waste form or in the resin itself control the release rate from the waste form.

A comparison of the absolute and fractional release rate data for the three decontamination radionuclides, ^{55}Fe , ^{60}Co , and ^{63}Ni , indicates that the lack of cement-solidified waste form structural integrity (does not hold together) does not appear to significantly affect releases from the solidified waste form. The average fractional release of ^{63}Ni is probably higher than that for the other decontamination radionuclides and indicates that this radionuclide is released at a higher rate than the others. The higher fractional release rates associated with ^{63}Ni may be due to the increased stability of nickel complexes, as the Irving-Williams correlation indicates that the stability of transition metal complexes fall in the order $\text{Ni}^{II} > \text{Co}^{II} > \text{Fe}^{II}$. These data suggest that the stability of the nickel complex with a chelating

agent may result in the higher average fractional release rates of ^{63}Ni relative to the other transition metals.

Other radionuclides for which analyses of the waste form and chelating agents were performed and measurable results were obtained were ^{14}C , ^{99}Tc , ^{129}I , ^{90}Sr , and ^{241}Pu . Carbon-14, ^{99}Tc , and ^{129}I were detectable in most samples, whereas ^{90}Sr was detectable in only a few of the leachate samples, and ^{241}Pu was not detectable in the leachates. The weighted average fractional release rates of ^{14}C are the lowest of any of the radionuclides measured. This is consistent with the results of Krishnamoorthy, who attributes the low release rate of ^{14}C measured in his study to the formation of insoluble hydrates and carbonates, which slow the release of this radionuclide. Further, he suggests that the fractional release rate of ^{60}Co should be slower than ^{14}C . This is inconsistent with our results in which ^{60}Co is released at a weighted average fractional release rate 20 times faster than ^{14}C .

This weighted average fractional release rate of ^{99}Tc is statistically the same as that for ^{55}Fe and ^{60}Co and suggests similar chemical and release rate behavior for ^{99}Tc . This might be expected because technetium is a metal and would be expected to form complexes with organic compounds such as chelating agents.

Fractional release rates for all radionuclides range from $3.5 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$ (^{129}I) to $3.4 \times 10^{-12} \text{ cm}^2 \cdot \text{s}^{-1}$ (^{14}C). The fractional release rates of iodine and cesium are the highest of all radionuclides present in the waste form and are statistically the same as that for picolinic acid ($5.8 \times 10^{-10} \text{ cm}^2 \cdot \text{s}^{-1}$), which suggests that the maximum diffusional fractional release rate from the waste form is between 5×10^{-10} and $1 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$.

Evaluation of the release rate data for the stable metals indicates that iron, cobalt, and chromium have similar release rate characteristics and that the release of these metals from the intact Peach Bottom-3 waste form is similar to that from the degraded FitzPatrick waste form. These data fur-

ther confirm the fact that the release of nickel is not dependant on waste-form structural stability. In addition, the average fractional release rate of nickel ($1 \times 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$) is about a factor of two greater than that for picolinic acid. These data suggest that the release of nickel and cobalt may not be controlled by the release of picolinic acid because they are released at a statistically faster rate than the picolinic acid.

In contrast to nickel, the data indicate that ^{55}Fe is released from the waste form faster than the elemental iron. This suggests that ^{55}Fe is in a different chemical form than the bulk of the elemental iron; however, kinetics suggest that ^{55}Fe should be in equilibrium with whatever chemical form the elemental iron is in. No explanation for this behavior is apparent.

Absolute release rates for iron, nickel, and chromium indicate that the release rates are within a factor of two for all elements. These data, when compared with the fractional release rate data, suggest that the release of these elements from the waste form is not dependant on the inventory in the waste form.

The concentrations of radionuclides, chelating agents, and stable metals in the liner were calculated as Ci/liner for radionuclides or kg/liner of stable metals or chelates. The summed radionuclide content is 56 Ci/liner. The primary decontamination radionuclides present in the liner, based on their measured concentration, are ^{54}Mn , ^{65}Zn , ^{60}Co , ^{55}Fe , ^{63}Ni , and ^{14}C . The summed activity of these radionuclides is 55 Ci or about 98% of the total activity.

Comparisons of the releases from LOMI waste forms that have been leached as part of this study and as part of previous studies indicate that the inventory of picolinic acid in the waste form does not appear to affect the fractional release rates because the inventory of picolinic acid in the Indian Point sample is an order of magnitude less than the other waste forms and yet the fractional release rate is greater than those for FitzPatrick and Peach Bottom-3. The fact that the CFR for Peach Bottom-3 is considerably better (lower) than that observed for other waste forms suggests

Conclusions

that changes in the formulation of the waste form may have improved the leaching properties of the waste form.

In summary, key conclusions from the Peach Bottom-3 study are that the compression test results and leachability indexes meet the requirements of the NRC's "Technical Position on Waste Forms," Revision 1, and that release rates of radionuclides, stable metals, and chelating agents do not appear, in general, to be affected by the structural stability of the waste form. These data suggest that waste form compression testing is of limited value in assessing actual waste-form stability. Other key points are that the apparent higher release rate for ^{63}Ni may be due to greater stability of complexes formed by this radionuclide and further suggests that there are chelant

affects on the transition metals. These effects are less apparent for ^{55}Fe and ^{60}Co , which form complexes that are less stable than those formed by ^{63}Ni . Also, the release rate behavior of ^{99}Tc is similar to that of ^{55}Fe and ^{60}Co and suggests similar chemistry for this radionuclide.

Another principal observation is that ^{14}C , which had the highest inventory (58%) of any radionuclide in the waste form, had the lowest leachability index, and indicates that this radionuclide is strongly retained in the cement matrix, probably as an insoluble hydrate or carbonate. In contrast, ^{129}I , a mobile anion, is released at rates similar to those for ^{137}Cs and has the lowest leachability index (8.4) or the highest release rate of all radionuclides measured.

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Appendix A

LOMI-NP-LOMI Decontamination Process

Appendix A

LOMI-NP-LOMI Decontamination Process

In most instances, chemical decontamination process formulas have been developed in a competitive environment; therefore, their exact chemical compositions are considered proprietary. The following description of the decontamination process used at the Peach Bottom Atomic Power Station Unit 3 from which solidified decontamination resin waste forms were collected was obtained from the literature and does not include information of a proprietary nature.

There are two general methods that have been developed to perform full system decontaminations of LWRs: concentrated chemical processes and dilute chemical processes. The concentrated processes use reagent concentrations of between 5 and 25 wt%, and the dilute processes that are currently employed use reagent concentrations of 1.0 wt% or less. Based on these concentrations, the quantity of reagents required for a concentrated process decontamination range from about 48 to 248 kg of reagent/m³ of primary system volume to be contaminated. A similar estimate for a dilute process decontamination is less than 10 kg/m³. During recent years, the dilute chemical processes have become the most widely used.

The decontamination processes used at the LWRs from which samples were obtained were dilute chemical, multi-step processes. They employed between 0.1 and 0.6 wt% reagent and were multi-step in the respect that they required an initial oxide removal step, a chrome removal step, and a final oxide-removal step. The processes were performed at low temperature [333 to 368 K (60 to 95°C)] and atmospheric pressure. The processes, in general, use a combination of organic acids and chelating agents to dissolve the oxide film from surfaces and suspend the resulting organo-metal complexes in solution. Corrosion inhibitors are sometimes added to reduce the attack of organic acids on carbon steel piping. Some oxide films have significant concentrations of chromium that adversely affect the dissolution of the oxide layer. Oxidizing treat-

ments are used on such films to condition them by rendering the chromium soluble through oxidation from +3 to +6 valence state.

The reagents are slurried or dissolved in concentrated form and injected directly into the primary system water and circulated for 1 to 3 days. During the process, the decontamination solution is passed through cation exchange resins to remove the corrosion and activation products and regenerate the reagents. Following completion of decontamination, mixed-bed resins are normally used to remove the residual metallic ions and the decontamination reagents.

The LOMI process^{1,2,3} was developed by the Central Electricity Generating Board in Britain and is now being marketed in the United States by Vectra Technologies (formerly Pacific Nuclear Services) and Westinghouse. The basic feature of the process is the use of V⁺² (as vanadous formate) to reduce the Fe⁺³ in oxide films to Fe⁺². The process involves electron stripping rather than attack of the film by acid. It can be a multi-step process that normally employs a chromium removal step following the initial LOMI process. Picolinic acid (as sodium picolinate) is used as a complexing agent and is injected into the coolant, and the coolant is circulated while being maintained at a temperature of between 353 and 368 K (80 and 95°C) under atmospheric pressure. Vanadous formate [V(HCOO)₂] is then injected to reduce Fe⁺³ to Fe⁺². The reaction destabilizes the oxide film releasing the metal ions to the solution. The excess picolinate in the reagent complexes with the metal ions and keeps them from redepositing on the system surfaces. The LOMI reagents and dissolved radionuclides and metal ions are removed from the system through treatment with ion-exchange resin columns. Both strong-acid cation and weak-base anion resin columns are usually employed to process the spent LOMI reagent solution.

The initial LOMI step is followed by the injection of nitric acid potassium permanganate (NP)

Appendix A

at a pH of 2.5 or alkaline potassium permanganate (AP), which is currently used at a pH of 10–11.5. These solutions render the chromium soluble, leaving behind an iron-rich deposit that can be dissolved using the LOMI reagents. The NP (or AP) reagents are removed on ion-exchange resin columns. Residual MnO₂ that is formed during the oxidation process is dissolved with oxalic acid, which is added directly to the permanganate solution and removed on a mixed-bed resin column. Following the oxalic acid rinse, the initial LOMI step can be repeated using a more dilute concentration of LOMI reagents. This decontamination process is commonly referred to as the LOMI-NP-LOMI (or LOMI-AP-LOMI) process. In either case, the spent reagents and corrosion

products removed during decontamination are processed through ion-exchange resins, and these resins constitute the decontamination waste product.

The LOMI-NP-LOMI process as described in References 4, 5, and 6 was used for the decontamination at Peach Bottom-3.^a The LOMI-NP-LOMI decontamination took 4–5 days, and a total of 39 Ci was removed.⁵ Figure A-1 from Reference 5 shows the release of radioactive material for each step.

a. Personal communication, John Remark, February 17, 1994.

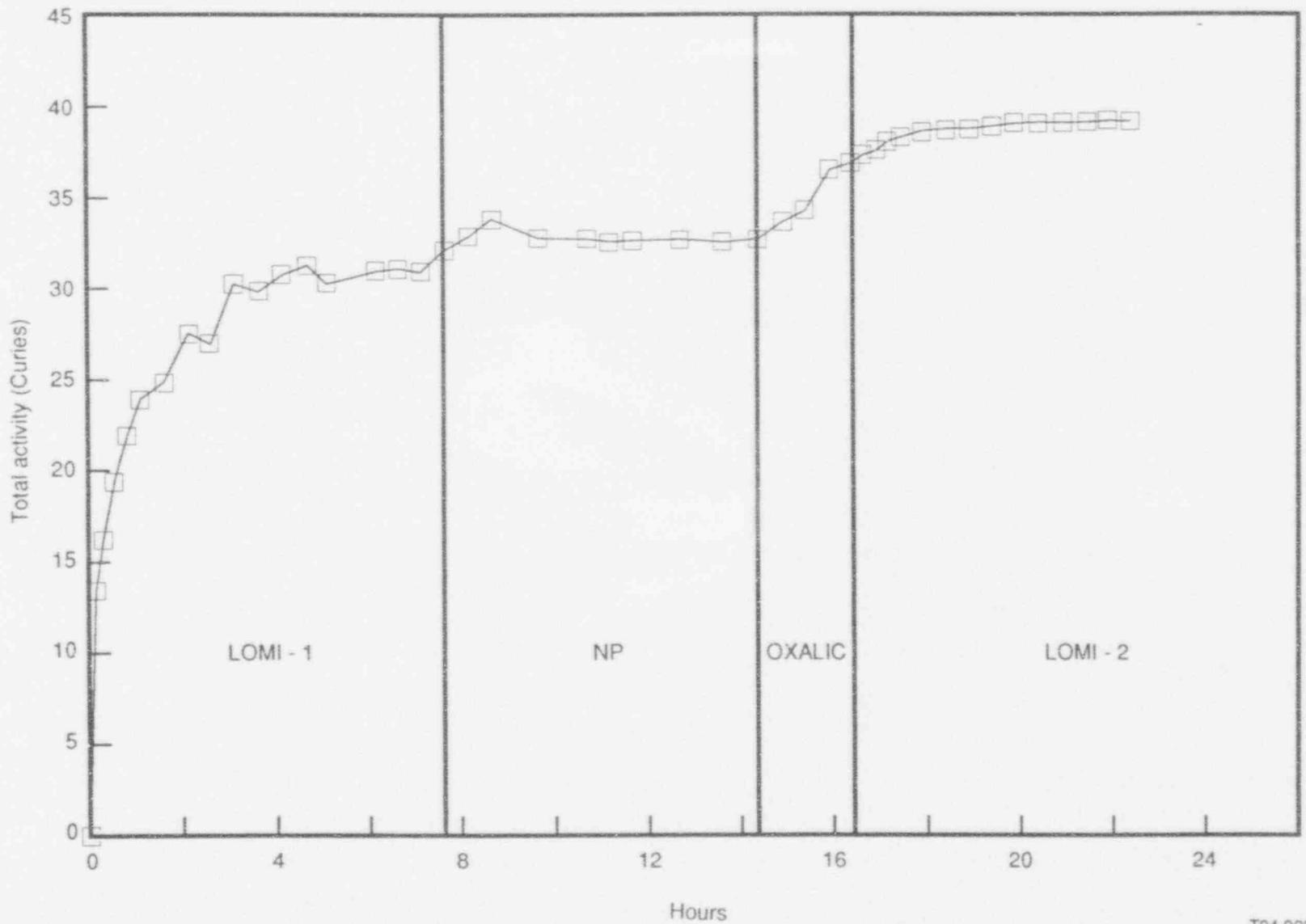


Figure A-1. Radionuclide release from Peach Bottom-3 LOMI-NP-LOMI decontamination process.

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Appendix B

**Summary of Solidification Performed at the
Peach Bottom Atomic Power Station Unit 3**

Appendix B

Summary of Solidification Performed at the Peach Bottom Atomic Power Station Unit 3

On October 20, 1989, Chem Nuclear Systems Inc. (CNSI) personnel prepared two process control program (PCP) specimens using 100-mL samples of wet ion-exchange resin that had been removed from liner 446828-15 after it had been mixed for about 30 minutes. The PCP specimens were prepared in 250-mL plastic disposable beakers that were equipped with air-tight lids. After the resin beads settled in the PCP containers, a thin layer of free-standing water, not more than 1/8-in. thick, covered the beads. The first step in preparing the PCP samples involved raising the pH of the resin. This initial pH adjustment was accomplished using a proprietary pH-adjusting agent. The agent was added to each PCP sample in 2-g increments, and after each addition, the sample was stirred for 3 minutes, and the pH of the resin was measured. When the pH-adjusting agent was stirred into the resin samples, the free-standing water in each sample turned a burnt-orange color. The initial pH of both of the PCP resin samples was about 4, and following the addition of 6 g of the pH-adjusting agent to each PCP specimen, the pH of both samples was 10.5. The pH of the two resin samples was monitored for about 20 minutes following the last addition of the pH-adjusting agent and, during this time, the pH of both samples remained unchanged. In all, 38-1/2 g of flyash was then slowly added to each PCP sample, and each sample was then stirred for several minutes. Finally, two different quantities of Portland Type I-P cement were then stirred into the two PCP samples. The quantities of Portland Type I-P cement that were added to the two samples, 48 g and 71 g, spanned the range of concentrations that had been certified by CNSI. In each case, the cement was added in 20-g increments every 10 minutes. When the final amounts of cement were stirred into the samples, they were sealed and placed in an oven that was maintained at 145°F, where they were baked overnight.

The two PCP specimens were removed from the oven after they had been baked between 18 and 24 hours and were then examined. The sample that contained 71 g of Portland cement was hard; however, the sample containing 48 g of Portland cement was somewhat soft. One could push the tip of a pencil into the surface of the latter PCP sample. Because the PCP specimen that contained the smaller quantity of Portland Type I-P cement had not set up as hard as the other PCP sample, five additional PCP specimens were prepared on October 23.

The procedure used on October 23 was the same as that previously used, except that the quantity of pH-adjusting agent added to each sample was increased to 8 g. Two of these PCP samples were prepared using the same quantities of Portland cement that had been used on October 20; the remaining three PCP samples were prepared using 50, 55, and 60 g of Portland cement. CNSI personnel stated that the quantity of pH-adjusting agent added to the PCP specimens was increased to help drive off the ammonia. They stated that the odor of ammonia was present when the PCP samples were prepared on October 20 and that the smell was particularly strong in the sample that contained the smaller quantity of Portland cement. The odor of ammonia was also quite strong during the preparation of the new PCP samples on October 23. When the final quantities of Portland cement were stirred into the five PCP samples, they were sealed, placed in an oven that was maintained at 145°F, and baked overnight to simulate the hydration exotherm.

The following morning, the PCP specimens were removed from the oven and examined. All five of the samples were hard and did not contain any free-standing water. Based on the quality of these PCP samples, the decision was made to proceed with the solidification of the three liners

Appendix B

using the same formula used to prepare the PCP sample containing the largest quantity of cement. EG&G Idaho personnel requested that liner 446828-15 be solidified first. Since it contained the highest concentrations of picolinic acid and activation metals, for our leach testing purposes, it was the best candidate of the three liners for sampling.

The solidification of the ion-exchange resins in liner 446828-15 commenced on October 24, 1989, at 10:46 a.m. when the pH-adjusting agent began to be added to the liner. A total of 550 lb of TSP was added to the liner over an 8-minute period. The pH-adjusting agent reduced the viscosity of the resin as was evidenced by the drop in the hydraulic pressure needed to rotate the mixing blades at 60 rpm. (The hydraulic pressure dropped from an initial value of about 2,000 psig to about 1,400 psig after the pH-adjusting agent was added.) Following the addition of the pH-adjusting agent, a total of 2,730 lb of flyash was then added to the liner from 11:10 a.m. to 12:07 p.m. When this addition was completed, the material in the liner was stirred for about 20 minutes without any new material being added to the liner. At 12:27 p.m., the addition of Portland Type I cement began. A total of 4,982 lb of Portland Type I cement was added to the liner between 12:27 p.m. and 1:52 p.m. After the last of the Portland cement had been added to the liner, the contents of the liner were mixed for a few minutes before the mixing motor was shut down. The quantities of resin and binder materials in the liner when the solidification operation was completed are summarized in Table 3.

After the fill-head was removed from the top of the liner, the resin/cement mixture inside the liner was sampled. This sampling tool, which was simply a plastic tube equipped with a plunger, was inserted into the resin/cement mixture five or six times to a depth of about 3 ft below the top surface of the mixture. Following each insertion, the material inside the tube was transferred to a plastic-lined bucket. About 2 gal of resin/cement mixture was collected. The resin/cement mixture was quite fluid, which made it possible to pour the material into the molds that were used to prepare

waste-form specimens. Between 2:35 and 3:05 p.m. on October 24, 19 waste-form samples were prepared, which were 2 in. in diameter and 4 in. long. Individual samples had contact exposure rates of about 100 mR/hour. The sample molds were sealed immediately after they were filled, and at 6:30 p.m. on October 24, they were all placed in an oven that was maintained at 145°F.

Three of the waste-form specimens were removed from the oven the following morning and were examined. All of the samples that were removed had some free-standing water on their top surfaces. They felt firm but were certainly not yet hard. These samples were then returned to the oven and all 19 specimens were baked at 145°F for a total time of about 72 hours. When these samples were examined by Peach Bottom-3 personnel during the week of October 30, they were hard, and no free-standing water was visible on their surfaces.

The waste resins in the remaining two liners, which were designated liners 446692-1 and 446828-10, were solidified after October 24 using the same solidification formula that had been used to solidify the ion-exchange resin in liner 446828-15. During the week of October 30, the resin/cement mixtures in all three liners were visually examined, and the hardness of the resin/cement monoliths were tested with a broom handle. The surfaces of the monoliths in liners 446692-1 and 446828-10 were impenetrable, but to everyone's surprise, the broom handle penetrated the material in liner 446828-15. A thin crust had formed over the top surface of the resin/cement mixture, but below the crust, the material had not yet solidified. CNSI personnel suggested that ammonia in the resin/cement mixture was retarding the setting of the cement. They recommended that the liner be vented to help remove the ammonia. The Philadelphia Electric Company followed the recommendation and began venting the liner the week of October 30. The liner was not examined again until December 21, 1989. On that date, the upper surface of the monolith was again probed with a broom handle, and this time, the broom handle did not penetrate the surface. During the week of January 15, 1990, the wall of liner 446828-15 was drilled at three dif-

ferent heights: about 4 in. from the top, near the center, and about 2 in. from the bottom. At the lower two locations, a soft, slightly damp material adhered to the grooves in the drill bit when the bit was withdrawn from the liner. Personnel from the Philadelphia Electric Company and the NRC who were present when the liner was drilled interpreted the presence of the material on the drill bit as meaning that the resin/cement mixture had not yet completely solidified.

On February 6, 1990, the condition of the material in liner 446828-15 was reexamined. The CNSI solidification supervisor who had supervised the solidification of all three liners during October 1989 personally conducted the examination. At a location on the liner wall opposite the location that had been previously drilled, 1.5-in.-diameter plugs were removed from the liner wall at three different heights. A hole saw was

employed to remove the plugs so that the surface of the resin/cement monolith would not be damaged during the process of removing the plugs. At all three locations where the plugs were removed, the surface of the monolith was found to be smooth and hard. The hardness of the surface of the monolith was tested by pounding the point of a chisel against the surface. The surface was found to be impenetrable. Following this examination, the CNSI solidification supervisor stated that he felt the liner had hardened sufficiently to allow it to be shipped to a disposal site. He also suggested that the results of the examination that had been performed during the week of January 15 might have been misinterpreted. His point was that solidified ion-exchange resin waste forms are not high-density concrete, and therefore, one would expect to be able to penetrate such waste forms using a high-speed drill.

Appendix C

Detailed Procedures for ^{14}C , ^{99}Tc , and ^{129}I Analysis

Appendix C

Detailed Procedures for ^{14}C , ^{99}Tc , and ^{129}I Analysis

Samples of resin wastes and leachates are typically analyzed for radionuclides specified in 10 CFR 61 (i.e., ^{14}C , ^{60}Co , ^{63}Ni , ^{90}Sr , ^{99}Tc , ^{129}I , ^{137}Cs , ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm) and other radionuclides using standard environmental analysis procedures. Radioanalytical procedures performed for the analysis of most of these radionuclides are specified in References 1 and 2; however, the procedures for ^{14}C , ^{99}Tc , and ^{129}I are not included as only the Peach Bottom-3 samples have been analyzed for these radionuclides.

Attachments 1 through 3 contain summaries of the analytical procedures used by B&W Nuclear Environmental Services to perform the analyses for ^{14}C , ^{99}Tc , and ^{129}I . All procedural steps are included with the exception of laboratory-specific information, calculational methods, and reporting requirements. Further information may be obtained from B&W Nuclear Environmental Services at the Lynchburg Technology Center in Lynchburg, Virginia.

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Attachment 1

B&W Nuclear Environmental Services, Inc.

LYNCHBURG TECHNOLOGY CENTER

TECHNICAL PROCEDURE

NUMBER TP-690 REV. NO. 0

TITLE TECHNETIUM-99 ANALYSIS

Appendix C

1.0 APPLICABILITY

This procedure applies to all Nuclear Environmental Laboratories personnel who are involved with radioactive waste sample analysis.

2.0 SCOPE

This procedure provides the steps necessary to identify and quantify the Technetium-99 in radioactive waste sent to burial sites for disposal. Based on the requirements set forth in the United States Code of Federal Regulations, Title 10 Part 61 (10 CFR 61), "Licensing Requirements for Land Disposal of Low Level Waste," radwaste samples must be analyzed for radionuclide concentration.

3.0 PROCEDURE SUMMARY

These samples shall be processed under chain-of-custody requirements. Interlaboratory transfers of samples shall be accompanied by Internal Chain-of-Custody (ICOC) forms.

Sample aliquots are obtained from the gamma scan fraction which was prepared in the Gamma Scan Preparation Section of TP-398. The Tc-99 aliquot undergoes an oxidation-reduction process and tributyl phosphate extraction. Care must be taken to prevent volatilization of Tc-99.

All experimental information shall be recorded on Benchsheet 690-1.

4.0 GLASSWARE

Before beginning the separation procedure it is strongly recommended that you gather together and label all of the glassware necessary for the separation of Tc. (All glassware used in this separation procedure shall have been cleaned according to TP-655.) It is also recommended that colored tape be used to color coordinate the spiked and unspiked samples of the same NEL number in order to reduce the possibility of cross-contamination. ALL LABELS MUST CONTAIN THE COMPLETE LABORATORY WORKING SAMPLE ID NUMBER. THIS NUMBER WILL CONSIST OF THE NEL # AND THE APPROPRIATE TAG NUMBERS (OR LETTERS OR SOME COMBINATION OF THE TWO) USED TO IDENTIFY THE ISOTOPE AND SEPARATION FRACTION.

The glassware listed below is the glassware that is expected to be used for each sample in a batch. Multiply the amount of glassware by the number of samples (including replicate, QC, and reagent blank) which will be analyzed. This listing does not include miscellaneous glassware used for reagents or chemicals.

- 4.1 Two beakers and watch glasses of appropriate size (Note: ≥ 100 mL beakers recommended for soil samples due to the vigorous reaction that occurs) if the hot plate digestion procedure is to be used (Step 9.5).
- 4.2 Two quartz crucibles if the furnace method of digestion is used (Step 9.5).
- 4.3 Two funnels for sample filtration (Step 9.5.3).
- 4.4 Two beakers of appropriate size for the sample filtrate (Step 9.5.3).
- 4.5 Two 125 mL separatory funnels (larger funnels may be necessary for larger sample sizes) (Step 9.5.3).

- 4.6 Two 15 mL polypropylene centrifuge tubes with caps (or equivalent) (Step 9.11).
- 4.7 Two 4 mL class A pipets (or an appropriate Eppendorf or class A pipet of suitable volume capacity) (Step 9.13).
- 4.8 Two glass liquid scintillation vials (Step 9.13).
- 4.9 Two glass liquid scintillation vials for efficiency standard and background (Step 9.15).

5.0 REAGENTS

- 5.1 **6.0 N Sulfuric Acid (H_2SO_4)** — Carefully add 167 mL of concentrated H_2SO_4 to a one liter volumetric flask containing 500 mL of reagent grade water and dilute to one liter with reagent grade water when the solution has cooled. Store in a plastic reagent bottle.
- 5.2 **6.0 N H_2SO_4** — 2 % HF (Sulfuric acid and Hydrofluoric acid) — Add 167 mL H_2SO_4 and 20 mL concentrated HF to 500 mL of reagent grade water and dilute one liter with reagent grade water. Cool. Store in a plastic bottle.
- 5.3 **4.0 N Hydrochloric Acid (HCl)** — Add 333 mL of concentrated HCl to 500 mL of reagent grade water and dilute to one liter with reagent grade water when cool. Store in a plastic bottle.
- 5.4 **9 N (H_2SO_4)** — Carefully add 250 mL of concentrated H_2SO_4 to a one liter volumetric flask containing 500 mL of reagent grade water and dilute to one liter with reagent grade water.
- 5.5 **Tributyl Phosphate (TBP) (equilibrated)** — Place 100.0 mL of TBP into a 250 mL separatory funnel and add 100.0 mL of 9 N H_2SO_4 , shake for 5 minutes, allow phases to separate, save TBP (the top layer) in a dark bottle, neutralize and discard the 9N H_2SO_4 .
- 5.6 **Te-99 Tracer** — NIST traceable or calibrated against a NIST traceable standard.
- 5.7 **Cobalt Carrier (5.0 mg Co/mL)** — Dissolve 20 g $CoCl_3 \cdot 6H_2O$ in 500 mL of 0.1 N HCl and dilute to one liter with 0.1 N HCl.
- 5.8 **Cesium Carrier (5.0 mg Cs/mL)** — Dissolve 6.3 g $CsCl_2$ in 500 mL of 0.1 N HCl and dilute to one liter with 0.1 N HCl.
- 5.9 **Manganese Carrier (5.0 mg Mn/mL)** — Dissolve 18.0 g $MnCl_2 \cdot 4H_2O$ in 500 mL of 0.1 N HCl and dilute to one liter with 0.1 N HCl.
- 5.10 **1% Potassium Permanganate (KMnO₄) (1 g/100 mL)** — Dissolve 5 g of KMNO₄ in 400 mL of reagent water and dilute to 500 mL with reagent grade water.
- 5.11 **0.1 N HCl** — Add 8.3 mL of concentrated HCl to 500 mL of reagent grade water and dilute to one liter with reagent grade water. Store in a plastic reagent bottle.

6.0 TECHNETIUM-99 SAMPLE PREPARATION AND ANALYSIS

Sample aliquots shall be removed from the gamma scan fraction (TP 398, Section 9) unless otherwise indicated by the Project Leader. Any deviations from this procedure shall be documented on Benchsheet 690-1.

Appendix C

Each sample in a batch is processed in duplicate as a spiked and as an unspiked sample. Each spiked sample contains a known amount of a NIST traceable Tc-99 standard solution of ~ 100000 dpm.

This separation should be performed in a fume hood as Technetium may be volatile.

- 6.1 Each batch of 10CFR61 samples processed must contain a QC sample, a reagent blank and a replicate of one sample for each batch of <= 10. If more than 10 but less than 20 samples are processed in a batch then a replicate of two samples must be processed.

- 6.2 **QC Sample** — A QC sample is a sample of a matrix similar to that of one of the samples being processed in the batch. Both the spiked and unspiked sample aliquots are spiked with a known amount of a NIST traceable Tc-99 radiotracer (~10,000–50,000 dpm). Whenever possible, use a QC stock sample. If a stock sample is not available, a QC sample may be prepared as follows:

- 6.2.1 Choose a matrix for the QC sample that will best represent a typical sample of the batch to be processed.

- 6.2.2 See Section 9.4 regarding recommended sample sizes.

- 6.2.3 Add a known amount of and NIST traceable Tc-99 standard to the QC sample matrix. Record the standard information, aliquot size, pipet ID and error on Benchsheet 690-1.

- 6.3 **Reagent Blank** — A reagent blank is comprised of an aliquot of reagent grade water. NO RADIOTRACER SPIKE IS ADDED TO THE UNSPIKED REAGENT BLANK!

- 6.3.1 Up to 100 mL of Reagent Grade DI water is used.

- 6.3.2 Record the sample ID, size (in grams (g) and in milliliters (mL)) and the balance and pipet IDs and errors Benchsheet 690-1.

- 6.4 Sample Aliquot size will vary depending on the sample matrix and the anticipated level of Tc-99 activity. See the Project Leader regarding the recommended sampling sizes. Record all sample ID, size (g or mL), balance or volumetric ID and error information on Benchsheet 690-1.

- 6.4.1 Single Phase Liquid Sample – typically use approximately 10–20 mL, but up to 100 mL of sample may be used in a 250 mL separatory funnel.

- 6.4.1.1 Accurately pipet a known volume of sample into a beaker of appropriate size or a quartz crucible (depending upon the digestion method used). Record the sample ID, the sample volume, the pipet ID and the pipet error on Benchsheet 690-1.

- 6.4.1.2 Prepare a duplicate for each sample adding ~100000 dpm of Tc-99 tracer. Record the sample ID, volume, pipet ID and error, the Tc-99 standard ID and the volume and pipet ID and error on Benchsheet 690-1.

- 6.4.2 Two Phase Liquid Sample – Typical sample sizes may be 10–20 mL, however up to 100 mL may be required for samples of low activity.

- 6.4.2.1 Weigh an empty beaker of appropriate size or a quartz crucible. Record the balance ID and error and the weight of the empty container (Tare Weight) on Benchsheet 690–1.
- 6.4.2.2 Pipet a known volume into the sample container.
- 6.4.2.3 Reweigh the container and sample.
- 6.4.2.4 Record the sample ID, the pipet ID and error, the weight and the volume on Benchsheet 690–1.
- 6.4.2.5 Prepare a duplicate of each sample adding ~100000 dpm of a NIST traceable Tc–99 radiotracer. In addition to the information requested above, record the standard ID, volume added, pipet ID and error on Benchsheet 690–1.
- 6.4.3 Solid Sample — Typical sample sizes may be 1–3 g, however a larger aliquot (~10 g) may be necessary for samples of expected low Tc–99 activity.
 - 6.4.3.1 Typical sizes: mixed Resin (1–3 g), filter paper (0.5 g) (depending on the amount of sample provided and its anticipated activity) and Soils (1–10 g). NIST traceable soil-type standards typically use 1–3 grams. Larger sample sizes may be recommended by the Project Leader.
 - 6.4.3.2 Weigh an empty beaker of appropriate size (100–150 mL recommended for soils) or a quartz crucible. Record the balance ID and error and the weight of the empty container (Tare weight) on Benchsheet 690–1.
 - 6.4.3.3 Transfer a sample aliquot to the empty container and reweigh. Record the sample ID and weight on Benchsheet 690–1.
 - 6.4.3.4 Prepare a duplicate of each sample adding ~100000 dpm of Tc–99 radiotracer. In addition to the information requested above, record the standard ID, volume added, pipet ID and error on Benchsheet 690–1.
- 6.5 **Sample Digestion** — Samples shall be digested using one of the following methods. Record all information on Benchsheet 690–1, including reagent lot numbers, which method of digestion was used and any deviations.
 - 6.5.1 Muffle Furnace Digestion: add 15 mL of concentrated ammonium hydroxide (NH_4OH) to the quartz crucible containing the sample. Slowly evaporate on a hot plate to dryness.
 - 6.5.1.1 Ignite in a muffle furnace at 400°C for two hours. Allow to cool.
 - 6.5.1.2 Add 15 mL of 6N H_2SO_4 and 2 grams of potassium persulfate ($\text{K}_2\text{S}_2\text{O}_8$). Boil the sample for 20 minutes.
 - 6.5.1.3 Proceed to Step 9.5.3.
 - 6.5.2 Hot Plate Digestion: for every 3 grams of sample add the following to the sample in the 150 mL beaker:

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6.5.2.1 5 mL of concentrated nitric acid (HNO_3), 5 mL DI water and ~1.5 g of $\text{K}_2\text{S}_2\text{O}_8$.

CAUTION: These chemicals should be added carefully as a vigorous reaction may occur.

6.5.2.2 Cover the beakers with a watch glass and gently reflux on a hot plate for > 1 hour. (The samples may also be allowed to sit overnight followed by a gentle reflux for ~1 hour.) **CAUTION:** The addition of heat may initiate a vigorous reaction. Observe the reflux process periodically to ensure samples do not overflow the beakers.

6.5.2.3 Proceed to Step 9.5.3.

6.5.3 Filter the cooled leachate through a Whatman #541 (or equivalent) filter paper into a beaker of an appropriate size.

6.5.3.1 Add 0.5 mL of Cesium and Manganese carriers and 1.0 mL of Cobalt carrier.

6.5.3.2 Wash the residue with sufficient 6 N H_2SO_4 to give a final volume of 45–50 mL.

6.5.3.3 Quantitatively transfer the sample to a 125 mL separatory funnel (or larger if needed) using 6N H_2SO_4 to effect the transfer.

6.5.3.4 Add 10 mL of concentrated H_2SO_4 and sufficient 1% KMnO_4 solution to maintain a pink color.

6.5.3.5 Proceed to Step 9.6.

6.6 **CAUTION:** Sample may become hot, wait until sample is cool to the touch before continuing.

6.7 Pipette 5.0 mL of concentrated tributyl phosphate (TBP) that has been previously equilibrated with 9 N H_2SO_4 into the separatory funnel (Step 8.5). Record reagent lot numbers, pipet ID and error on Benchsheet 690-1.

6.8 Shake for two minutes, allow phases to separate, drain the aqueous phase (the lower phase) and discard in the appropriate radioactive waste container.

6.9 Wash the TBP with 25 mL of 6 N sulfuric acid – 2 % hydrofluoric acid (H_2SO_4 –HF), shake for approximately two minutes. Allow the phases to separate and drain the aqueous phase (the lower phase) and discard in the appropriate radioactive waste container.

6.10 Wash the TBP solution with 25 mL of 4 N hydrochloric acid (HCl) and shake for about two minutes. Allow the phases to separate for 10 minutes so that sufficient extract (TBP) will be available for counting.

6.11 Drain and discard the aqueous phase and transfer the extracts (TBP) to a 15 mL centrifuge tube.

- 6.12 Centrifuge for 2 to 3 minutes to obtain a clear extract
- 6.13 Accurately pipette 4.0 mL (or an amount reasonably available) of extract into a scintillation vial and add 12.0 mL of scintillation cocktail. Record the volume of TBP recovered (pipet ID and error) and cocktail lot number on Benchsheet 690-1.
- 6.14 If a prepared efficiency standard and liquid scintillation background sample are available, proceed to Step 9.17.
- 6.15 Prepare a counting efficiency standard by placing a known amount of a NIST traceable Tc-99 standard (~100000 dpm), enough TBP to give a 4 mL total volume and 12.0 mL of scintillation cocktail into a scintillation vial. Label the vial with an efficiency ID. Record the sample ID, the standard ID, volume added, reagent lot numbers, pipet ID and error information on Benchsheet 690-1.
- 6.16 Prepare a background by placing 4.0 mL of TBP and 12.0 mL of scintillation cocktail into a scintillation vial. Label the vial as a sample background. Record the sample ID, the reagent lot numbers, pipet ID and error information on Benchsheet 690-1.
- 6.17 Transfer the samples, efficiency standard and background to the counting room for liquid scintillation analysis with a completed ICOC.

Attachment 2

B&W Nuclear Environmental Services, Inc.

LYNCHBURG TECHNOLOGY CENTER

TECHNICAL PROCEDURE

NUMBER TP-693 REV. NO. 0

TITLE I-129 ANALYSIS

1.0 APPLICABILITY

This procedure applies to all Nuclear Environmental Laboratories personnel who are involved with radiochemical separations.

2.0 SCOPE

This procedure provides the steps necessary to identify and quantify the Iodine-129 in radioactive waste sent to burial sites for disposal. Based on the requirements set forth in the United States Code of Federal Regulations, Title 10 Part 61 (10 CFR 61), "Licensing Requirements for Land Disposal of Low Level Waste," radwaste samples must be analyzed for radionuclide concentration.

3.0 SUMMARY OF METHOD

The sample is fused with sodium hydroxide (NaOH), oxidized with sodium hypochlorite (NaOCl) and extracted using either carbon tetrachloride (CCl₄) or chloroform (CHCl₃). It is then back-extracted into sodium bisulfite (NaHSO₃) and precipitated using silver nitrate (AgNO₃). The I-129 is quantitated using a Low Energy Photon Spectrometer (LEPS) detector at 29.7 keV or 39.5 keV. The sample yield is determined chemically with a natural iodine carrier. The sample yield is determined chemically with a natural iodine tracer measured gravimetrically or by ion chromatography.

- 3.1 Sample aliquots are obtained from the gamma scan fraction which was prepared in TP-398. The I-129 aliquot undergoes a solvent extraction separation.
- 3.2 All experimental information shall be recorded on Benchsheet 693-1 or the appropriate Notebook.

4.0 GLASSWARE

Before beginning the separation procedure it is strongly recommended that you gather together and label all of the glassware necessary for the separation of iodine. (All glassware used in this separation procedure shall have been cleaned according to TP-655.) ALL LABELS MUST CONTAIN THE COMPLETE WORKING LABORATORY SAMPLE NUMBER.

The glassware listed below is the glassware that is expected to be used for each sample in a batch. This listing does not include miscellaneous glassware to be used for reagents or chemicals.

SAMPLES

- 4.1 One nickel crucible.
- 4.2 One filter funnel.
- 4.3 One 250 mL beaker.
- 4.4 Two 250 mL separatory funnels (or appropriate size).
- 4.5 One 25 mL volumetric flask.
- 4.6 One 50 mL beaker.

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- 4.7 Two liquid scintillation vials.
- 4.8 One 1 1/4" flat planchet.
- 4.9 One plastic petri dish.
- 4.10 One 10 mL volumetric flask.

EFFICIENCY STANDARDS

- 4.11 Two — Three 50 mL beakers.
- 4.12 Two — Three 1 1/4" flat planchets.
- 4.13 Two — Three plastic petri dishes.

CARRIER

- 4.14 One 25 mL volumetric flask.
- 4.15 One 10 mL volumetric flask.

5.0 REAGENTS

- 5.1 **Iodine Carrier (10 mg I/mL)** — Dissolve 13.0 g potassium iodide (KI) in 500 mL of reagent grade water and dilute to one liter with reagent grade water.
- 5.2 **5% Sodium Hypochlorite (NaOCl)** — Purchase a prepared solution from an approved vendor.
- 5.3 **5% Sodium Hydroxide (NaOH)** — Dissolve 50 g of NaOH in 400 mL of reagent grade water and dilute to one liter with reagent grade water after allowing the solution to cool.
- 5.4 **20% Sodium Bisulfite (NaHSO₃)** — PREPARE FRESH JUST PRIOR TO USE — Dissolve 1.04 g NaHSO₃ in 5 mL of reagent grade water and dilute to 10 mL with reagent grade water.
- 5.5 **0.1 N Silver Nitrate Solution (AgNO₃)** — Purchase a prepared solution from an approved vendor.
- 5.6 **I-129 Tracer** — NIST traceable or calibrated against a NIST traceable standard.
- 5.7 Ethyl Alcohol (EtOH).
- 5.8 Sodium Hydroxide (NaOH).

6.0 IODINE-129 SAMPLE PREPARATION AND ANALYSIS

Sample aliquots shall be removed from the gamma scan fraction unless otherwise indicated by the project leader. Any deviations from this procedure shall be documented on Benchsheet 693-1 or in the appropriate notebook.

- 6.1 Each sample batch processed must contain a QC sample, a reagent blank, and a replicate of one sample for each batch of ≤ 10 . If more than 10 but less than 20 samples are processed in a batch then a replicate of two samples must be processed.

6.2 **Laboratory Control Sample** — A laboratory control sample is a sample comprised of an approximation of the matrix of one of the samples being processed in the batch. It is spiked with a known amount of I-129 radiotracer. Whenever possible, use a laboratory control stock sample. If no laboratory control available, the laboratory control sample may be prepared as follows.

6.2.1 Choose a matrix for the laboratory control sample that will best represent a typical sample of the batch to be processed.

6.2.2 See Section 10.4 regarding recommended sample sizes.

6.2.3 Add a known amount of a NIST traceable I-129 standard to the laboratory control sample matrix. Record the laboratory control sample preparation information.

6.3 **Reagent Blank** — A reagent blank is comprised of an aliquot of reagent grade water. NO RADIOTRACER SPIKE IS ADDED!

6.3.1 Up to 250 mL of Reagent Grade DI water is used. Record all sample information.

6.4 **Sample Aliquot** size will vary depending on the sample matrix and the anticipated level of I-129 activity. See the Project Leader regarding the recommended sampling sizes. Record all sample information.

6.4.1 **For a Single Phase Liquid Sample** — Shake the sample to homogenize. Liquid samples typically use approximately 10–20 mL, but up to 250 mL of sample may be used in a 500 mL beaker. Add 1.0 mL iodide carrier (approximately 18 mg) and proceed with Step 10.6 of this procedure. Record the carrier information.

6.4.2 **If the Sample is a Two-Phase Liquid** — Shake the sample to homogenize. Pipet a known aliquot of 10–20 mL or up to 40 mL into a nickel crucible. Record all sample information.

6.4.3 **If the Sample is a Solid** — accurately weigh an appropriate amount of the sample into a nickel crucible. Typical sample sizes may be 1–3 g, however a larger aliquot (~10 g) may be necessary for samples of expected low I-129 activity. Record all sample information.

6.4.3.1 **Mixed Resin** — typically use 1–3 g.

6.4.3.2 **Filter paper** — typically use 0.5 g, depending on the amount of sample provided and its anticipated activity.

6.4.3.3 **Soils** — A clean, characterized soil sample typically uses 1–5 grams. NIST traceable soil-type standards typically use ~1 gram. Larger sample sizes may be recommended by the Project Leader.

6.5 **Sample Preparation** — Record all information on Benchsheet 693-1 or the appropriate Notebook.

6.5.1 Add 1.0 mL of iodide carrier (approximately 18 mg) to the sample.

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- 6.5.2 Add 4.0 mL of DI water, 3.0 mL of ethyl alcohol and 10.0 g of sodium hydroxide (NaOH).
 - 6.5.3 Heat gently on a hot plate or equivalent, until all alcohol has been evaporated.
 - 6.5.4 Place the crucible in a furnace and slowly raise the temperature to 600°C over a 2–3 hour period. Hold the temperature at 600°C for 2 hours.
- NOTE:** If using a 75 mL nickel crucible, place the crucible on a tray prior to placing it in the muffle furnace. It has been found that when the crucible bottom rests on the bottom of the furnace, the combination of heat and chemicals causes a weakening (possible leaks) in the crucible.
- 6.5.5 Cool the crucible and add approximately 15 mL of reagent grade water. Boil to dissolve the melt.
 - 6.5.6 Filter the dissolved liquid through a Whatman # 41 or equivalent filter paper, into a 250 mL beaker.
 - 6.5.7 Repeat Steps 10.5.5 and 10.5.6 until all of the melt has been dissolved and filtered. If the sample will not completely dissolve proceed to Step 10.5.8. If complete dissolution is attained proceed to Step 10.6.
 - 6.5.8 Return the sample (in the filter paper) to the crucible and add 15 mL of 5% NaOH solution.
 - 6.5.9 Heat the solution in the crucible for about 30 minutes to dissolve the melt.
 - 6.5.10 Filter the solution through a Whatman 41 or equivalent filter paper. Combine the filtrate with that from Step 10.5.6.
 - 6.5.11 Stir the filtered solution for ~20 minutes.
- 6.6 **CAUTION:** The following step MUST be performed in a hood. While stirring, add 20 mL of 5% sodium hypochlorite (NaOCl or household bleach) solution. Next add concentrated nitric acid (HNO_3) until the pH of the solution is about 1. If the sample was treated with NaOH over 20 mL of concentrated HNO_3 will be required. When proper pH has been reached, all visible reaction will cease and solution will become yellowish to clear in color.
- SPECIAL NOTE:** This step must be performed in a fume hood because toxic chlorine gas is evolved. This-oxidizes the iodide to iodine so that it may be extracted by the carbon tetrachloride or chloroform. At this time chloride contaminants should be removed by oxidation of the chloride to chlorine gas - which volatilizes. Bromide should remain in its oxidation state to be separated during the extraction to follow.
- 6.7 Transfer the solution to a 250 mL separatory funnel labeled A.
- NOTE:** If the sample is a liquid with a volume greater than 100 mL use a 500 mL separatory funnel. Effect the transfer with DI H_2O .
- 6.8 Add 2 grams of hydroxylamine hydrochloride (H_2NOHHCl) and 50.0 mL of carbon tetrachloride (CCl_4) or chloroform (CHCl_3) to the separatory funnel. Stopper only after bubbling stops. Gently agitate and release pressure before proceeding to Step 10.8.

NOTE: Due to new restrictions on the use of CCl_4 , conversion to another extractant is necessary. See the Project Leader to determine when this conversion is to be made and the extractant to be used if other than CCl_4 .

- 6.9 Shake the funnel for 2 minutes and drain the organic phase (lower layer) into a 250 mL separatory funnel labeled B. It may take several minutes for complete phase separation to occur.

NOTE: The organic phase will become pink in the presence of iodine.

- 6.10 Add 1 g of hydroxylamine hydrochloride and 50 mL of CCl_4 to the aqueous phase in funnel A and re-extract by shaking for 2 minutes.

- 6.11 Allow phase separation to occur. Drain the organic phase (lower phase) into funnel B. Discard the aqueous phase in funnel A into the appropriate radioactive waste container.

- 6.12 Add 20 mL of DI water and 10 drops of freshly prepared 20% sodium bisulfite (NaHSO_3) to the funnel B containing the organic phase and shake for 2 minutes. This will strip the iodine from the organic phase into the aqueous phase. Drain and discard the organic phase into the appropriate radioactive waste container.

- 6.13 Transfer the aqueous phase into a clean labeled 25 mL volumetric flask. Dilute to the mark with DI water. Transfer the solution into a 50 mL beaker. Remove a 1.0 mL aliquot and transfer to a syringe fitted with an AcrodiscTM filter (0.45 μm , 10 mm). Filter into a labelled scintillation vial for use later in the IC yield analysis. Record all information.

- 6.14 Add 1 mL of concentrated HNO_3 and 2 mL of 0.1 N silver nitrate (AgNO_3) solution to the beaker. Mix well and allow to stand for ~5 minutes.

- 6.15 Weigh a labelled 1 1/4" planchet with a 0.45 μm glass fiber millipore or equivalent filter along with a piece of two sided tape. Record the weight information. (The sample will be mounted on the BOTTOM of the planchet, therefore label the top side of the planchet.)

- 6.16 Using a 25 mm filter funnel, filter with suction through the preweighed 0.45 μm glass fiber filter using DI water to effect the transfer. Wash the precipitate with 5 mL of DI water.

- 6.17 Secure the filter paper on the preweighed planchet with the two sided tape. Weigh the planchet to the nearest 0.1 mg. Air dry the sample for ~30 minutes. Record the weight.

- 6.18 Cover the planchet with x-ray film using tape to hold the film in planchet. Label the tape.

- 6.19 If no prepared efficiency standards are available, prepare 2-3 I-129 standard planchets (one for each available LEPS detector) as follows:

- 6.19.1 Add 25 mL of DI water, 1.0 mL of iodide carrier (from the same carrier stock solution as used in the analysis of the samples) and an aliquot of an NIST traceable I-129 radiotracer solution of ~40,000 dpm. Record all standard preparation information.

- 6.19.2 Proceed with Steps 10.13–10.17.

7.0 YIELD ANALYSIS

- 7.1 Remove 0.2 mL of the yield aliquot from Step 10.12 and dilute to 10 mL with DI water. Transfer the solution through an AcrodiscTM/syringe (0.45 µm, 10 mm acrodisc) filtration apparatus to a clean labelled scintillation vial for transfer to the analytical laboratory using a completed ICOC form for analysis by ion chromatography. Record all yield information and error information.
- 7.2 Prepare a 100% iodide carrier solution as follows:
 - 7.2.1 Add 20 mL of DI water and 1.0 mL of the iodide carrier stock solution used for the sample analyses to a 25 mL volumetric flask. Record the volumetric error.
 - 7.2.2 Transfer the solution to a 50 mL beaker. Remove 1.0 mL of solution to a labeled scintillation vial.
 - 7.2.3 Place a 0.2 mL of the aliquot from Step 11.2.1 in a 10 mL volumetric flask and dilute to the mark with DI water. Transfer the solution to a clean, labeled scintillation vial and transfer to the analytical laboratory with the samples from Step 11.1. Record all carrier information.
- 7.3 Transfer the samples and standards to the counting room along with a completed ICOC form.

Attachment 3

B&W Nuclear Environmental Services, Inc.

LYNCHBURG TECHNOLOGY CENTER

TECHNICAL PROCEDURE

NUMBER TP-691 REV. NO. 0

TITLE Carbon-14/Tritium Analysis for 10 CFR 61

Appendix C

1.0 SUMMARY OF METHOD

Sample aliquots are obtained from the gamma scan fraction which was prepared in TP-398. The C-14/H-3 aliquot undergoes oxidation, separation, and collection as carbon dioxide and water respectively.

All experimental information shall be recorded on Benchsheet 691-1 or in the appropriate notebook.

2.0 EQUIPMENT

All reusable glassware used in this separation procedure shall be cleaned according to TP-655.

- 2.1 One ion-exchange column (at least 0.5X15 cm) with stopcock, per H-3 sample (including blanks and recovery samples).
- 2.2 Six liquid scintillation vials per sample (including blanks and recovery samples).
- 2.3 Four liquid scintillation vials for efficiency and background standards.
- 2.4 Oxidizer, Packard Instruments, or equivalent.
- 2.5 Packard "combust-o-cones" or equivalent.

3.0 REAGENTS

- 3.1 IRN-150 resin or equivalent. Crack the resin slightly. Condition the resin with reagent grade water. Shake, then allow the resin to settle. Decant and discard the water along with any fine resin particles. Store the resin in reagent grade water as a slurry.
- 3.2 Packard Permafluor E/Permafluor V or equivalent C-14 cocktail.
- 3.3 Packard Carbosorb C-14 cocktail or equivalent.
- 3.4 Instagel scintillation cocktail or equivalent.
- 3.5 C-14 Standard — NIST traceable or calibrated against a NIST traceable standard.
- 3.6 H-3 Standard — NIST traceable or calibrated against a NIST traceable standard.
- 3.7 Methanol
- 3.8 Industrial grade nitrogen or equivalent.
- 3.9 Industrial grade oxygen or equivalent.

4.0 C-14/H-3 SAMPLE PREPARATION

Sample aliquots shall be removed from the gamma scan fraction prepared in TP-398 unless otherwise indicated by the Project Leader. Any deviations from this procedure shall be documented on Benchsheet 691-1 or in the appropriate Notebook.

- 4.1 Each sample batch processed must contain a laboratory control sample, a reagent blank, and a replicate of one sample for each batch of ≤ 10 . If more than 10 but fewer than 20 samples are processed in a batch, then a replicate of at least two samples must be processed.
- 4.2 **Laboratory Control Sample** — This sample is comprised of an approximation of the matrix of one of the samples in the batch. It is spiked with a known amount of C-14/H-3 radiotracer. Whenever possible, use a QC stock solution. If no QC stock solution is available, a laboratory control sample may be prepared as follows:
 - 4.2.1 Choose a matrix that will best represent a typical sample of the batch to be processed.
 - 4.2.2 See Section 10.4 regarding recommended sample sizes.
 - 4.2.3 Add a known amount of a NIST traceable C-14/H-3 standard to the appropriate matrix. Record all information.
- 4.3 **Reagent Blank** — A reagent blank is comprised of a combusto-cone and combust-aid pad or equivalent. NO RADIOTRACER SPIKE IS ADDED!
- 4.4 **Sample Preparation** — Record all information on Benchsheet 691-1 or the appropriate Notebook.
 - 4.4.1 For a Single Phase Liquid — Shake the sample to homogenize. Pipet up to 0.5 mL into a prepared combusto cone just before analysis. Record all sample information.
 - 4.4.2 For a Two-Phase Liquid — Shake the sample to homogenize. Pipet up to 0.5 mL into a prepared, preweighed combusto cone just before analysis. Immediately reweigh the cone containing the sample. Record all sample information.
 - 4.4.3 For a solid sample — Accurately weigh a prepared combusto-cone. Place up to 0.5 g of homogenized sample into the cone and immediately reweigh. Record all sample information.
- 4.5 **Sample Analysis**
 - 4.5.1 OXIDIZER START UP: Turn on the Nitrogen and Oxygen gas (at the bottle and at the instrument).
 - 4.5.2 Check the methanol bottle to insure that the level is adequate (~2/3 full) and the tube is submerged.

CAUTION: DO NOT OVERFILL THE BOTTLE.
 - 4.5.3 With the power off, remove the overflow trap and replace the glass wool contained therein. Reconnect the trap. CAUTION: TO PREVENT PERSONAL INJURY THE POWER MUST BE OFF BEFORE PERFORMING THIS TASK.
 - 4.5.4 Turn the oxidizer power on and allow ~10 minutes for the instrument to warm up. During this time check all fluid levels and make additions as necessary. To facilitate cap removal, be sure that the vent/pressure switch is in the vent position and the pumps are off. Each reservoir should be at least half full before operation begins.

- 4.5.5 After the warm up period, check the fluid dispensing levels by first switching the vent/pressure switch to the on position then toggle each reservoir on/off switch to the on position one at a time to check the volume dispensed. For example, turn on the reagent grade water, leaving the Permafluor and Carbosorb off. Initiate a burn cycle and collect the eluate. Repeat this sequence with the Permafluor and Carbosorb until all liquid levels are producing the desired volume. The optimum volumes are as follows:

REAGENT GRADE WATER = 5 mL

PERMAFLUOR = 10 mL

CARBOSORB = 5 mL

Once all fluids are yielding the desired volume, switch all on/off switches to the on position. The instrument is now ready for operation.

4.6 Preanalysis Blank

- 4.6.1 PREANALYSIS BLANK ALIQUOT: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time. Place a combusto-cone and pad into the combustion basket and initiate the burn cycle.
- 4.6.2 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.6.3 PREANALYSIS BLANK #1: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.
- 4.6.4 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.6.5 PREANALYSIS BLANK #2: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.
- 4.6.6 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must undergo a cation exchange clean-up column before analysis.

4.7 Sample Aliquot

- 4.7.1 SAMPLE ALIQUOT: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time. Place the combusto-cone containing the sample into the combustion basket and initiate the burn cycle.

- 4.7.2 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.7.3 SAMPLE BLANK #1: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time.
- 4.7.4 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.7.5 SAMPLE BLANK #2: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time.
- 4.7.6 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.7.7 Repeat Section 10.7 for each sample in the batch then proceed to Step 10.8.

4.8 Carbon-14 Recovery

- 4.8.1 C-14 RECOVERY ALIQUOT: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time. Place a combusto-cone with pad into the combustion basket. Pipet the appropriate amount of C-14 tracer (~20,000 dpm) into the cone and initiate the burn cycle.
- 4.8.2 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.8.3 C-14 RECOVERY BLANK #1: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.
- 4.8.4 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.8.5 C-14 RECOVERY BLANK #2: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.

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- 4.8.6 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.

4.9 Tritium Recovery

- 4.9.1 H-3 RECOVERY ALIQUOT: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time. Place a combusto-cone with pad into the combustion basket. Pipet the appropriate amount of H-3 tracer (~20,000 dpm) into the cone and initiate the burn cycle.
- 4.9.2 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.9.3 H-3 RECOVERY BLANK #1: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.
- 4.9.4 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.9.5 H-3 RECOVERY BLANK #2: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.
- 4.9.6 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.

4.10 Postanalysis Blank

- 4.10.1 POSTANALYSIS BLANK ALIQUOT: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time. Place a combusto-cone with pad into the combustion basket and initiate the burn cycle.
- 4.10.2 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.10.3 POSTANALYSIS BLANK #1: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.

- 4.10.4 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.
- 4.10.5 POSTANALYSIS BLANK #2: Remove the caps from two scintillation vials and place the vials into the sample collection slots on the oxidizer. Set the oxidizer burn time for the desired length of time and initiate the burn cycle.
- 4.10.6 Label the caps, removed from the scintillation vials, with the appropriate working lab numbers. When the burn cycle is complete, secure the caps on the appropriate vials. The C-14 fraction is ready for analysis. The H-3 fraction must be passed through a cation exchange clean-up column before analysis.

4.11 Efficiency Standards

- 4.11.1 If no prepared efficiency standards are available, prepare a C-14 and/or a H-3 standard as follows: For CARBON-14, pipet a known amount (~20,000 dpm) of the appropriate NIST traceable standard into a labeled scintillation vial containing 10 mL of Permafluor and 5 mL of Carbo-sorb (or equivalent). Shake well. For TRITIUM pipet a known amount (~20,000 dpm) of the appropriate NIST traceable standard into a labeled scintillation vial containing 10 mL of reagent grade water. Add 10 mL of Instagel or equivalent. Shake well.

4.12 Background Standards

- 4.12.1 If no prepared background standards are available, prepare a C-14 and/or a H-3 standard as follows: For CARBON-14, pipet 10 mL of Permafluor and 5 mL of Carbo-sorb into a labeled scintillation vial. Shake well. For TRITIUM pipet 10 mL of reagent grade water and 10 mL of Instagel or equivalent into a labeled scintillation vial. Shake well.

4.13 Tritium Purification

- 4.13.1 For each of the pre and postanalysis blanks, sample aliquots and recovery aliquots, prepare a 10 cm resin bed using prepared IRN-150 resin (see reagents section for preparation guidelines). Condition the column with 10 mL of reagent grade water.
- 4.13.2 Place a clean scintillation vial under the column. Load the sample aliquot on the column and elute to the top of the resin.
- 4.13.3 Load BLANK #1 on the column and elute to the top of the resin. Rinse the scintillation vial (for Blank #1) twice, with ~1 mL of reagent grade water. Continue collecting the eluate in the initial scintillation vial until 10 mL has been collected.
- 4.13.4 When 10 mL of eluate has been collected, replace the collection vial with the rinsed, BLANK #1, scintillation vial. To the initial collection vial, add 10 mL of instagel (or equivalent). Place the appropriately labeled cap on the vial and shake well.
- 4.13.5 Elute the rinse solution to the top of the resin.

- 4.13.6 Continue collecting the eluate into the BLANK #1 vial until 10 mL of elute has been collected. Load BLANK #2 on the column and elute to the top of the resin. Rinse the scintillation vial (for Blank #2) twice, with ~1 mL of reagent grade water. Continue collecting the eluate in the BLANK #1 scintillation vial until 10 mL has been collected.
- 4.13.7 When 10 mL of eluate has been collected, replace the collection vial with the rinsed, BLANK #2, scintillation vial. To the BLANK #2 collection vial, add 10 mL of instagel (or equivalent). Place the appropriately labeled cap on the vial and shake well.
- 4.13.8 Rinse the column with reagent water grade until 10 mL of eluate has been collected in the BLANK #2 vial.
- 4.13.9 Add 10 mL of Instagel or equivalent to the vial (BLANK #2). Place the appropriately labeled cap on the vial and shake well.
- 4.14 Wipe the exterior of all vials with a lint-free cloth to remove any particles that may interfere with the scintillation analysis.
- 4.15 Transfer all samples, efficiency and background standards to the Counting Room for Liquid Scintillation beta analysis. All transfers must be accompanied by a completed ICOC form.

Appendix D
Leaching Data for Peach Bottom

Table D-1. Peach Bottom sample #4 cumulative fraction releases, release rates, effective diffusivities, and leachability indexes.

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm ⁻² ·s ⁻¹)	RELEASE RATE (cm ⁻² ·s ⁻¹)	RELEASE RATE (Ci·yr ⁻¹)	INCREMENTAL RELEASE RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² ·s ⁻¹)	LEACH INDEX
1. (30a)	3.47E-04	6.91E-03	---	---	---	---	---	---	---
2. (2hr)	8.44E-02	4.40E-02	3.90E-08	3.49E-11	1.92E-04	3.94E-05	3.94E-05	1.32E-13	1.29E+01
3. (7hr)	2.93E-01	1.04E-02	3.73E-09	3.34E-12	1.83E-06	9.36E-08	4.88E-05	9.81E-15	1.40E+01
4. (1d)	1.00E+00	1.80E-03	1.89E-10	1.70E-13	9.30E-07	1.61E-06	5.04E-05	8.77E-17	1.61E+01
5. (2d)	2.00E+00	7.38E-03	5.49E-10	4.92E-13	2.70E-08	6.61E-06	5.70E-05	1.81E-15	1.47E+01
6. (3d)	3.00E+00	8.05E-03	4.51E-10	4.04E-13	2.21E-08	5.42E-06	6.24E-05	2.07E-15	1.47E+01
7. (4d)	4.00E+00	3.56E-02	2.65E-09	2.37E-12	1.30E-05	3.19E-05	9.43E-05	1.01E-13	1.30E+01
8. (5d)	5.00E+00	2.26E-02	1.68E-09	1.51E-12	8.27E-06	2.03E-05	1.15E-04	5.24E-14	1.33E+01
Mean:		6.89E-09	6.17E-12	3.39E-06			4.27E-14	1.41E+01	
Standard Deviation:		1.32E-08	1.18E-11	6.47E-06			5.01E-14	1.07E+00	
FILE NAME: Fe-55									
SAMPLE ID: PEACH BOTTOM #4									
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm ⁻² ·s ⁻¹)	RELEASE RATE (cm ⁻² ·s ⁻¹)	RELEASE RATE (Ci·yr ⁻¹)	INCREMENTAL RELEASE RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² ·s ⁻¹)	LEACH INDEX
1. (30a)	3.47E-04	1.74E-03	---	---	---	---	---	---	---
2. (2hr)	8.44E-02	8.02E-03	7.11E-09	4.69E-10	3.49E-05	5.29E-04	5.29E-04	2.37E-11	1.08E+01
3. (7hr)	2.93E-01	1.88E-03	8.73E-10	4.44E-11	3.31E-06	1.24E-04	6.53E-04	1.74E-12	1.18E+01
4. (1d)	1.00E+00	1.69E-03	1.78E-10	1.17E-11	8.73E-07	1.11E-04	7.64E-04	4.18E-13	1.24E+01
5. (2d)	2.00E+00	1.94E-05	1.44E-12	9.50E-14	7.08E-09	1.28E-06	7.86E-04	8.75E-17	1.82E+01
6. (3d)	3.00E+00	1.41E-04	1.05E-11	6.92E-13	5.15E-08	9.29E-06	7.75E-04	8.08E-15	1.42E+01
7. (4d)	4.00E+00	7.23E-04	5.38E-11	3.55E-12	2.64E-07	4.76E-05	8.23E-04	2.25E-13	1.26E+01
8. (5d)	5.00E+00	5.53E-04	4.12E-11	2.71E-12	2.02E-07	3.65E-06	8.59E-04	1.70E-13	1.26E+01
Mean:		1.15E-09	7.60E-11	5.68E-08			3.75E-12	1.29E+01	
Standard Deviation:		2.44E-09	1.61E-10	1.20E-05			8.16E-12	1.86E+00	
FILE NAME: Co-60									
SAMPLE ID: PEACH BOTTOM #4									
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm ⁻² ·s ⁻¹)	RELEASE RATE (cm ⁻² ·s ⁻¹)	RELEASE RATE (Ci·yr ⁻¹)	INCREMENTAL RELEASE RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² ·s ⁻¹)	LEACH INDEX
1. (30a)	3.47E-04	1.18E-02	---	---	---	---	---	---	---
2. (2hr)	8.44E-02	1.08E-01	9.53E-08	2.54E-10	4.68E-04	2.87E-04	2.87E-04	6.98E-12	1.12E+01
3. (7hr)	2.93E-01	9.53E-02	3.41E-08	9.09E-11	1.67E-04	2.54E-04	5.42E-04	7.33E-12	1.11E+01
4. (1d)	1.00E+00	1.41E-01	1.48E-08	3.95E-11	7.28E-05	3.76E-04	9.17E-04	4.77E-12	1.19E+01
5. (2d)	2.00E+00	1.13E-01	8.40E-09	2.24E-11	4.13E-05	3.01E-04	1.22E-03	3.76E-12	1.14E+01
6. (3d)	3.00E+00	8.08E-02	6.00E-09	1.60E-11	2.95E-05	2.15E-04	1.43E-03	3.27E-12	1.15E+01
7. (4d)	4.00E+00	6.25E-02	4.68E-09	1.24E-11	2.29E-05	1.67E-04	1.60E-03	2.76E-12	1.18E+01
8. (5d)	5.00E+00	4.84E-02	3.60E-09	9.61E-12	1.77E-05	1.29E-04	1.73E-03	2.13E-12	1.17E+01
Mean:		2.38E-08	6.36E-11	1.17E-04			4.43E-12	1.14E+01	
Standard Deviation:		3.08E-08	8.21E-11	1.51E-04			1.88E-12	1.86E-01	
FILE NAME: Ni-63									
SAMPLE ID: PEACH BOTTOM #4									
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm ⁻² ·s ⁻¹)	RELEASE RATE (cm ⁻² ·s ⁻¹)	RELEASE RATE (Ci·yr ⁻¹)	INCREMENTAL RELEASE RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² ·s ⁻¹)	LEACH INDEX
1. (30a)	3.47E-04	6.52E-04	---	---	---	---	---	---	---
2. (2hr)	8.44E-02	6.04E-03	5.35E-09	8.15E-10	2.63E-06	9.20E-04	9.20E-04	7.17E-11	1.01E+01
3. (7hr)	2.93E-01	6.76E-03	2.42E-09	3.68E-10	1.19E-05	1.03E-03	1.95E-03	1.20E-10	9.92E+00
4. (1d)	1.00E+00	8.30E-03	8.74E-10	1.33E-10	4.29E-06	1.27E-03	3.22E-03	5.41E-11	1.03E+01
5. (2d)	2.00E+00	5.34E-03	3.97E-10	6.06E-11	1.95E-06	8.14E-04	4.03E-03	2.75E-11	1.06E+01
6. (3d)	3.00E+00	3.72E-03	2.77E-10	4.22E-11	1.36E-06	5.67E-04	4.60E-03	2.27E-11	1.08E+01
7. (4d)	4.00E+00	2.82E-03	1.95E-10	2.97E-11	9.58E-07	3.99E-04	5.00E-03	1.58E-11	1.08E+01
8. (5d)	5.00E+00	4.21E-03	3.13E-10	4.78E-11	1.54E-06	6.42E-04	5.64E-03	5.26E-11	1.03E+01
Mean:		1.40E-09	2.14E-10	6.89E-06			5.21E-11	1.04E+01	
Standard Deviation:		1.76E-09	2.69E-10	8.67E-06			3.34E-11	2.85E-01	

Appendix D

Table D-1. (continued).

FILE NAME: Sr-90
SAMPLE ID: PEACH BOTTOM #4

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm⁻²·s⁻¹)	RELEASE RATE (cm⁻²·s⁻¹)	RELEASE RATE (Ci·yr⁻¹)	INCREMENTAL RELEASE (Ci)	CUMULATIVE RELEASE (Ci)	EFFECTIVE DIFFUSIVITY (cm²·s⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	3.43E-06	---	---	---	---	1.64E-03	1.64E-03	2.28E-10
2. (2hr)	8.44E-02	1.57E-05	1.39E-11	1.45E-00	8.82E-08	1.64E-03	1.64E-03	2.28E-10	9.64E+00
3. (7hr)	2.93E-01	N/D	N/D	N/D	N/D	N/D	1.64E-03	N/D	N/D
4. (1d)	1.00E+00	N/D	N/D	N/D	N/D	N/D	1.64E-03	N/D	N/D
5. (2d)	2.00E+00	3.48E-06	2.59E-13	2.71E-11	1.27E-09	3.64E-04	2.00E-03	5.50E-12	1.13E+01
6. (3d)	3.00E+00	N/D	N/D	N/D	N/D	N/D	2.00E-03	N/D	N/D
7. (4d)	4.00E+00	3.47E-06	2.58E-13	2.70E-11	1.27E-09	3.63E-04	2.37E-03	1.31E-11	1.09E+01
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	N/D	2.37E-03	N/D	N/D
Mean:		4.80E-12	5.02E-10	2.36E-08			8.21E-11	1.06E+01	
Standard Deviation:		5.42E-12	5.72E-10	3.15E-08			1.03E-10	5.91E-01	

FILE NAME: Tc-99
SAMPLE ID: PEACH BOTTOM #4

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm⁻²·s⁻¹)	RELEASE RATE (cm⁻²·s⁻¹)	RELEASE RATE (Ci·yr⁻¹)	INCREMENTAL RELEASE (Ci)	CUMULATIVE RELEASE (Ci)	EFFECTIVE DIFFUSIVITY (cm²·s⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	N/D	---	---	---	---	1.26E-04	1.26E-04	1.35E-12
2. (2hr)	8.44E-02	5.95E-04	5.27E-10	1.12E-10	2.59E-06	1.26E-04	1.26E-04	1.19E+01	
3. (7hr)	2.93E-01	5.41E-04	1.93E-10	4.10E-11	9.50E-07	1.15E-04	2.41E-04	1.49E-12	1.18E+01
4. (1d)	1.00E+00	1.24E-03	1.30E-10	2.77E-11	6.40E-07	2.63E-04	5.04E-04	2.33E-12	1.16E+01
5. (2d)	2.00E+00	5.22E-03	3.88E-10	8.24E-11	1.91E-06	1.11E-03	1.61E-03	5.08E-11	1.03E+01
6. (3d)	3.00E+00	1.01E-03	7.53E-11	1.60E-11	3.70E-07	2.14E-04	1.83E-03	3.24E-12	1.15E+01
7. (4d)	4.00E+00	8.31E-04	6.19E-11	1.31E-11	3.04E-07	1.76E-04	2.00E-03	3.08E-12	1.15E+01
8. (5d)	5.00E+00	6.28E-04	4.68E-11	9.92E-12	2.30E-07	1.33E-04	2.14E-03	2.27E-12	1.16E+01
Mean:		2.03E-10	4.31E-11	9.99E-07			9.23E-12	1.15E+01	
Standard Deviation:		1.71E-10	3.64E-11	8.42E-07			1.70E-11	4.97E-01	

FILE NAME: I-129
SAMPLE ID: PEACH BOTTOM #4

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi·cm⁻²·s⁻¹)	RELEASE RATE (cm⁻²·s⁻¹)	RELEASE RATE (Ci·yr⁻¹)	INCREMENTAL RELEASE (Ci)	CUMULATIVE RELEASE (Ci)	EFFECTIVE DIFFUSIVITY (cm²·s⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	N/D	---	---	---	---	1.26E-04	1.26E-04	1.35E-12
2. (2hr)	8.44E-02	1.87E-04	1.48E-10	2.44E-08	7.27E-07	2.76E-02	2.76E-02	6.45E-08	7.19E+00
3. (7hr)	2.93E-01	N/D	N/D	N/D	N/D	N/D	2.76E-02	N/D	N/D
4. (1d)	1.00E+00	3.93E-05	4.13E-12	6.83E-10	2.03E-08	6.49E-03	3.41E-02	1.42E-09	8.85E+00
5. (2d)	2.00E+00	2.58E-04	1.92E-11	3.17E-09	9.41E-08	4.25E-02	7.66E-02	7.50E-08	7.12E+00
6. (3d)	3.00E+00	N/D	N/D	N/D	N/D	N/D	7.66E-02	N/D	N/D
7. (4d)	4.00E+00	1.91E-05	1.42E-12	2.35E-10	6.98E-09	3.15E-03	7.98E-02	9.85E-10	9.01E+00
8. (5d)	5.00E+00	1.01E-04	7.51E-12	1.24E-09	3.69E-08	1.67E-02	9.64E-02	3.55E-08	7.45E+00
Mean:		3.80E-11	5.95E-09	1.77E-07			3.55E-08	7.92E+00	
Standard Deviation:		5.63E-11	9.30E-09	2.77E-07			3.08E-08	8.28E-01	

FILE NAME: Cr
SAMPLE ID: PEACH BOTTOM #4

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug·cm⁻²·s⁻¹)	RELEASE RATE (cm⁻²·s⁻¹)	RELEASE RATE (g·yr⁻¹)	INCREMENTAL RELEASE (g)	CUMULATIVE RELEASE (g)	EFFECTIVE DIFFUSIVITY (cm²·s⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	N/D	---	---	---	---	1.57E-03	1.57E-03	2.08E-10
2. (2hr)	8.44E-02	4.00E+01	3.55E-05	1.39E-09	1.74E-01	1.57E-03	1.57E-03	9.68E+00	
3. (7hr)	2.93E-01	4.09E+01	1.46E-05	5.79E-10	7.19E-02	1.60E-03	3.17E-03	2.90E-10	9.54E+00
4. (1d)	1.00E+00	6.57E+01	6.92E-06	2.71E-10	3.40E-02	2.57E-03	5.74E-03	2.23E-10	9.65E+00
5. (2d)	2.00E+00	6.61E+01	4.92E-06	1.93E-10	2.42E-02	2.59E-03	8.33E-03	2.77E-10	9.56E+00
6. (3d)	3.00E+00	5.57E+01	4.15E-06	1.62E-10	2.04E-02	2.18E-03	1.05E-02	3.35E-10	9.48E+00
7. (4d)	4.00E+00	4.86E+01	3.62E-06	1.42E-10	1.78E-02	1.90E-03	1.24E-02	3.58E-10	9.45E+00
8. (5d)	5.00E+00	4.70E+01	3.50E-06	1.37E-10	1.72E-02	1.84E-03	1.42E-02	4.32E-10	9.37E+00
Mean:		1.05E-05	4.09E-10	5.14E-02			3.03E-10	9.53E+00	
Standard Deviation:		1.08E-05	4.24E-10	5.32E-02			7.25E-11	1.04E-01	

Table D-1. (continued).

FILE NAME: Fe SAMPLE ID: PEACH BOTTOM #4										
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² s ⁻¹)	RELEASE RATE (cm ⁻² s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	3.26E+01	---	---	---	---	---	---	---	
2. (2hr)	8.44E-02	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	
3. (7hr)	2.93E-01	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	
4. (1d)	1.00E+00	2.77E+01	2.91E-06	6.69E-13	1.43E-02	5.36E-06	6.36E-06	1.36E-15	1.49E+01	
5. (2d)	2.00E+00	N/D	N/D	N/D	N/D	6.36E-06	N/D	N/D	N/D	
6. (3d)	3.00E+00	2.61E+01	1.94E-06	4.46E-13	9.55E-03	5.99E-06	1.24E-06	2.53E-15	1.46E+01	
7. (4d)	4.00E+00	2.43E+01	1.81E-06	4.15E-13	8.88E-03	5.58E-06	1.79E-06	3.09E-15	1.45E+01	
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	1.79E-05	N/D	N/D	N/D	
Mean:		2.22E-06	5.10E-13	1.09E-02			2.33E-15	1.47E+01		
Standard Deviation:		4.92E-07	1.13E-13	2.42E-03			7.17E-16	1.51E-01		
FILE NAME: Co SAMPLE ID: PEACH BOTTOM #4										
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² s ⁻¹)	RELEASE RATE (cm ⁻² s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	1.54E+01	---	---	---	---	---	---	---	
2. (2hr)	8.44E-02	2.09E+01	1.87E-05	5.74E-09	9.28E-02	6.43E-03	6.43E-03	3.53E-09	8.45E+00	
3. (7hr)	2.93E-01	3.20E+01	1.14E-05	3.52E-09	5.70E-02	9.88E-03	1.63E-02	1.10E-08	7.98E+00	
4. (1d)	1.00E+00	2.94E+01	3.09E-06	9.51E-10	1.54E-02	9.05E-03	2.53E-02	2.75E-09	8.56E+00	
5. (2d)	2.00E+00	1.04E+01	7.77E-07	2.39E-10	3.87E-03	3.21E-03	2.86E-02	4.29E-10	9.37E+00	
6. (3d)	3.00E+00	1.22E+01	9.06E-07	2.79E-10	4.51E-03	3.75E-03	3.23E-02	9.90E-10	9.00E+00	
7. (4d)	4.00E+00	1.56E+01	1.16E-06	3.58E-10	5.78E-03	4.81E-03	3.71E-02	2.29E-09	8.64E+00	
8. (5d)	5.00E+00	1.61E+01	1.42E-06	4.38E-10	7.08E-03	5.89E-03	4.30E-02	4.42E-09	8.35E+00	
Mean:		5.35E-06	1.85E-09	2.66E-02			3.82E-09	8.82E+00		
Standard Deviation:		6.47E-06	1.99E-09	3.22E-02			3.25E-09	4.22E-01		
FILE NAME: Ni SAMPLE ID: PEACH BOTTOM #4										
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² s ⁻¹)	RELEASE RATE (cm ⁻² s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	N/D	---	---	---	---	---	---	---	
2. (2hr)	8.44E-02	4.87E+01	4.32E-05	2.90E-09	2.12E-01	3.27E-03	3.27E-03	9.07E-10	9.04E+00	
3. (7hr)	2.93E-01	4.63E+01	1.65E-05	1.11E-09	8.12E-02	3.11E-03	6.38E-03	1.09E-09	8.96E+00	
4. (1d)	1.00E+00	5.19E+01	5.46E-05	3.67E-10	2.88E-02	3.49E-03	9.87E-03	4.10E-10	9.39E+00	
5. (2d)	2.00E+00	5.22E+01	3.88E-06	2.61E-10	1.91E-02	3.51E-03	1.34E-02	5.10E-10	9.29E+00	
6. (3d)	3.00E+00	5.04E+01	3.76E-06	2.52E-10	1.85E-02	3.39E-03	1.88E-02	8.10E-10	9.09E+00	
7. (4d)	4.00E+00	5.03E+01	3.75E-06	2.52E-10	1.84E-02	3.38E-03	2.01E-02	1.13E-09	8.95E+00	
8. (5d)	5.00E+00	4.35E+01	3.24E-06	2.18E-10	1.59E-02	2.92E-03	2.31E-02	1.09E-09	8.96E+00	
Mean:		1.14E-05	7.66E-10	5.60E-02			8.51E-10	9.10E+00		
Standard Deviation:		1.37E-05	9.19E-10	6.72E-02			2.70E-10	1.62E-01		
FILE NAME: Zn SAMPLE ID: PEACH BOTTOM #4										
LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² s ⁻¹)	RELEASE RATE (cm ⁻² s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	5.15E+01	---	---	---	---	---	---	---	
2. (2hr)	8.44E-02	3.31E+01	2.93E-05	1.58E-09	1.44E-01	1.79E-03	1.79E-03	2.71E-10	9.57E+00	
3. (7hr)	2.93E-01	N/D	N/D	N/D	N/D	1.79E-03	N/D	N/D	N/D	
4. (1d)	1.00E+00	2.25E+01	2.37E-06	1.28E-10	1.16E-02	1.22E-03	3.01E-03	5.00E-11	1.03E+01	
5. (2d)	2.00E+00	2.61E+01	1.94E-06	1.05E-10	9.54E-03	1.41E-03	4.42E-03	8.20E-11	1.01E+01	
6. (3d)	3.00E+00	N/D	N/D	N/D	N/D	4.42E-03	4.42E-03	N/D	N/D	
7. (4d)	4.00E+00	N/D	N/D	N/D	N/D	4.42E-03	N/D	N/D	N/D	
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	4.42E-03	N/D	N/D	N/D	
Mean:		1.12E-05	6.06E-10	5.50E-02			1.35E-10	9.98E+00		
Standard Deviation:		1.28E-05	6.92E-10	6.29E-02			9.74E-11	3.08E-01		

Appendix D

Table D-1. (continued).

FILE NAME: B
SAMPLE ID: PEACH BOTTOM #4

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE RELEASE (g*yr ⁻¹)	CUMULATIVE RELEASE (g*yr ⁻¹)	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	3.43E+01	---	---	---	---	---	---	---
2. (2hr)	8.44E-02	3.19E+01	2.77E-05	6.05E-09	1.36E-01	6.82E-03	6.82E-03	3.94E-09	8.40E+00
3. (7hr)	2.93E-01	N/D	N/D	N/D	N/D	N/D	6.82E-03	N/D	N/D
4. (1d)	1.00E+00	3.46E+01	3.64E-06	7.93E-10	1.79E-02	7.54E-03	1.44E-02	1.92E-09	8.72E+00
5. (2d)	2.00E+00	3.48E+01	2.59E-06	5.64E-10	1.27E-02	7.58E-03	2.19E-02	2.38E-09	8.82E+00
6. (3d)	3.00E+00	3.48E+01	2.59E-06	5.64E-10	1.27E-02	7.58E-03	2.95E-02	4.05E-09	8.39E+00
7. (4d)	4.00E+00	N/D	N/D	N/D	N/D	N/D	2.95E-02	N/D	N/D
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	N/D	2.95E-02	N/D	N/D
Mean:			9.14E-06	1.99E-09	4.49E-02			3.07E-09	8.53E+00
Standard Deviation:			1.08E-05	2.34E-09	5.28E-02			9.39E-10	1.40E-01

FILE NAME: Picolinic Acid
SAMPLE ID: PEACH BOTTOM #4

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE RELEASE (g*yr ⁻¹)	CUMULATIVE RELEASE (g*yr ⁻¹)	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	N/D	---	---	---	---	---	---	---
2. (2hr)	8.44E-02	3.20E+04	2.84E-02	2.08E-09	1.39E+02	2.35E-03	2.35E-03	4.66E-10	9.33E+00
3. (7hr)	2.93E-01	3.26E+04	1.18E-02	8.53E-10	5.72E+01	2.39E-03	4.73E-03	6.45E-10	9.19E+00
4. (1d)	1.00E+00	6.14E+04	6.46E-03	4.74E-10	3.18E+01	4.50E-03	9.23E-03	6.84E-10	9.17E+00
5. (2d)	2.00E+00	5.81E+04	4.32E-03	3.17E-10	2.12E+01	4.26E-03	1.35E-02	7.52E-10	9.12E+00
6. (3d)	3.00E+00	3.64E+04	2.71E-03	1.98E-10	1.33E+01	2.66E-03	1.62E-02	5.01E-10	9.30E+00
7. (4d)	4.00E+00	2.85E+04	2.12E-03	1.55E-10	1.04E+01	2.09E-03	1.82E-02	4.31E-10	9.37E+00
8. (5d)	5.00E+00	2.21E+04	1.64E-03	1.21E-10	8.08E+00	1.62E-03	1.99E-02	3.35E-10	9.48E+00
Mean:			8.18E-03	6.00E-10	4.02E+01			5.45E-10	9.28E+00
Standard Deviation:			8.84E-03	6.48E-10	4.34E+01			1.40E-10	1.16E-01

Table D-2. Peach Bottom sample #8 cumulative fraction releases, release rates, effective diffusivities, and leachability indexes.

FILE NAME: C-14 SAMPLE ID: Peach Bottom #8										
LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	1.22E-02	---	---	---	---	---	---	---	
2. (2hr)	8.40E-02	1.18E-01	9.48E-08	8.55E-11	5.16E-04	1.07E-04	1.07E-04	1.09E-12	1.20E+01	
3. (7hr)	2.44E-01	9.35E-02	3.91E-08	3.53E-11	2.13E-04	8.45E-05	1.92E-04	1.37E-12	1.19E+01	
4. (1d)	9.39E-01	1.29E-01	1.24E-08	1.12E-11	6.78E-05	1.16E-04	3.08E-04	4.81E-13	1.23E+01	
5. (2d)	1.93E+00	1.18E-01	8.01E-09	7.23E-12	4.36E-05	1.07E-04	4.15E-04	5.19E-13	1.23E+01	
6. (3d)	2.92E+00	8.49E-02	5.75E-09	5.19E-12	3.13E-05	7.87E-05	4.91E-04	4.62E-13	1.23E+01	
7. (4d)	3.91E+00	7.01E-02	4.75E-09	4.29E-12	2.59E-05	6.33E-05	5.55E-04	4.48E-13	1.24E+01	
8. (5d)	4.90E+00	1.47E-01	9.98E-09	9.02E-12	5.44E-05	1.33E-04	5.88E-04	2.55E-12	1.16E+01	
Mean:			2.50E-08	2.25E-11	1.38E-04			9.88E-13	1.21E+01	
Standard Deviation:			3.05E-08	2.75E-11	1.66E-04			7.20E-13	2.75E-01	
FILE NAME: Fe-55 SAMPLE ID: Peach Bottom #8										
LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	1.91E-01	---	---	---	---	---	---	---	
2. (2hr)	8.40E-02	1.18E-01	9.30E-08	6.17E-09	5.07E-04	7.72E-03	7.72E-03	5.70E-09	8.24E+00	
3. (7hr)	2.44E-01	6.63E-02	2.77E-08	1.84E-09	1.51E-04	4.40E-03	1.21E-02	3.71E-09	8.43E+00	
4. (1d)	9.39E-01	5.27E-02	5.09E-09	3.38E-10	2.78E-05	3.50E-03	1.56E-02	4.36E-10	9.36E+00	
5. (2d)	1.93E+00	2.29E-02	1.55E-09	1.03E-10	8.43E-06	1.52E-03	1.71E-02	1.04E-10	9.98E+00	
6. (3d)	2.92E+00	1.04E-02	7.01E-10	4.65E-11	3.82E-06	6.87E-04	1.78E-02	3.70E-11	1.04E+01	
7. (4d)	3.91E+00	4.93E-03	3.34E-10	2.21E-11	1.82E-06	3.27E-04	1.81E-02	1.19E-11	1.09E+01	
8. (5d)	4.90E+00	4.80E-03	3.26E-10	2.16E-11	1.77E-06	3.18E-04	1.85E-02	1.46E-11	1.08E+01	
Mean:			1.84E-09	1.22E-09	1.00E-04			1.43E-09	9.74E+00	
Standard Deviation:			3.18E-09	2.11E-09	1.73E-04			2.14E-09	1.02E+00	
FILE NAME: Co-60 SAMPLE ID: Peach Bottom #8										
LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	1.21E-02	---	---	---	---	---	---	---	
2. (2hr)	8.40E-02	1.64E-01	1.31E-07	3.52E-10	7.13E-04	4.41E-04	4.41E-04	1.85E-11	1.07E+01	
3. (7hr)	2.44E-01	9.41E-02	3.94E-08	1.06E-10	2.14E-04	2.53E-04	6.93E-04	1.23E-11	1.09E+01	
4. (1d)	9.39E-01	2.03E-01	1.96E-08	5.28E-11	1.07E-04	5.47E-04	1.24E-03	1.06E-11	1.10E+01	
5. (2d)	1.93E+00	1.78E-01	1.20E-08	3.23E-11	6.55E-06	4.77E-04	1.72E-03	1.09E-11	1.10E+01	
6. (3d)	2.92E+00	1.18E-01	8.01E-09	2.15E-11	4.37E-06	3.18E-04	2.04E-03	7.94E-12	1.11E+01	
7. (4d)	3.91E+00	1.06E-01	7.19E-09	1.93E-11	3.92E-05	2.88E-04	2.32E-03	9.06E-12	1.10E+01	
8. (5d)	4.90E+00	2.34E-01	1.59E-08	4.26E-11	8.64E-05	6.28E-04	2.95E-03	5.69E-11	1.02E+01	
Mean:			3.33E-08	8.95E-11	1.81E-04			1.80E-11	1.09E+01	
Standard Deviation:			4.11E-08	1.10E-10	2.24E-04			1.62E-11	2.72E-01	
FILE NAME: Ni-63 SAMPLE ID: Peach Bottom #8										
LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	2.57E-05	---	---	---	---	---	---	---	
2. (2hr)	8.40E-02	1.11E-04	8.89E-11	1.36E-11	4.55E-07	1.71E-05	1.71E-05	2.78E-14	1.38E+01	
3. (7hr)	2.44E-01	7.02E-05	2.94E-11	4.50E-12	1.50E-07	1.08E-05	2.78E-05	2.22E-14	1.37E+01	
4. (1d)	9.39E-01	9.24E-05	8.93E-12	1.37E-12	4.57E-08	1.42E-06	4.20E-06	7.17E-15	1.41E+01	
5. (2d)	1.93E+00	3.77E-05	2.55E-12	3.91E-13	1.31E-08	5.78E-06	4.78E-06	1.51E-15	1.48E+01	
6. (3d)	2.92E+00	1.54E-05	1.04E-12	1.60E-13	5.34E-09	2.36E-06	5.02E-06	4.38E-16	1.54E+01	
7. (4d)	3.91E+00	N/D	N/D	N/D	N/D	N/D	5.02E-06	N/D	N/D	
8. (5d)	4.90E+00	N/D	N/D	N/D	N/D	N/D	5.02E-06	N/D	N/D	
Mean:			2.52E-11	4.01E-12	1.34E-07			1.18E-14	1.43E+01	
Standard Deviation:			3.30E-11	5.05E-12	1.69E-07			1.12E-14	6.91E-01	

Appendix D

Table D-2. (continued).

FILE NAME: Sr-90
SAMPLE ID: Peach Bottom #8

LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{Ci} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	1.94E-05	---	---	---	---	---	---	---
2. (2hr)	8.40E-02	7.73E-05	6.17E-11	6.52E-09	3.37E-07	8.16E-03	8.16E-03	6.36E-09	8.20E+00
3. (7hr)	2.44E-01	2.59E-05	1.09E-11	1.15E-09	5.91E-08	2.74E-03	1.09E-02	1.44E-09	8.84E+00
4. (1d)	9.39E-01	3.50E-05	3.39E-12	3.57E-10	1.85E-08	3.70E-03	1.46E-02	4.88E-10	9.31E+00
5. (2d)	1.93E+00	N/D	N/D	N/D	N/D	N/D	1.46E-02	N/D	N/D
6. (3d)	2.92E+00	N/D	N/D	N/D	N/D	N/D	1.46E-02	N/D	N/D
7. (4d)	3.91E+00	1.09E-05	7.41E-13	7.83E-11	4.04E-09	1.16E-03	1.58E-02	1.48E-10	9.83E+00
8. (5d)	4.90E+00	6.91E-06	4.69E-13	4.95E-11	2.55E-09	7.29E-04	1.65E-02	7.67E-11	1.01E+01
Mean:			1.54E-11	1.63E-09	8.41E-08			1.70E-09	9.26E+00
Standard Deviation:			2.35E-11	2.48E-09	1.28E-07			2.38E-09	6.88E-01

FILE NAME: Tc-99
SAMPLE ID: Peach Bottom #8

LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{Ci} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	1.54E-03	---	---	---	---	---	---	---
2. (2hr)	8.40E-02	2.53E-03	2.02E-09	4.39E-10	1.04E-05	5.42E-04	5.42E-04	2.81E-11	1.06E+01
3. (7hr)	2.44E-01	1.95E-03	8.15E-10	1.74E-10	4.17E-06	4.17E-04	9.59E-04	3.33E-11	1.05E+01
4. (1d)	9.39E-01	1.01E-01	9.80E-09	2.10E-09	5.02E-05	2.17E-02	2.26E-02	1.68E-09	7.78E+00
5. (2d)	1.93E+00	2.65E-03	1.80E-10	3.84E-11	9.20E-07	5.68E-04	2.32E-02	1.46E-11	1.08E+01
6. (3d)	2.92E+00	2.74E-03	1.86E-10	3.97E-11	9.51E-07	5.86E-04	2.38E-02	2.70E-11	1.06E+01
7. (4d)	3.91E+00	1.26E-03	8.53E-11	1.83E-11	4.37E-07	2.70E-04	2.41E-02	8.08E-12	1.11E+01
8. (5d)	4.90E+00	6.48E-03	4.39E-10	9.38E-11	2.25E-06	1.38E-03	2.55E-02	2.76E-10	9.56E+00
Mean:			1.93E-09	4.13E-10	9.89E-06			2.45E-09	1.01E+01
Standard Deviation:			3.27E-09	7.00E-10	1.67E-05			5.84E-09	1.05E+00

FILE NAME: Sb-125
SAMPLE ID: Peach Bottom #8

LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{Ci} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	2.32E-04	---	---	---	---	---	---	---
2. (2hr)	8.40E-02	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
3. (7hr)	2.44E-01	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
4. (1d)	9.39E-01	9.72E-04	9.39E-11	7.91E-11	5.12E-07	8.19E-04	8.19E-04	2.39E-11	1.06E+01
5. (2d)	1.93E+00	1.09E-03	7.38E-11	6.22E-11	4.02E-07	9.19E-04	1.74E-03	3.83E-11	1.04E+01
6. (3d)	2.92E+00	6.49E-04	4.40E-11	3.70E-11	2.40E-07	5.47E-04	2.28E-03	2.35E-11	1.06E+01
7. (4d)	3.91E+00	7.22E-04	4.89E-11	4.12E-11	2.66E-07	6.09E-04	2.89E-03	4.11E-11	1.04E+01
8. (5d)	4.90E+00	9.69E-04	6.58E-11	5.54E-11	3.58E-07	8.17E-04	3.71E-03	9.62E-11	1.00E+01
Mean:			6.53E-11	5.80E-11				4.46E-11	1.04E+01
Standard Deviation:			1.80E-11	1.51E-11				2.68E-11	2.23E-01

FILE NAME: I-129
SAMPLE ID: Peach Bottom #8

LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{Ci} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	2.74E-05	---	---	---	---	---	---	---
2. (2hr)	8.40E-02	5.86E-06	4.68E-12	7.79E-10	2.55E-08	9.75E-04	9.75E-04	9.09E-11	1.00E+01
3. (7hr)	2.44E-01	1.47E-05	6.13E-12	1.02E-09	3.34E-08	2.44E-03	3.41E-03	1.14E-09	8.94E+00
4. (1d)	9.39E-01	N/D	N/D	N/D	N/D	3.41E-03	3.41E-03	N/D	N/D
5. (2d)	1.93E+00	4.15E-06	2.81E-12	4.68E-10	1.53E-08	8.91E-03	1.03E-02	2.17E-09	8.66E+00
6. (3d)	2.92E+00	3.57E-05	2.42E-12	4.02E-10	1.32E-08	5.94E-03	1.63E-02	2.77E-09	8.56E+00
7. (4d)	3.91E+00	2.09E-05	1.41E-12	2.35E-10	7.70E-09	3.47E-03	1.97E-02	1.34E-09	8.87E+00
8. (5d)	4.90E+00	4.10E-06	2.78E-12	4.85E-10	1.52E-08	6.83E-03	2.66E-02	6.72E-09	8.17E+00
Mean:			3.37E-12	5.61E-10	1.84E-08			2.37E-09	8.88E+00
Standard Deviation:			1.57E-12	2.61E-10	8.54E-09			2.12E-09	5.78E-01

Table D-2. (continued).

FILE NAME: Cs-137
 SAMPLE ID: Peach Bottom #8

LEACH ID	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE ($\mu\text{Ci} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{Ci} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	3.25E-04	---	---	---	---	---	---	---
2. (2hr)	8.40E-02	7.79E-04	6.22E-10	1.14E-09	3.39E-08	1.43E-03	1.43E-03	1.95E-10	9.71E+00
3. (7hr)	2.44E-01	8.11E-04	3.39E-10	6.22E-10	1.85E-08	1.49E-03	2.92E-03	4.24E-10	9.37E+00
4. (1d)	9.39E-01	1.46E-03	1.41E-10	2.59E-10	7.68E-07	2.68E-03	5.59E-03	2.55E-10	9.59E+00
5. (2d)	1.93E+00	1.12E-03	7.55E-11	1.39E-10	4.12E-07	2.05E-03	7.64E-03	1.90E-10	9.72E+00
6. (3d)	2.92E+00	1.11E-03	7.51E-11	1.38E-10	4.09E-07	2.04E-03	9.67E-03	3.25E-10	9.49E+00
7. (4d)	3.91E+00	2.38E-03	1.61E-10	2.95E-10	8.77E-07	4.36E-03	1.40E-02	2.11E-09	8.88E+00
8. (5d)	4.90E+00	2.38E-03	1.61E-10	2.95E-10	8.79E-07	4.36E-03	1.84E-02	2.74E-09	8.56E+00
Mean:									
Standard Deviation:									
8.92E-10 9.30E+00									
9.88E-10 4.48E-01									

FILE NAME: Fe
 SAMPLE ID: Peach Bottom #8

LEACH ID	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE ($\text{ug} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{g} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	2.03E+03	---	---	---	---	---	---	---
2. (2hr)	8.40E-02	1.10E+03	8.81E-04	2.04E-10	4.80E+00	2.55E-04	2.55E-04	6.20E-12	1.12E+01
3. (7hr)	2.44E-01	3.28E+02	1.37E-04	3.17E-11	7.47E-01	7.57E-05	3.30E-04	1.10E-12	1.20E+01
4. (1d)	9.39E-01	1.03E+02	1.00E-05	2.31E-12	5.45E-02	2.39E-05	3.54E-04	2.03E-14	1.37E+01
5. (2d)	1.93E+00	1.38E+02	9.33E-06	2.18E-12	5.09E-02	3.19E-05	3.88E-04	4.61E-14	1.33E+01
6. (3d)	2.92E+00	8.62E+01	5.84E-06	1.35E-12	3.18E-02	1.99E-05	4.08E-04	3.11E-14	1.35E+01
7. (4d)	3.91E+00	5.17E+01	3.50E-06	8.09E-13	1.91E-02	1.10E-05	4.18E-04	1.50E-14	1.38E+01
8. (5d)	4.90E+00	8.62E+01	5.85E-06	1.35E-12	3.19E-02	1.99E-05	4.38E-04	5.72E-14	1.32E+01
Mean:									
Standard Deviation:									
1.50E-04 3.47E-11 8.20E-01									
3.02E-04 6.97E-11 1.64E+00									
1.07E-12 1.30E+01									
2.13E-12 9.13E-01									

Appendix D

Table D-3. Peach Bottom sample #12 cumulative fraction releases, release rates, effective diffusivities, and leachability indexes.

FILE NAME: C-14
SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	7.80E-02	----	----	----	----	----	----	----
2. (2hr)	8.37E-02	1.48E-02	1.29E-08	1.17E-11	6.43E-05	1.34E-05	1.34E-05	1.54E-15	1.38E+01
3. (7hr)	2.92E-01	9.61E-03	3.39E-09	3.08E-12	1.89E-05	8.72E-06	2.21E-05	8.71E-15	1.41E+01
4. (1d)	1.00E+00	7.68E-03	7.98E-10	7.24E-13	3.97E-06	6.97E-06	2.91E-06	1.66E-15	1.48E+01
5. (2d)	2.00E+00	9.85E-03	7.29E-10	6.57E-13	3.80E-06	8.94E-06	3.80E-06	3.35E-15	1.45E+01
6. (3d)	3.00E+00	1.07E-02	7.84E-10	7.11E-13	3.90E-06	9.68E-06	4.77E-05	6.88E-15	1.42E+01
7. (4d)	4.00E+00	4.29E-03	3.15E-10	2.80E-13	1.57E-06	3.89E-06	5.18E-05	1.52E-15	1.48E+01
8. (5d)	5.00E+00	3.38E-03	2.48E-10	2.25E-13	1.23E-06	3.08E-06	5.47E-05	1.21E-15	1.49E+01
Mean:		2.74E-09	2.49E-12	1.36E-05			5.50E-15	1.44E+01	
Standard Deviation:		4.27E-09	3.88E-12	2.13E-05			4.83E-15	3.95E-01	

FILE NAME: Fe-55
SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	7.88E-04	----	----	----	----	----	----	----
2. (2hr)	8.37E-02	8.63E-04	7.61E-10	5.08E-11	3.79E-05	5.76E-05	5.76E-05	2.86E-13	1.25E+01
3. (7hr)	2.92E-01	5.81E-03	2.05E-09	1.37E-10	1.02E-05	3.88E-04	4.45E-04	1.71E-11	1.08E+01
4. (1d)	1.00E+00	2.44E-03	2.53E-10	1.59E-11	1.26E-05	1.63E-04	6.08E-04	8.98E-13	1.20E+01
5. (2d)	2.00E+00	2.88E-03	2.11E-10	1.41E-11	1.05E-06	1.92E-04	8.00E-04	1.55E-12	1.18E+01
6. (3d)	3.00E+00	1.26E-02	9.28E-10	6.19E-11	4.82E-06	8.43E-04	1.64E-03	5.08E-11	1.03E+01
7. (4d)	4.00E+00	2.62E-03	1.93E-10	1.29E-11	9.59E-07	1.75E-04	1.82E-03	3.07E-12	1.15E+01
8. (5d)	5.00E+00	2.58E-03	1.90E-10	1.26E-11	9.43E-07	1.72E-04	1.99E-03	3.83E-12	1.14E+01
Mean:		6.55E-10	4.37E-11	3.26E-06			1.11E-11	1.15E+01	
Standard Deviation:		6.39E-10	4.24E-11	3.16E-06			1.70E-11	7.05E-01	

FILE NAME: Co-60
SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	3.29E-02	----	----	----	----	----	----	----
2. (2hr)	8.37E-02	1.00E-01	8.85E-08	2.39E-10	4.40E-04	2.71E-04	2.71E-04	6.38E-12	1.12E+01
3. (7hr)	2.92E-01	9.11E-02	3.22E-08	8.70E-11	1.60E-04	2.47E-04	5.18E-04	6.92E-12	1.12E+01
4. (1d)	1.00E+00	1.29E-01	1.34E-08	3.62E-11	6.85E-05	3.49E-04	8.67E-04	4.13E-12	1.14E+01
5. (2d)	2.00E+00	1.11E-01	8.17E-09	2.21E-11	4.06E-05	3.01E-04	1.17E-03	3.79E-12	1.14E+01
6. (3d)	3.00E+00	8.24E-02	6.08E-09	1.64E-11	3.01E-06	2.23E-04	1.39E-03	3.54E-12	1.15E+01
7. (4d)	4.00E+00	6.41E-02	4.71E-09	1.27E-11	2.35E-06	1.74E-04	1.56E-03	3.02E-12	1.15E+01
8. (5d)	5.00E+00	5.56E-02	4.08E-09	1.10E-11	2.09E-06	1.50E-04	1.71E-03	2.91E-12	1.15E+01
Mean:		2.24E-08	6.07E-11	1.12E-04			4.38E-12	1.14E+01	
Standard Deviation:		2.84E-08	7.59E-11	1.42E-04			1.49E-12	1.38E-01	

FILE NAME: Ni-63
SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE LEACH TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (Ci*yr ⁻¹)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	5.59E-03	----	----	----	----	----	----	----
2. (2hr)	8.37E-02	7.13E-03	6.29E-09	9.71E-10	3.13E-05	1.10E-03	1.10E-03	1.05E-10	9.98E+00
3. (7hr)	2.92E-01	5.97E-03	2.11E-09	3.25E-10	1.05E-05	9.22E-04	2.02E-03	9.57E-11	1.00E+01
4. (1d)	1.00E+00	9.09E-03	9.42E-10	1.46E-10	4.69E-06	1.40E-03	3.43E-03	6.70E-11	1.02E+01
5. (2d)	2.00E+00	6.79E-03	4.99E-10	7.70E-11	2.48E-06	1.05E-03	4.47E-03	4.51E-11	1.03E+01
6. (3d)	3.00E+00	4.42E-03	3.25E-10	5.01E-11	1.62E-06	6.82E-04	5.16E-03	3.32E-11	1.05E+01
7. (4d)	4.00E+00	3.70E-03	2.72E-10	4.20E-11	1.35E-06	5.72E-04	5.73E-03	3.28E-11	1.05E+01
8. (5d)	5.00E+00	3.06E-03	2.25E-10	3.47E-11	1.12E-06	4.73E-04	6.20E-03	2.88E-11	1.05E+01
Mean:		1.52E-09	2.35E-10	7.58E-06			5.85E-11	1.03E+01	
Standard Deviation:		2.04E-09	3.15E-10	1.02E-06			2.93E-11	2.15E-01	

Table D-3. (continued).

FILE NAME: Sr-90
 SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE ACTIVITY LEACHED (days) (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹) (cm ⁻² *s ⁻¹)	RELEASE RATE (Cl*yr ⁻¹)	INCREMENTAL RELEASE (Cl*yr ⁻¹)	CUMULATIVE RELEASE (Cl*yr ⁻¹)	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	N/D	----	----	----	----	----
2. (2hr)	8.37E-02	5.22E-06	4.60E-12	4.68E-10	2.29E-06	5.54E-04	5.54E-04
3. (7hr)	2.92E-01	3.47E-06	1.22E-12	1.30E-10	6.09E-09	3.68E-04	9.22E-04
4. (1d)	1.00E+00	8.58E-07	8.89E-14	9.43E-12	4.42E-10	9.10E-06	1.01E-03
5. (2d)	2.00E+00	N/D	N/D	N/D	N/D	1.01E-03	2.81E-13
6. (3d)	3.00E+00	N/D	N/D	N/D	N/D	1.01E-03	N/D
7. (4d)	4.00E+00	3.46E-06	2.54E-13	2.70E-11	1.27E-09	3.67E-04	1.38E-03
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	1.38E-03	N/D
Mean:		1.54E-12	1.54E-10	7.68E-09		1.39E-11	1.12E+01
Standard Deviation:		1.82E-12	1.93E-10	9.05E-09		9.30E-12	7.86E-01

FILE NAME: Tc-99
 SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE ACTIVITY LEACHED (days) (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹) (cm ⁻² *s ⁻¹)	RELEASE RATE (Cl*yr ⁻¹)	INCREMENTAL RELEASE (Cl*yr ⁻¹)	CUMULATIVE RELEASE (Cl*yr ⁻¹)	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	N/D	----	----	----	----	----
2. (2hr)	8.37E-02	6.84E-05	5.98E-11	1.29E-11	2.98E-07	1.47E-05	1.47E-05
3. (7hr)	2.92E-01	7.65E-04	2.70E-10	5.80E-11	1.34E-06	1.64E-04	1.79E-04
4. (1d)	1.00E+00	3.52E-03	3.65E-10	7.85E-11	1.82E-06	7.56E-04	9.35E-04
5. (2d)	2.00E+00	5.06E-03	3.72E-10	8.00E-11	1.85E-06	1.09E-03	2.02E-03
6. (3d)	3.00E+00	2.97E-03	2.19E-10	4.70E-11	1.09E-06	6.39E-04	2.66E-03
7. (4d)	4.00E+00	1.80E-04	1.32E-11	2.84E-12	6.58E-08	3.87E-05	2.70E-03
8. (5d)	5.00E+00	1.67E-03	1.22E-10	2.63E-11	6.10E-07	3.58E-04	3.06E-03
Mean:		2.03E-10	4.36E-11	1.01E-06		1.39E-11	1.15E+01
Standard Deviation:		1.32E-10	2.84E-11	6.59E-07		1.68E-11	1.21E+00

FILE NAME: I-129
 SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE ACTIVITY LEACHED (days) (uCi)	RELEASE RATE (uCi*cm ⁻² *s ⁻¹) (cm ⁻² *s ⁻¹)	RELEASE RATE (Cl*yr ⁻¹)	INCREMENTAL RELEASE (Cl*yr ⁻¹)	CUMULATIVE RELEASE (Cl*yr ⁻¹)	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	1.06E-05	----	----	----	----	----
2. (2hr)	8.37E-02	5.44E-05	5.63E-11	9.42E-09	2.80E-07	1.08E-02	9.94E-09
3. (7hr)	2.92E-01	N/D	N/D	N/D	N/D	1.08E-02	N/D
4. (1d)	1.00E+00	5.85E-05	6.91E-12	1.16E-09	3.44E-08	1.11E-02	2.19E-02
5. (2d)	2.00E+00	9.40E-06	6.90E-13	1.16E-10	3.43E-09	1.57E-03	2.35E-02
6. (3d)	3.00E+00	5.84E-05	4.30E-12	7.19E-10	2.14E-08	9.78E-03	3.33E-02
7. (4d)	4.00E+00	N/D	N/D	N/D	N/D	3.33E-02	6.82E-09
8. (5d)	5.00E+00	1.44E-04	1.06E-11	1.78E-09	5.28E-08	2.42E-02	5.74E-08
Mean:		1.58E-11	2.64E-09	7.84E-08		1.93E-08	8.33E+00
Standard Deviation:		2.05E-11	3.44E-09	1.02E-07		2.82E-08	9.31E-01

FILE NAME: Cr
 SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE QUANTITY LEACHED (days) (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹) (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE (g*yr ⁻¹)	CUMULATIVE RELEASE (g*yr ⁻¹)	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX
1. (30s)	3.47E-04	4.56E+01	----	----	----	----	----
2. (2hr)	8.37E-02	6.81E+01	5.83E-05	2.31E-09	2.90E-01	2.62E-03	5.93E-10
3. (7hr)	2.92E-01	6.07E+01	2.14E-05	8.49E-10	1.07E-01	2.41E-03	6.60E-10
4. (1d)	1.00E+00	8.05E+01	8.36E-06	3.31E-10	4.16E-02	3.20E-03	3.47E-10
5. (2d)	2.00E+00	7.66E+01	5.63E-06	2.23E-10	2.80E-02	3.04E-03	3.87E-10
6. (3d)	3.00E+00	6.61E+01	4.86E-06	1.93E-10	2.42E-02	2.62E-03	4.89E-10
7. (4d)	4.00E+00	6.23E+01	4.58E-06	1.81E-10	2.28E-02	2.47E-03	5.11E-10
8. (5d)	5.00E+00	5.92E+01	4.34E-06	1.72E-10	2.16E-02	2.35E-03	5.15E+00
Mean:		1.54E-05	6.09E-10	7.64E-02		5.43E-10	9.28E+00
Standard Deviation:		1.84E-05	7.30E-10	9.16E-02		1.28E-10	1.10E-01

Appendix D

Table D-3. (continued).

FILE NAME: Fe SAMPLE ID: PEACH BOTTOM #12										
LEACH ID	CUMULATIVE LEACH TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE RATE	CUMULATIVE RELEASE RATE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	N/D	----	----	----	----	----	----	----	
2. (2hr)	8.37E-02	5.04E+01	4.45E-05	1.04E-11	2.21E-01	1.17E-05	1.17E-05	1.19E-14	1.39E+01	
3. (7hr)	2.92E-01	6.07E+01	2.14E-05	4.98E-12	1.07E-01	1.41E-05	2.59E-06	2.27E-14	1.36E+01	
4. (1d)	1.00E+00	7.20E+01	7.47E-06	1.74E-12	3.72E-02	1.68E-05	4.26E-06	9.55E-15	1.40E+01	
5. (2d)	2.00E+00	5.05E+01	3.71E-06	8.63E-13	1.85E-02	1.17E-05	5.44E-05	5.79E-15	1.42E+01	
6. (3d)	3.00E+00	4.17E+01	3.07E-06	7.14E-13	1.53E-02	9.72E-06	6.41E-05	6.73E-15	1.42E+01	
7. (4d)	4.00E+00	4.33E+01	3.18E-06	7.40E-13	1.58E-02	1.01E-05	7.42E-05	1.02E-14	1.40E+01	
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	N/D	7.42E-05	N/D	N/D	
Mean:			1.39E-05	3.23E-12	6.91E-02			1.11E-14	1.40E+01	
Standard Deviation:			1.51E-05	3.52E-12	7.52E-02			5.58E-15	1.91E-01	
 FILE NAME: Co SAMPLE ID: PEACH BOTTOM #12										
LEACH ID	CUMULATIVE LEACH TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE RATE	CUMULATIVE RELEASE RATE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	1.69E+00	----	----	----	----	----	----	----	
2. (2hr)	8.37E-02	1.36E+01	1.23E-05	3.83E-09	6.11E-02	4.34E-03	4.34E-03	1.63E-09	8.79E+00	
3. (7hr)	2.92E-01	1.74E+01	6.12E-06	1.91E-09	3.05E-02	5.41E-03	9.75E-03	3.33E-09	8.48E+00	
4. (1d)	1.00E+00	8.58E+00	8.89E-07	2.77E-10	4.42E-03	2.67E-03	1.24E-02	2.43E-10	9.61E+00	
5. (2d)	2.00E+00	1.57E+01	1.15E-06	3.59E-10	5.73E-03	4.88E-03	1.73E-02	1.00E-09	9.00E+00	
6. (3d)	3.00E+00	2.78E+01	2.05E-06	6.38E-10	1.02E-02	8.68E-03	2.60E-02	5.36E-09	8.27E+00	
7. (4d)	4.00E+00	2.42E+01	1.78E-06	5.55E-10	8.88E-03	7.55E-03	3.35E-02	5.72E-09	8.24E+00	
8. (5d)	5.00E+00	2.26E+01	1.66E-06	5.18E-10	8.26E-03	7.05E-03	4.06E-02	6.41E-09	8.19E+00	
Mean:			3.70E-06	1.15E-09	1.84E-02			3.38E-09	8.88E+00	
Standard Deviation:			3.86E-06	1.20E-09	1.92E-02			2.30E-09	4.81E-01	
 FILE NAME: Ni SAMPLE ID: PEACH BOTTOM #12										
LEACH ID	CUMULATIVE LEACH TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE RATE	CUMULATIVE RELEASE RATE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	N/D	----	----	----	----	----	----	----	
2. (2hr)	8.37E-02	7.63E+01	6.90E-05	4.70E-09	3.44E-01	5.33E-03	5.33E-03	2.45E-09	8.61E+00	
3. (7hr)	2.92E-01	6.94E+01	2.45E-05	1.67E-09	1.22E-01	4.72E-03	1.01E-02	2.54E-09	8.59E+00	
4. (1d)	1.00E+00	9.26E+01	9.60E-06	6.54E-10	4.78E-02	6.30E-03	1.54E-02	1.35E-09	8.87E+00	
5. (2d)	2.00E+00	6.96E+01	5.11E-06	3.48E-10	2.55E-02	4.74E-03	2.11E-02	9.42E-10	9.03E+00	
6. (3d)	3.00E+00	7.48E+01	5.50E-06	3.74E-10	2.74E-02	5.09E-03	2.82E-02	1.85E-09	8.73E+00	
7. (4d)	4.00E+00	6.40E+01	4.70E-06	3.20E-10	2.34E-02	4.36E-03	3.05E-02	1.90E-09	8.72E+00	
8. (5d)	5.00E+00	6.98E+01	5.11E-06	3.48E-10	2.54E-02	4.74E-03	3.53E-02	2.89E-09	8.54E+00	
Mean:			1.76E-05	1.20E-09	8.78E-02			1.99E-09	8.73E+00	
Standard Deviation:			2.20E-01	1.50E-09	1.09E-01			6.40E-10	1.59E-01	
 FILE NAME: Zn SAMPLE ID: PEACH BOTTOM #12										
LEACH ID	CUMULATIVE LEACH TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug*cm ⁻² *s ⁻¹)	RELEASE RATE (cm ⁻² *s ⁻¹)	RELEASE RATE (g*yr ⁻¹)	INCREMENTAL RELEASE RATE	CUMULATIVE RELEASE RATE	EFFECTIVE DIFFUSIVITY (cm ² *s ⁻¹)	LEACH INDEX	
1. (30s)	3.47E-04	2.37E+01	----	----	----	----	----	----	----	
2. (2hr)	8.37E-02	3.83E+01	3.37E-05	1.85E-09	1.68E-01	2.10E-03	2.10E-03	3.80E-10	9.42E+00	
3. (7hr)	2.92E-01	3.99E+01	1.41E-05	7.72E-10	7.01E-02	2.19E-03	4.29E-03	5.45E-10	9.26E+00	
4. (1d)	1.00E+00	4.29E+01	4.45E-06	2.44E-10	2.21E-02	2.35E-03	6.84E-03	1.88E-10	9.73E+00	
5. (2d)	2.00E+00	2.96E+01	2.17E-06	1.19E-10	1.08E-02	1.62E-03	8.26E-03	1.10E-10	9.96E+00	
6. (3d)	3.00E+00	2.26E+01	1.66E-06	9.11E-11	8.27E-03	1.24E-03	9.50E-03	1.09E-10	9.96E+00	
7. (4d)	4.00E+00	2.60E+01	1.91E-06	1.05E-10	9.49E-03	1.42E-03	1.09E-02	2.03E-10	9.69E+00	
8. (5d)	5.00E+00	2.78E+01	2.04E-06	1.12E-10	1.02E-02	1.53E-03	1.24E-02	3.00E-10	9.52E+00	
Mean:			8.58E-06	4.70E-10	4.27E-02			2.62E-10	9.65E+00	
Standard Deviation:			1.11E-05	6.07E-10	5.51E-02			1.47E-10	2.44E-01	

Table D-3. (continued).

FILE NAME: B
SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE LEACH TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE ($\mu\text{g} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{g} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	N/D	---	---	---	---	---	---	---
2. (2hr)	8.37E-02	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
3. (7hr)	2.92E-01	3.47E+01	1.22E-05	2.70E-09	6.09E-02	7.66E-03	7.66E-03	6.68E-09	8.17E+00
4. (1d)	1.00E+00	3.77E+01	3.91E-06	8.64E-10	1.95E-02	8.33E-03	1.60E-02	2.36E-09	8.63E+00
5. (2d)	2.00E+00	3.83E+01	2.81E-06	6.21E-10	1.40E-02	8.45E-03	2.44E-02	3.00E-09	8.52E+00
6. (3d)	3.00E+00	N/D	N/D	N/D	N/D	N/D	2.44E-02	N/D	N/D
7. (4d)	4.00E+00	N/D	N/D	N/D	N/D	N/D	2.44E-02	N/D	N/D
8. (5d)	5.00E+00	N/D	N/D	N/D	N/D	N/D	2.44E-02	N/D	N/D
Mean:			6.32E-06	1.40E-09	3.15E-02			4.01E-09	8.44E+00
Standard Deviation:			4.21E-06	9.29E-10	2.09E-02			1.91E-09	1.93E-01

FILE NAME: Picolinic Acid
SAMPLE ID: PEACH BOTTOM #12

LEACH ID	CUMULATIVE LEACH TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE ($\mu\text{g} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{cm}^{-2} \cdot \text{s}^{-1}$)	RELEASE RATE ($\text{g} \cdot \text{yr}^{-1}$)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ($\text{cm}^2 \cdot \text{s}^{-1}$)	LEACH INDEX
1. (30s)	3.47E-04	N/D	---	---	---	---	---	---	---
2. (2hr)	8.37E-02	2.89E+04	2.55E-02	1.89E-09	1.27E+02	2.14E-03	2.14E-03	3.97E-10	9.40E+00
3. (7hr)	2.92E-01	3.02E+04	1.06E-02	7.91E-10	5.30E+01	2.24E-03	4.39E-03	5.72E-10	9.24E+00
4. (1d)	1.00E+00	6.04E+04	6.26E-03	4.65E-10	3.12E+01	4.48E-03	8.87E-03	6.83E-10	9.17E+00
5. (2d)	2.00E+00	5.87E+04	4.17E-03	3.10E-10	2.07E+01	4.21E-03	1.31E-02	7.45E-10	9.13E+00
6. (3d)	3.00E+00	3.79E+04	2.79E-03	2.07E-10	1.39E+01	2.82E-03	1.50E-02	5.65E-10	9.25E+00
7. (4d)	4.00E+00	2.84E+04	2.08E-03	1.55E-10	1.04E+01	2.11E-03	1.80E-02	4.45E-10	9.35E+00
8. (5d)	5.00E+00	2.31E+04	1.70E-03	1.26E-10	8.45E+00	1.72E-03	1.97E-02	3.81E-10	9.42E+00
Mean:			7.59E-03	5.63E-10	3.78E+01			5.41E-10	9.28E+00
Standard Deviation:			7.84E-03	5.82E-10	3.90E+01			1.30E-10	1.05E-01

Appendix D

Table D-4. Radionuclide concentrations in leachates (decay corrected to May 19, 1993).

Analyte	Units	PB #4		PB#8		PB#12	
Prerinse (30s)							
C-14	(uCi/ml)	4.03E-06	± 3.70E-07	7.07E-06	± 1.90E-06	4.67E-05	± 4.20E-06
Fe-55	(uCi/ml)	1.01E-06	± 1.06E-08	1.11E-04	± 2.01E-06	4.65E-07	± 8.48E-09
Co-60	(uCi/ml)	6.90E-06	± 6.18E-07	7.04E-06		1.95E-05	± 1.44E-06
Ni-63	(uCi/ml)	3.80E-07	± 2.00E-08	1.49E-08	± 2.98E-09	3.31E-06	± 1.80E-07
Sr-90	(uCi/ml)	2.00E-09	± 2.00E-09	1.12E-08	± 2.93E-09	-2.00E-09	± 2.00E-09
Tc-99	(uCi/ml)	-8.60E-08	± 8.60E-09	8.92E-07	± 4.90E-08	-2.48E-08	± 2.40E-09
I-129	(uCi/ml)	<1.47E-07		1.59E-08	± 2.00E-09	6.30E-09	± 3.40E-09
Cs-137	(uCi/ml)	N/D		1.93E-07		1.00E-06	± 2.00E-07
Total alpha*	(uCi/ml)	2.00E-09	± 2.00E-09			8.00E-10	± 1.50E-09
Cr	(ug/ml)	N/D		<0.03		2.70E-07	± 4.00E-03
Fe	(ug/ml)	1.90E-02	± 4.00E-03	1.18E+00		N/D	
Co	(ug/ml)	9.00E-03	± 8.00E-03	N/D		1.00E-03	± 5.00E-03
Zn	(ug/ml)	3.00E-02	± 5.00E-03	5.00E-02		1.40E-02	± 3.00E-03
B	(ug/ml)	2.00E-02	± 5.00E-03	1.00E-02		N/D	
Ni	(ug/ml)	N/D		<0.03		N/D	
PO4	(ug/ml)	8.00E-02	± 1.00E-02	6.71E+02	as P (total)	<0.50	
SO4	(ug/ml)	8.60E-01	± 4.00E-02	<1.00		3.68E+00	± 1.80E-01
Picolinic Acid	(ug/ml)	N/D		<6.16E+01		N/D	
2 Hours							
C-14	(uCi/ml)	2.53E-05	± 2.20E-06	6.87E-05	± 3.60E-06	8.49E-06	± 7.90E-07
Fe-55	(uCi/ml)	4.61E-06	± 8.48E-08	6.75E-05	± 1.58E-06	4.96E-07	± 8.48E-09
Co-60	(uCi/ml)	6.18E-05	± 4.12E-06	9.50E-05		5.77E-05	± 4.12E-06
Ni-63	(uCi/ml)	3.47E-06	± 1.90E-07	5.45E-08	± 1.59E-08	4.10E-06	± 2.00E-07
Sr-90	(uCi/ml)	9.00E-09	± 1.40E-08	4.48E-08	± 1.18E-08	3.00E-09	± 2.00E-09
Tc-99	(uCi/ml)	3.42E-07	± 2.90E-08	1.47E-06	± 4.50E-08	3.93E-08	± 3.60E-09
I-129	(uCi/ml)	9.10E-07	± 2.40E-08	3.40E-09	± 1.80E-09	3.70E-08	± 2.10E-08
Cs-137	(uCi/ml)	N/D		4.62E-07		9.00E-07	± 2.00E-07
Total alpha*	(uCi/ml)	2.00E-09	± 2.00E-09			1.00E-09	± 2.00E-09
Cr	(ug/ml)	2.30E-02	± 4.00E-03	<0.03		3.80E-02	± 3.00E-03
Fe	(ug/ml)	N/D		6.40E-01		2.90E-02	± 4.00E-03
Co	(ug/ml)	1.20E-02	± 7.00E-03	N/D		8.00E-03	± 5.00E-03
Zn	(ug/ml)	1.90E-02	± 4.00E-03	<0.03		2.20E-02	± 4.00E-03
B	(ug/ml)	1.80E-02	± 4.00E-03	<0.01		N/D	
Ni	(ug/ml)	2.80E-02	± 5.00E-03	<0.03		4.50E-02	± 5.00E-03
PO4	(ug/ml)	6.00E-02	± 1.00E-02	<0.50	as P (total)	<0.50	
SO4	(ug/ml)	1.60E+00	± 8.00E-02	<1.00		6.20E+00	± 3.00E-01
Picolinic Acid	(ug/ml)	1.84E+01		<6.16E+01		1.66E+01	

Table D-4. (continued).

Analyte	Units	PB #4		PB#8		PB#12	
7 Hours							
C-14	(uCi/ml)	5.87E-06	± 5.30E-02	5.42E-05	± 3.37E-06	5.54E-06	± 5.20E-07
Fe-55	(uCi/ml)	1.06E-06	± 1.80E-08	3.84E-05	± 1.59E-06	3.35E-06	± 6.36E-08
Co-60	(uCi/ml)	5.36E-05	± 4.12E-06	5.46E-05		5.25E-05	± 4.12E-06
Ni-63	(uCi/ml)	3.80E-06	± 2.00E-07	4.07E-08	± 9.93E-09	3.44E-06	± 1.90E-07
Sr-90	(uCi/ml)	-2.00E-09	± 1.00E-09	1.50E-08	± 2.93E-09	2.00E-09	± 1.00E-09
Tc-99	(uCi/ml)	3.04E-07	± 2.60E-08	1.13E-06	± 4.00E-06	4.41E-07	± 3.80E-08
I-129	(uCi/ml)	<4.02E-08		8.50E-09	± 1.80E-09	<1.08E-08	
Cs-137	(uCi/ml)	N/D		4.81E-07		N/D	
Total alpha*	(uCi/ml)	1.00E-09	± 2.00E-09			8.00E-10	± 1.90E-09
Cr	(ug/ml)	2.30E-02	± 4.00E-03	<0.03		3.50E-02	± 4.00E-03
Fe	(ug/ml)	N/D		1.90E-01		3.50E-02	± 5.00E-03
Co	(ug/ml)	1.80E-02	± 9.00E-03	N/D		1.00E-02	± 7.00E-03
Zn	(ug/ml)	N/D		<0.03		2.30E-02	± 4.00E-03
B	(ug/ml)	N/D		<0.01		2.00E-02	± 4.00E-03
Ni	(ug/ml)	2.60E-02	± 5.00E-03	<0.03		4.00E-02	± 7.00E-03
PO4	(ug/ml)	<0.50		<0.50	as P (total)	<0.50	
SO4	(ug/ml)	7.00E+01	± 3.00E+00	<1.00		8.30E+00	± 4.00E-01
Picolinic Acid	(ug/ml)	1.83E+01		<6.16E+01		1.74E+01	
1 Day							
C-14	(uCi/ml)	3.04E-06	± 2.80E-07	7.46E-05	± 3.70E-06	4.48E-06	± 4.20E-07
Fe-55	(uCi/ml)	9.76E-07	± 1.70E-08	3.05E-05	± 1.77E-06	1.42E-06	± 2.12E-08
Co-60	(uCi/ml)	8.14E-05	± 6.18E-06	1.18E-04		7.52E-05	± 5.15E-06
Ni-63	(uCi/ml)	4.80E-06	± 3.00E-07	5.36E-08	± 1.39E-08	5.30E-06	± 3.00E-07
Sr-90	(uCi/ml)	-9.00E-10	± 1.00E-09	2.03E-08	± 2.93E-09	5.00E-10	± 1.00E-09
Tc-99	(uCi/ml)	7.16E-07	± 6.00E-08	5.88E-05	± 1.60E-06	2.05E-06	± 1.40E-07
I-129	(uCi/ml)	2.27E-08	± 6.30E-09	<1E-09		3.88E-08	± 9.20E-09
Cs-137	(uCi/ml)	N/D		1.26E-06		8.00E-07	± 2.00E-07
Total alpha*	(uCi/ml)	2.00E-09	± 2.00E-09			3.00E-09	± 2.00E-09
Cr	(ug/ml)	3.80E-02	± 6.00E-03	<0.03		4.70E-02	± 3.00E-03
Fe	(ug/ml)	1.60E-02	± 4.00E-03	6.00E-02		4.20E-02	± 4.00E-03
Co	(ug/ml)	1.70E-02	± 8.00E-03	N/D		5.00E-03	± 6.00E-03
Zn	(ug/ml)	1.30E-02	± 4.00E-03	<0.03		2.50E-02	± 4.00E-03
B	(ug/ml)	2.00E-02	± 5.00E-03	<0.01		2.20E-02	± 4.00E-03
Ni	(ug/ml)	3.00E-02	± 8.00E-03	<0.03		5.40E-02	± 5.00E-03
PO4	(ug/ml)	<0.50		<0.50	as P (total)	<0.50	
SO4	(ug/ml)	1.13E+01	± 6.00E-01	9.33E+00		1.06E+01	± 5.00E-01
Picolinic Acid	(ug/ml)	3.55E+01		<6.16E+01		3.52E+01	

Appendix D

Table D-4. (continued).

Analyte	Units	PB #4	PB#8		PB#12					
2 Days										
C-14	(uCi/ml)	4.24E-06	±	4.00E-07	6.86E-05	±	3.57E-06	5.66E-06	±	5.30E-07
Fe-55	(uCi/ml)	1.11E-08	±	2.12E-10	1.32E-05	±	7.95E-07	1.65E-06	±	3.18E-08
Co-60	(uCi/ml)	6.49E-05	±	5.15E-06	1.03E-04			6.39E-05	±	5.15E-06
Ni-63	(uCi/ml)	3.07E-06	±	1.70E-07	2.18E-08	±	4.97E-09	3.90E-06	±	2.00E-07
Sr-90	(uCi/ml)	2.00E-09	±	1.00E-09	<1.0E-09			-5.00E-10	±	1.50E-09
Tc-99	(uCi/ml)	3.00E-06	±	2.30E-07	1.54E-06	±	4.60E-08	2.91E-06	±	2.10E-07
I-129	(uCi/ml)	1.48E-07	±	3.10E-08	2.41E-08	±	2.60E-09	5.40E-09	±	7.50E-09
Cs-137	(uCi/ml)	N/D			8.65E-07			N/D		
Total alpha*	(uCi/ml)	1.10E-09	±	2.00E-09				1.00E-09	±	3.00E-09
Cr	(ug/ml)	3.80E-02	±	6.00E-03	<0.03			4.40E-02	±	3.00E-03
Fe	(ug/ml)	N/D			8.00E-02			2.90E-02	±	3.00E-03
Co	(ug/ml)	6.00E-03	±	6.00E-03	N/D			9.00E-03	±	6.00E-03
Zn	(ug/ml)	1.50E-02	±	4.00E-03	<0.03			1.70E-02	±	5.00E-03
B	(ug/ml)	2.00E-02	±	5.00E-03	<0.01			2.20E-02	±	4.00E-03
Ni	(ug/ml)	3.00E-02	±	5.00E-03	<0.03			4.00E-02	±	6.00E-03
PO4	(ug/ml)	<0.50			<0.50	as P (total)		<0.50		
SO4	(ug/ml)	1.04E+01	±	5.00E-01	7.97E+00			1.18E+01	±	6.00E-01
Picolinic Acid	(ug/ml)	3.34E+01			<6.16E+01			3.26E+01		
3 Days										
C-14	(uCi/ml)	3.48E-06	±	3.30E-07	4.93E-05	±	3.13E-06	6.13E-06	±	5.80E-07
Fe-55	(uCi/ml)	8.10E-08	±	1.38E-09	6.00E-06	±	5.29E-07	7.26E-06	±	1.27E-07
Co-60	(uCi/ml)	4.64E-05	±	3.09E-06	6.86E-05			4.74E-05	±	3.09E-06
Ni-63	(uCi/ml)	2.14E-06	±	1.20E-07	8.94E-09	±	3.97E-09	2.54E-06	±	1.40E-07
Sr-90	(uCi/ml)	-2.00E-09	±	2.00E-09	<1E-09			-5.00E-09	±	1.00E-09
Tc-99	(uCi/ml)	5.81E-07	±	5.00E-08	1.59E-06	±	5.90E-08	1.71E-06	±	1.20E-07
I-129	(uCi/ml)	<7.96E-09			2.07E-08	±	3.80E-09	3.36E-08	±	9.90E-09
Cs-137	(uCi/ml)	N/D			6.62E-07			N/D		
Total alpha*	(uCi/ml)	1.00E-09	±	3.00E-09				2.00E-09	±	2.00E-09
Cr	(ug/ml)	3.20E-02	±	3.00E-03	<0.03			3.80E-02	±	3.00E-03
Fe	(ug/ml)	1.50E-02	±	4.00E-03	5.00E-02			2.40E-02	±	4.00E-03
Co	(ug/ml)	7.00E-03	±	5.00E-03	N/D			1.60E-02	±	8.00E-03
Zn	(ug/ml)	N/D			<0.03			1.30E-02	±	4.00E-03
B	(ug/ml)	2.00E-02	±	5.00E-03	<0.01			N/D		
Ni	(ug/ml)	2.90E-02	±	5.00E-03	<0.03			4.30E-02	±	6.00E-03
PO4	(ug/ml)	<0.50			<0.50	as P (total)		<0.50		
SO4	(ug/ml)	8.10E+00	±	4.00E-01	6.69E+00			8.20E+00	±	4.00E-01
Picolinic Acid	(ug/ml)	2.09E+01			<6.16E+01			2.18E+01		

Table D-4. (continued).

Analyte	Units	PB #4		PB#8		PB#12				
4 Days										
C-14	(uCi/ml)	2.05E-05	±	1.90E-06	4.07E-05	±	1.37E-06	2.48E-06	±	2.40E-07
Fe-55	(uCi/ml)	4.17E-07	±	7.42E-09	2.86E-06	±	6.70E-07	1.52E-06	±	3.18E-08
Co-60	(uCi/ml)	3.61E-05	±	3.09E-06	6.16E-05			3.71E-05	±	3.09E-06
Ni-63	(uCi/ml)	1.51E-06	±	8.00E-08	<5E-09			2.14E-06	±	1.20E-07
Sr-90	(uCi/ml)	2.00E-09	±	1.00E-09	6.35E-09	±	3.32E-09	2.00E-09	±	1.00E-09
Tc-99	(uCi/ml)	4.79E-07	±	4.20E-08	7.31E-07	±	4.60E-08	1.04E-07	±	8.20E-08
I-129	(uCi/ml)	1.10E-08	±	1.00E-08	1.21E-08	±	2.60E-09	<1.90E-07		
Cs-137	(uCi/ml)	N/D			6.58E-07			N/D		
Total alpha*	(uCi/ml)	1.00E-09	±	2.00E-09				2.00E-09	±	2.00E-09
Cr	(ug/ml)	2.80E-02	±	4.00E-03	<0.03			3.60E-02	±	3.00E-03
Fe	(ug/ml)	1.40E-02	±	4.00E-03	3.00E-02			2.50E-02	±	4.00E-03
Co	(ug/ml)	9.00E-03	±	7.00E-03	N/D			1.40E-02	±	7.00E-03
Zn	(ug/ml)	N/D			3.00E-02			1.50E-02	±	4.00E-03
B	(ug/ml)	N/D			2.00E-01			N/D		
Ni	(ug/ml)	2.90E-02	±	5.00E-03	<0.03			3.70E-02	±	6.00E-03
PO4	(ug/ml)	<0.50			3.00E-02	as P (total)		<0.50		
SO4	(ug/ml)	6.80E+00	±	3.00E-01	8.00E+00			6.20E+00	±	3.00E-01
Picolinic Acid	(ug/ml)	1.64E+01			<6.16E+01			1.64E+01		
5 Days										
C-14	(uCi/ml)	1.30E-05	±	1.20E-06	8.53E-05	±	1.71E-06	1.94E-06	±	1.80E-07
Fe-55	(uCi/ml)	3.18E-07	±	5.30E-09	2.78E-06	±	9.74E-07	1.48E-06	±	3.18E-08
Co-60	(uCi/ml)	2.78E-05	±	2.06E-06	1.36E-04			3.19E-05	±	2.06E-06
Ni-63	(uCi/ml)	2.42E-06	±	1.30E-07	<5E-09			1.76E-06	±	9.00E-08
Sr-90	(uCi/ml)	>4.00E-09	±	2.00E-09	4.01E-09	±	1.95E-09	-2.00E-09	±	2.00E-09
Tc-99	(uCi/ml)	3.61E-07	±	3.20E-08	3.75E-06	±	1.32E-07	9.58E-07	±	6.90E-08
I-129	(uCi/ml)	5.80E-08	±	1.50E-08	2.38E-08	±	2.90E-09	8.30E-08	±	3.10E-08
Cs-137	(uCi/ml)	N/D			1.41E-06			N/D		
Total alpha*	(uCi/ml)	1.30E-09	±	1.70E-09				1.00E-09	±	2.00E-09
Cr	(ug/ml)	2.70E-02	±	4.00E-03	<0.03			3.40E-02	±	4.00E-03
Fe	(ug/ml)	N/D			5.00E-02			N/D		
Co	(ug/ml)	1.10E-02	±	6.00E-03	N/D			1.30E-02	±	7.00E-03
Zn	(ug/ml)	N/D			7.00E-02			1.60E-02	±	3.00E-03
B	(ug/ml)	N/D			6.00E-02			N/D		
Ni	(ug/ml)	2.50E-02	±	6.00E-03	<0.03			4.00E-02	±	8.00E-03
PO4	(ug/ml)	<0.50			6.00E-02	as P (total)		<0.50		
SO4	(ug/ml)	5.30E+00	±	3.00E-01	1.11E+01			5.30E+00	±	3.00E-01
Picolinic Acid	(ug/ml)	1.27E+01			<6.16E+01			1.33E+01		

* The activity of the nuclides was at or below the detection limit of about 0.004 pCi/mL. The value reported is the sum of all actinide activity, except protactinium, found in the sample.

Appendix D

Table D-5. Leach test results for James A. FitzPatrick mixed-bed resin waste-form specimen leached in deionized water.^a

Nuclide	CFR	Release rate		Average effective diffusivity (cm ² · s ⁻¹)	Leachability index
		(μ Ci · cm ⁻² · s ⁻¹)	(cm ⁻² · s ⁻¹) ^b		
⁵⁴ Mn	5.6 (-4)	1.0 ± 0.5 (-9)	6 ± 3 (-12)	3 ± 1 (-13)	13.3 ± 0.3
⁵⁵ Fe	6.1 (-3) ^c	7 ± 5 (-10)	7 ± 5 (-11)	3 ± 2 (-11)	11.6 ± 0.8
⁵⁸ Co	1.2 (-2)	9 ± 2 (-9)	5 ± 1 (-11)	2.6 ± 0.7 (-11)	10.8 ± 0.2
⁶⁰ Co	1.7 (-2)	5 ± 1 (-8)	7 ± 2 (-11)	5 ± 1 (-11)	10.5 ± 0.2
⁶³ Ni	9.2 (-2) ^c	1.3 ± 0.4 (-8)	6 ± 1 (-10)	4 ± 1 (-9)	8.9 ± 0.3
⁶⁵ Zn	2.3 (-3)	4 ± 2 (-10)	1.0 ± 0.5 (-11)	1.2 ± 0.4 (-12)	12.3 ± 0.2
⁹⁰ Sr	— ^d	— ^d	— ^d	— ^d	— ^d
¹²⁵ Sb	<7.7 (-3)	<7.9 (-11)	<5.1 (-11)	<3.6 (-11)	>11.0
¹³⁴ Cs	7.3 (-1)	8 ± 3 (-9)	6 ± 2 (-9)	3.6 ± 0.7 (-7)	6.6 ± 0.2
¹³⁷ Cs	6.4 (-1)	1.5 ± 0.5 (-8)	5 ± 2 (-9)	1.5 ± 0.5 (-7)	7.1 ± 0.2
²⁴¹ Pu	— ^e	6.7 (-12)	6.9 (-13)	2.2 (-15)	14.7
Acid or metal		(μ g/cm ² · s)			
Picolinic acid	5.4 (-1)	8 ± 3 (-3)	3 ± 1 (-9)	1.4 ± 0.8 (-7)	7.5 ± 0.3
Chromium	<6.6 (-3) ^c	<1.6 (-6)	<5.7 (-11)	<2.4 (-11)	>11.2
Iron	<3.3 (-3) ^c	<1.0 (-5)	<2.6 (-11)	<5.4 (-12)	>12.0
Cobalt	<2.5 (-2) ^c	<8.0 (-6)	<2.2 (-10)	<3.5 (-10)	>10.0
Nickel	5.0 (-2) ^c	5 ± 2 (-6)	3 ± 1 (-10)	1.1 ± 0.5 (-9)	9.4 ± 0.4

a. Results are for only the first 90 days of leaching.

b. Fraction of initial inventory released per square centimeter of specimen surface area per second.

c. Not all leachate samples were analyzed for this radionuclide or metal. CFR value was estimated by interpolating measured incremental release rates.

d. Concentrations of radionuclide in leachate samples were not determined.

e. Only the leachate corresponding to leaching interval number three was analyzed for ²⁴¹Pu. Leaching results are those determined for leaching interval number three.

Appendix E

Radionuclide, Chelating Agent, and Stable Metal Inventory in Peach Bottom-3 Liner 446828-15

Appendix E

Radionuclide, Chelating Agent, and Stable Metal Inventory in Peach Bottom-3 Liner 446828-15

Table E-1 summarizes the radionuclide, chelating agent, and stable metal inventories in Peach Bottom-3 liner #446828. All inventories are based on the measured radionuclide content in the cemented waste form with the exception of those listed below. These radionuclide concentrations were taken from the resin samples due to apparent problems with the analysis of the waste-form samples. The total mass of material in the liner was 7,180 kg, and the radionuclide concentrations have been extrapolated to this mass based on the data summarized in Table 6. The liner was a L14-170 liner with a capacity of 4.79m³.

The concentrations of radionuclides, chelating agents, and stable metals in the liner is presented in Table E-1 as Ci/liner for radionuclides or kg of stable metals or chelates. The summed radionuclide content is 56 Ci/liner. Resin results for

⁵⁴Mn, ¹²⁵Sb, ¹³⁷Cs, ²⁴²Cm, ¹⁴C, and ⁹⁹Tc were multiplied by the resin-to-total-material ratio (0.5) for inclusion in the total. The primary decontamination radionuclides present in the resins based on their measured concentration are ⁵⁴Mn, ⁶⁵Zn, ⁶⁰Co, ⁵⁵Fe, ⁶³Ni, and ¹⁴C. The summed activity of these radionuclides is 55 Ci or about 98% of the total activity. Carbon-14 makes up about 58% of the total activity. The dominant decontamination radionuclides ⁶⁰Co and ⁵⁵Fe make up about 31% and 1.9% of the total activity. In contrast, the fission products ⁹⁰Sr, ⁹⁹Tc, ¹²⁹I, and ¹³⁷Cs collectively constitute about 0.3% of the total activity. The concentrations of the transuranic isotopes are also low and sum to a total of 1.1×10^{-2} Ci (0.02% of the total activity). Greater than 87% of the transuranic activity was ²⁴¹Pu.

Appendix E

Table E-1. Radionuclide, stable metal, and chelating agent inventory in Peach Bottom-3 liner #446828-15 (L14-170 with a capacity of 4.79m³) (Decay date 10/25/89).

Radionuclide	Peach Bottom-3 liner inventory (Ci or g)
⁵⁴ Mn	1.5E-0 ± 0.2E-1
⁵⁵ Fe	1.0E 0 ± 3.9E-1
⁶⁰ Co	1.7E+1 ± 8.6E-3
⁶³ Ni	1.9E-1 ± 2.2E-2
⁶⁵ Zn	3.4E 0 ± 1.1E 0
¹²⁵ Sb	6.1E-2 ± 3.5E-3
¹³⁷ Cs	1.3E-2 ± 1.8E-3
⁹⁰ Sr	3.0E-4 ± 5.4E-5
²³⁸ Pu	5.9E-4 ± 3.7E-5
²³⁹ Pu	1.8E-4 ± 1.1E-5
²⁴¹ Pu	9.9E-3 ± 1.0E-3
²⁴¹ Am	7.2E-4 ± 5.0E-5
²⁴² Cm	2.9E-5 ± 3.3E-6
²⁴⁴ Cm	1.3E-4 ± 1.6E-4
¹⁴ C	3.1E+1 ± 3.1E-1
⁹⁹ Tc	1.3E-1 ± 3.0E-3
¹²⁹ I	1.7E-4 ± 2.5E-5
Chromium ^a	7.4E+2 ± 1.4E+1
Iron ^a	2.1E+1 ± 1.1E 0
Zinc ^a	1.1E0 ± 8.6E-1
Nickel ^a	5.2E0 ± 2.1E-1
Cobalt	1.1E-1
Boron ^b	1.3E-1
Picolinic acid ^c	3.9E-1

a. Analyses performed using inductively coupled plasma spectroscopy elemental analysis methods.

b. Analyses performed using ion chromatography.

c. Analysis performed using picolinic acid titration. Due to uncertainties in the solidified waste analysis, the loading in the liner will be used.

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(See instructions on the reverse)

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D. W. Akers, N. C. Kraft, J. W. Mandler

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Idaho National Engineering Laboratory
EG&G Idaho, Inc
P.O. Box 1625
Idaho Falls, Idaho 83415

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10. SUPPLEMENTARY NOTES

11. ABSTRACT (200 words or less)

As part of a study being performed for the Nuclear Regulatory Commission (NRC), small-scale waste-form specimens were collected during a low oxidation-state transition-metal ion (LOMI)-nitric permanganate (NP)-LOMI solidification performed in October 1989 at the Peach Bottom Atomic Power Station Unit 3. The purpose of this program was to evaluate the performance of cement-solidified decontamination waste to meet the low-level waste stability requirements defined in the NRC's "Technical Position on Waste Form," Revision 1. The samples were acquired and tested because little data have been obtained on the physical stability of actual cement-solidified decontamination ion-exchange resin waste forms and on the leachability of radionuclides and chelating agents from those waste forms. The Peach Bottom waste-form specimens were subjected to compressive strength, immersion, and leach testing in accordance with the NRC's "Technical Position on Waste Form," Revision 1.

Results of this study indicate that the specimens withstood the compression tests (>500 psi) before and after immersion testing and leaching, and that the leachability indexes for all radionuclides, including ¹⁴C, ⁹⁹Tc, and ¹²⁹I, are well above the leachability index requirement of 6.0, required by the NRC's "Technical Position on Waste Form," Revision 1.

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