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# The Leachability of Decontamination Ion-Exchange Resins Solidified in Cement at Operating Nuclear Power Plants

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Prepared by C.V. McIsaac, J.W. Mandler

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Prepared for  
U.S. Nuclear Regulatory  
Commission

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## ABSTRACT

Data are presented on the release of radionuclides, stable metals, and organic reagents from decontamination ion-exchange resin wastes solidified in Portland cement. Both solidified and unsolidified decontamination resin waste samples were collected from five commercial light water reactors following chemical decontaminations of primary coolant systems. The decontaminations were performed using the Can-Decon, AP/Citrox, Dow NS-1, and LOMI processes. Samples of unsolidified decontamination resin waste were analyzed for concentrations of 10 CFR Part 61 radionuclides, selected transition metals, and organic chelating and complexing agents. The organic acids included oxalic acid, citric acid, formic acid, picolinic acid, ethylenediaminetetraacetic acid (EDTA), and diethylenetriaminepentaacetic acid (DPTA). Seven solidified resin waste samples were leached in deionized water following the ANS 16.1 leach-test procedure. Release rates and leachability indexes of radionuclides, transition metals, and organic acids were determined.

Analytical methods are described which were used to measure organic acids in resin wastes and in leachate solutions generated during leach-testing of waste form samples. Gas-liquid and ion chromatography methods were developed which allowed detection of organic acids in the tenths-of-a-ppm range.

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## ACRONYMS

AP	alkaline permanganate
BNL	Brookhaven National Laboratory
BWR	boiling water reactor
CFR	cumulative fraction release
DTPA	diethylenetriaminepentaacetic acid
EDTA	ethylenediaminetetraacetic acid
FID	flame ionization detector
GLC	gas-liquid chromatography
HPGe	hyper-pure germanium
HPLC	high-performance liquid chromatography
IAEA	International Atomic Energy Agency
IC	ion chromatography
ICP-AES	inductively coupled plasma-atomic emission spectroscopy
INEL	Idaho National Engineering Laboratory
LOMI	Low Oxidation-State Transition-Metal Ion
LWR	light water reactor
NRC	U.S. Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PCP	process control program
PCRS	primary coolant recirculation system
PWR	pressurized water reactor
RWCS	reactor water clean-up system
VES	vinyl ester-styrene
WINCO	Westinghouse Idaho Nuclear Co.

## EXECUTIVE SUMMARY

The U.S. Nuclear Regulatory Commission (NRC) regulation 10 CFR Part 61, "Licensing Requirements for Land Disposal of Radioactive Waste," establishes a waste classification system based on the radionuclide concentrations in the wastes. The regulation requires that higher-activity wastes, which are designated Class B and C wastes, be stabilized prior to disposal at a land burial site. Lower-activity waste, designated as Class A waste, may be disposed of in separate disposal units at the disposal site without having been stabilized. However, Section 61.56 of 10 CFR Part 61 establishes minimum requirements for all classes of waste that include the following: (a) liquid waste must be solidified or packaged in sufficient absorbent material to absorb twice the volume of the liquid, and (b) solid waste containing liquid shall contain as little free-standing and noncorrosive liquid as is reasonably achievable, but in no case shall the liquid exceed 1% of the volume.

Ion-exchange resins used to process decontamination reagents at nuclear power stations following chemical decontaminations of primary coolant system (PCS) components are routinely solidified at reactor sites using Portland cement as the solidification agent. Solidification of the ion-exchange resins provides structural stability and a slower release of radionuclides. Structural stability is intended to ensure that the waste does not degrade and promote slumping, collapse, or other failure of the cover over the disposal trench and thereby lead to water infiltration. Test procedures to demonstrate stability and leachability of waste forms are described in the "Technical Position on Waste Form," prepared by the NRC Low-Level Waste Licensing Branch, which stipulates that in order to assure that the solidification process will consistently produce a product which is acceptable for disposal and will meet disposal site license conditions, a process control program (PCP) should be used. General requirements for PCPs are provided in the NRC Standard Review Plan 11.4, "Solid Waste Management Systems," and its accompanying Branch Technical Position ETSB 11-3, "Design Guidance for Solid Waste Management Systems Installed in Light-Water-Cooled Nuclear Power Plants," (NUREG-0800).

The NRC Branch Technical Position on Waste Form specifies that waste specimens be prepared based on the proposed waste streams to be solidified and based on the range of waste stream chemistries expected. It stipulates that these solidified waste specimens should have compressive strengths of at least  $3.4 \times 10^5$  Pa (50 psig) when tested in accordance with ASTM C39 and leach-

ability indexes greater than 6 when leach-tested for a minimum of 90 days in accordance with the ANS 16.1 procedure.

The major effort of the leaching study being performed at INEL is to establish a data base on the release of radionuclides, stable metals, and organic chelating and complexing agents from decontamination ion-exchange resin wastes solidified in cement at operating light water reactors (LWRs). During 1984 and 1985, unsolidified and solidified resin waste specimens were collected from five LWRs—one pressurized water reactor (PWR) and four boiling water reactors (BWRs). Primary coolant system components of the LWRs had been decontaminated using four different chemical decontamination processes. The Low Oxidation-State Transition-Metal Ion (LOMI) process was used to decontaminate a steam generator channel head at the PWR; and the Can-Decon, Dow NS-1, and AP/Citrox processes were separately used to decontaminate the primary coolant recirculation systems at the four BWRs. In each case, samples of decontamination resin waste were collected from the solidification liner both prior to and following the addition of cement and additives. Following curing, the cement-solidified waste-form samples, which were solid right-circular cylinders, were typically about 5 cm in diameter by 10 cm long.

Seven solidified resin waste specimens were leached in deionized water following the standard procedure ANS 16.1. Before the release rates of organic acids could be determined, analytical procedures were developed for measuring their concentrations in resin wastes and in leachate solutions obtained during the course of leach-testing solidified waste specimens. Organic acids included oxalic, citric, formic, and picolinic acids, ethylenediaminetetraacetic acid (EDTA), and diethylenetriaminepentaacetic acid (DTPA). Leaching results were used to determine the release rates, effective diffusivities, and leachability indexes of radionuclides identified in 10 CFR Part 61, selected stable metals, and organic acids using the methods described in ANS 16.1.

### Concentrations of Radionuclides, Stable Metals, and Organic Acids in Resin Wastes

Of the radionuclides detected in the resin wastes,

$^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{63}\text{Ni}$  were the major activities. Concentrations of these radionuclides ranged from about 0.1 to 20  $\mu\text{Ci}/\text{cm}^3$  of wet resin. Cobalt-60 made up from 24% to 59% of the total radionuclide inventories of the resins;  $^{55}\text{Fe}$  from 15% to 62%;  $^{63}\text{Ni}$  from 3% to 43%; and  $^{54}\text{Mn}$  from 4% to 8%. The concentrations of  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  were, on the other hand, relatively low, their maximum concentrations being  $5.9 \times 10^{-4}$ ,  $1.6 \times 10^{-1}$ , and  $1.3 \times 10^{-1}$   $\mu\text{Ci}/\text{cm}^3$  of wet resin, respectively. Among the transuranics,  $^{241}\text{Pu}$  exhibited the highest activity;  $^{241}\text{Pu}$  concentrations ranged from  $5 \times 10^{-3}$  to  $3.5 \times 10^{-1}$   $\mu\text{Ci}/\text{cm}^3$ . The concentrations of transuranics in the resin waste obtained from Millstone-1 were typically an order of magnitude higher than corresponding concentrations in other resin wastes which, excluding  $^{241}\text{Pu}$ , normally ranged from  $1 \times 10^{-6}$  to  $1 \times 10^{-4}$   $\mu\text{Ci}/\text{cm}^3$ . Omitting  $^{58}\text{Co}$ , which has a half-life of only 70.9 days, total radionuclide concentrations in the resins ranged from about 1.7 to 31.7  $\mu\text{Ci}/\text{cm}^3$ . Excluding the result for the mixed-bed resin obtained from Cooper Station, the average loading on the remaining five resin waste samples was  $17 \pm 4$   $\mu\text{Ci}/\text{cm}^3$ .

Iron and nickel were the major transition metals in the resin wastes, while chromium and cobalt were present at trace levels. Concentrations of iron ranged from about  $5 \times 10^2$  to  $1 \times 10^4$   $\mu\text{g}/\text{cm}^3$  wet resin, while concentrations of nickel ranged from  $7.4 \times 10^1$  to about  $2 \times 10^3$   $\mu\text{g}/\text{cm}^3$  wet resin. Maximum concentrations of chromium and cobalt were  $3.5 \times 10^2$  and  $6 \times 10^1$   $\mu\text{g}/\text{cm}^3$ , respectively.

Concentrations of organic reagents in resin wastes were measured using gas-liquid chromatography. Prior to analysis, the involatile organic acids in the resins were first converted to their volatile methyl ester forms using boron trifluoride in methanol. The ranges of concentrations of organic acids in the resins, expressed as  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin, were: oxalic acid,  $2.7 \times 10^2$  to  $1.6 \times 10^4$ ; citric acid,  $9.6 \times 10^1$  to  $2.5 \times 10^4$ ; and EDTA,  $3.7 \times 10^3$  to  $6.1 \times 10^3$ . The concentration of DTPA in the resin collected from Pilgrim Station was  $3.6 \times 10^2$   $\mu\text{g}/\text{cm}^3$ , and the concentration of picolinic acid in the resin obtained from Indian Point-3 Station was  $4.1 \times 10^3$   $\mu\text{g}/\text{cm}^3$ . The total reagent loadings of the resin wastes ranged from 0.04 wt. % for the cation resin sample obtained from Cooper Station to 4.1 wt. % for the mixed-bed resin sample obtained from Cooper Station. The reagent loadings of other resins were typically between 1 and 2 wt. %.

## Waste Form Stability During Leaching

Of the seven cement-solidified resin waste-form samples which were leach-tested in deionized water, two completely decomposed into loose granular rubble within 8 h following immersion in leachant and two others deteriorated to a lesser extent over the course of 90 days of leaching. The samples which completely decomposed were the cation and mixed-bed resin waste forms collected from Cooper Station. The primary coolant recirculation system at Cooper Station had been decontaminated using the AP/Citrox process, which employs alkaline permanganate, citric acid, and oxalic acid. The two samples which suffered modest deterioration lost not more than 5% of their masses during leaching, but the leaching results indicate that both spallation and cracking occurred. These two samples were obtained from Millstone-1 Station, which had been decontaminated using the Can-Decon process. This decontamination process employs citric acid, oxalic acid, and EDTA.

The observed deterioration of the Millstone-1 waste forms might be the result of the condition of the cement/resin paste at the time of their collection. The cement/resin mixture had begun to set up when the waste form samples were being collected; and, as a result, following curing the surfaces of the two waste forms were coarser than usual and their upper ends were concave and irregular.

The complete decomposition of the Cooper Citrox waste forms following immersion in deionized water might have been caused by different failure mechanisms. The Cooper mixed-bed resin contained relatively high concentrations of citric acid (i.e.,  $2.5 \times 10^4$   $\mu\text{g}/\text{cm}^3$ ). The citric acid might have combined with calcium in the cement to form insoluble calcium citrate precipitates. The formation of calcium citrate would have interfered with the hydration of the cement, producing a waste form lacking structural stability. The failure of the Cooper cation resin waste form, which contained relatively small quantities of reagents, may have been due to the swelling of cation resin beads as a result of ion-exchange processes.

## Release of Radionuclides

The leaching results show that, with the exception of  $^{137}\text{Cs}$ , only very small fractions of the inventories of the radionuclides contained in the decontamination resin waste forms were released from the forms during leach-

testing. The total quantity of  $^{54}\text{Mn}$  released from any waste form ranged from less than 0.005% to less than 0.2%, where the results are expressed as percent of initial inventory. Excluding the results for the Dow NS-1 waste form obtained from Pilgrim Station, release fractions for  $^{60}\text{Co}$  ranged from 0.03% to 0.2%. The fraction of  $^{60}\text{Co}$  released from the Pilgrim sample was much higher than usual, being about 5%. Similar results for  $^{125}\text{Sb}$  ranged from 0.2% to less than 7%.

For all waste-form samples except the Dow NS-1 sample obtained from Pilgrim, the total quantities of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ , and  $^{125}\text{Sb}$  released can be accounted for if only the first 30  $\mu\text{m}$  of the surface layer of each sample was totally depleted of activity. Even in the case of the Pilgrim sample, which released about 5% of its inventory of  $^{60}\text{Co}$ , a surface totally depleted of  $^{60}\text{Co}$  to a depth of 0.5 mm would account for the total quantity of  $^{60}\text{Co}$  released from the waste form. These results indicate that it is possible that the observed releases of radionuclides other than  $^{137}\text{Cs}$  were the result of release mechanisms operating exclusively on or very near the surfaces of the waste forms. The operative release mechanisms might have included corrosion and dissolution of the waste-form surfaces and diffusion from just beneath the surfaces. The sharp declines in release rates that were observed over the course of leaching are consistent with significant depletion of activities from the surfaces of the waste forms.

While releases of most radionuclides were generally very small, five of the seven resin waste forms released their entire inventories of  $^{137}\text{Cs}$  during leaching. The waste forms which were the exceptions released about 36% and 0.4% of their initial inventories of  $^{137}\text{Cs}$ . The waste form which released an unusually small fraction of its  $^{137}\text{Cs}$  inventory was the Dow NS-1 sample obtained from Pilgrim. It is not known if special additives were used during the solidification of the Pilgrim sample, but the anomalously low  $^{137}\text{Cs}$  release suggests that additives such as zeolites, clay, or silica were employed to reduce the leachability of  $^{137}\text{Cs}$ . The results show that while significantly less  $^{137}\text{Cs}$  was released from the Pilgrim sample compared to the quantities released from other waste-form samples, the releases of transition metal radionuclides were much higher than usual. The high  $^{137}\text{Cs}$  release fractions measured for most of the waste forms indicates that  $^{137}\text{Cs}$  remained soluble following cement hydration.

Using the methods specified in ANS 16.1, leachability indexes of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ ,  $^{137}\text{Cs}$ , and  $^{241}\text{Pu}$  were determined for each of the seven waste-form specimens which were leach-tested. With the exception of the results for  $^{58}\text{Co}$  for the waste-form specimen obtained from Pilgrim Station and the results for  $^{125}\text{Sb}$  for the cation resin

waste-form specimen obtained from Cooper Station, the measured leachability indexes were all greater than 6, which, as was previously mentioned, is the lower limit considered acceptable according to the NRC Branch Technical Position on Waste Form. Leachability indexes of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{125}\text{Sb}$ , and  $^{241}\text{Pu}$  were typically greater than 10. Similar results for  $^{90}\text{Sr}$  ranged from 8.0 to 11.8, while leachability indexes of  $^{137}\text{Cs}$  varied from 6.1 to 13.4. Radionuclide leachability indexes determined for the cation and mixed-bed resin waste-form specimens obtained from Cooper Station, which both completely decomposed during initial phases of leaching, were similar to corresponding leachability indexes measured for the specimens which maintained their physical integrity during leaching.

A previous study conducted at the Idaho National Engineering Laboratory (INEL) determined the releases of  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$  from boric acid and sulfate evaporator wastes which had been solidified in cement at two operating LWRs. The average leachability indexes of  $^{60}\text{Co}$  and  $^{90}\text{Sr}$  for the majority of the decontamination resin waste forms were consistently higher than the corresponding leachability indexes determined for the boric acid and sulfate waste forms. The leachability indexes of  $^{137}\text{Cs}$  for the majority of the decontamination resin waste forms were comparable to the  $^{137}\text{Cs}$  leachability indexes determined for the evaporator waste forms. Assuming that the evaporator wastes were free of organic reagents, the results of this study indicate that the organic reagents contained in the solidified decontamination resin waste forms did not adversely affect their leaching characteristics.

## Release of Organic Acids

The concentrations of organic acids in leachates were measured using gas-liquid and ion chromatography analysis methods. The leaching results show that the total quantities of oxalic acid, citric acid, and EDTA which were released from the Can-Decon and Citrox waste forms during leaching were all quite small. Cumulative release fractions of organic acids for these waste forms ranged from 0.8% to 2.4% of initial inventories. However, organic acids were readily released from the Dow NS-1 waste form obtained from Pilgrim Station and from the LOMI waste form collected from Indian Point-3 Station. Results for the Pilgrim waste form indicated that between 30% and 40% of the original inventories of oxalic acid, citric acid, and DTPA in the sample was released during leaching. The results also indicated that the entire inventory of

EDTA was leached from the Pilgrim waste form during 48 days of continuous leaching. In the case of the LOMI waste form obtained from Indian Point-3, 83% of the formic acid and 60% of the picolinic acid originally in the waste form were released from the sample during 46 days of leaching. Measurements indicated that the release rates of organic acids from the Pilgrim and Indian Point-3 waste forms remained comparatively high even during the later, longer leaching intervals.

The trends in the releases of organic acids were, in many instances, similar to observed trends in releases of radionuclides. Release rates of both species generally decreased significantly as elapsed leaching time increased, with decreases in release rates being more pronounced during the longer leaching intervals. In the case of the Cooper mixed bed resin Citrox waste form, which disintegrated completely within 1 h following immersion in leachant, releases of both radionuclides and organic acids decreased significantly after 5 days of leaching, indicating that surface effect release

mechanisms dominated the release of both species during the initial leaching intervals. Thereafter, the releases of both species appeared to have been diffusion controlled.

In almost all cases, considerably larger fractions of the inventories of organic acids were released compared to the fractions of the inventories of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ , and  $^{125}\text{Sb}$  which were released. Conversely, in several instances, the cumulative release fraction of  $^{137}\text{Cs}$  was much higher than the cumulative release fractions of the organic acids. The specific chemical forms of the radionuclides when released from the waste forms are not known. Because the waste forms are chemically complex media, radionuclides may exist combined with binder materials as cationic or anionic ligands; or they may be complexed with organic acids. Future work should focus on determining the specific chemical forms of the radionuclides, both inside the waste forms and in solution in leachates.

# THE LEACHABILITY OF DECONTAMINATION ION-EXCHANGE RESINS SOLIDIFIED IN CEMENT AT OPERATING NUCLEAR POWER PLANTS

## INTRODUCTION

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

Two generic processes have been developed for chemical decontamination of primary system components: concentrated processes (5 to 25 wt. % chemicals in solution) and dilute processes (1 wt. % or less chemicals in solution). Examples of chemical processes that can be used in both concentrated and dilute forms are NS-1, developed by Dow Chemical Company,<sup>a</sup> and AP/Citrox. The Can-Decon and Low Oxidation-State Transition-Metal Ion (LOMI) processes are examples of dilute reagent processes (normally less than 0.2% reagent by weight). The reagents used in these processes include chelating or complexing agents such as ethylenediaminetetraacetic acid (EDTA), diethylenetriaminepentaacetic acid (DTPA), citric acid, oxalic acid, picolinic acid, and formic acid. Chelating agents and organic acids are used in decontamination formulas because they form strong complexes with actinides, lanthanides, heavy metals, and transition metals. Generally, these chemical decontamination solutions, once used, are treated with ion-exchange resins to extract the soluble activity; and it is these resins that constitute the final waste to be processed and disposed of.

Wastes from these chemical decontamination processes contain relatively large quantities of chelates or complexing agents in addition to large inventories of

activation and corrosion products. A potential problem with decontamination wastes disposed of in shallow land burial sites is the increased solubility of organo-radionuclide complexes. During recent years, a number of studies have focused on the issue of chelate-enhanced mobilization of low-level radioactive waste. A definite association of industrially used chelating agents with increased movement of certain radionuclides through soil columns was reported in 1967.<sup>1</sup> In 1977, chelated radionuclide migration was documented at the Chalk River Nuclear Facility in Canada<sup>2</sup> and at Oak Ridge National Laboratory (ORNL) in Tennessee.<sup>3</sup> EDTA was subsequently identified as the main organic compound associated with  $^{60}\text{Co}$  and trace amounts of  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{244}\text{Cm}$  at the ORNL site.<sup>4</sup> Laboratory studies and analyses of actual water collected from the site indicated that the presence of organic chelants in the leachates was responsible for diminished shale and soil adsorption of  $^{60}\text{Co}$ .<sup>4</sup> In 1981, EDTA was again identified in radioactive waste containing  $^{238}\text{Pu}$ , this time in trench leachates from the Maxey Flats burial ground in Kentucky.<sup>5</sup>

If organo-radionuclide complexes are formed after decontamination reagents and radionuclides have leached from waste forms in a burial trench, they may migrate rapidly away from the trench because of the reduction in the sorption of these species by the geome-dia.<sup>6</sup> The actual hazard posed to the public by disposal of chelated decontamination resin wastes will depend on a number of factors. Leachability is a function of the radionuclide, resin waste chemistry, solidification agent, and 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, complexing 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 leachability of actual decontamination resin waste forms that represent commonly used decontamination processes.

The U. S. Nuclear Regulatory Commission (NRC) is concerned with the safe disposal of decontamination

a. Mention of specific products and/or manufacturers in this document implies neither endorsement or preference nor disapproval by the U.S. Government, any of its agencies, or EG&G Idaho, Inc., of the use of a specific product for any purpose.



wastes containing chelating agents and has made provisions for the disposal of chelated decontamination wastes in regulation 10 CFR Part 61, "Licensing Requirements for Land Disposal of Radioactive Waste".<sup>7</sup> Section 61.2 of the regulation defines chelating agents as amine polycarboxylic acids (e.g., EDTA, DTPA), hydroxy-carboxylic acids, and polycarboxylic acids (e.g., citric acid, carboxylic acid, and glucinic acid). Section 61.56 provides requirements on the stability of waste forms which must be met for waste forms to be acceptable for near-surface disposal. Liquid wastes must be solidified, while resin wastes may be solidified or dewatered and packaged in high-integrity containers. Low-level wastes from LWRs are generally classified as Class A or B according to 10 CFR 61 and are routinely solidified at LWR sites using hydraulic cement. Solidification provides dimensional stability and a slower release of radionuclides. Test procedures to demonstrate waste-form dimensional stability and leachability are specified in the NRC Low-Level Waste Licensing Branch "Technical Position on Waste Form,"<sup>8</sup> in which a numerical leachability index, based on an effective bulk diffusivity, is defined.

Prior to July, 1984, it was common practice to solidify ion-exchange resins with standard cement formulations, regardless of the liquid waste those resins had been used to treat. Solidification in the field could proceed after a sample of waste had been satisfactorily solidified. A recent NRC requirement on decontamination waste forms is stated in the generic NRC letter dated April 30, 1984:<sup>9</sup>

"We believe that different EDTA-based decontamination solutions will have generally different effects on solidification chemistry. Testing should be done to demonstrate stability for each decontamination solution or resin to be produced."

The results of numerous investigations reported in the literature indicate that the rate of radionuclide release from a waste form is influenced by poorly understood interactions related to the properties of the solid and the leaching system. Among the factors that are known to influence the leachability of cement-solidified waste forms are the chemical composition of the cement used, waste-to-binder ratio, 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 leachant composition, pH, volume, and residence time also influence leachability.

Table 1 summarizes some of the important aspects of fourteen leaching studies that have been performed on specimens of solidified resins or decontamination solutions.<sup>10-24</sup> More specific information regarding

the sizes of the samples, the types of resins and solidification agents used, and the types of leach tests that were employed are provided in Appendix A, Table A-1. In these studies, solidified resin specimens were subjected to leach tests following either a modified International Atomic Energy Agency (IAEA) procedure<sup>25</sup> or the ANS 16.1 leach test procedure.<sup>26</sup> Both of these methods are nonequilibrium tests in the respect that the solidified resin waste sample is completely immersed over an extended period of time in a large volume of leachate that is periodically replaced with new leachant. These tests are designed to allow interlaboratory comparison of leach test results.

Ten of the fourteen leach studies were conducted using waste forms fabricated in the laboratory using simulated resin waste. The types of resins used in these studies to make simulated resin waste included those representative of the common forms of decontamination resin wastes, namely, cation, anion, and mixed-bed ion-exchange resins. The cation resin used in the majority of the studies was Amberlite IRN-77, manufactured by Rohm and Haas. This resin has a polystyrene-divinylbenzene matrix with sulfonic acid exchange groups and is supplied in the  $H^+$  form. Two types of anion exchange were commonly used: Amberlite IRN-78 (Rohm and Haas), which is a strong-base anion exchange resin having a polystyrene-divinylbenzene matrix with a quaternary ammonium functionality ( $OH^-$  form), and Ionac A-365, manufactured by Sybron, which is a weak-base polyacrylate-based anion exchange resin in the free base and  $OH^-$  forms. Simulated mixed-bed resin waste was made by adding either IRN-78 or Ionac A-365 anion exchange resin to IRN-77 cation exchange resin. Simulated resin wastes were normally doped with  $^{137}Cs$ ,  $^{85}Sr$ , and/or  $^{60}Co$  prior to solidification.

In only two cases were studies conducted using resins loaded with decontamination reagents. In these two studies,<sup>10,11</sup> anion and mixed-bed exchange resins were loaded with chelating and complexing agents to simulate decontamination resin wastes; however, neither radioactive tracers nor transition metals were added to these simulated decontamination resin wastes. In most cases, the anion exchange resins were equilibrated with enough organic acid or chelating agent to exhaust 50% of the exchange capacity. These simulated decontamination resin wastes were solidified in either Portland I cement or vinyl ester-styrene (VES) using waste-to-binder weight ratios of about 0.6 or 2.0, respectively, where the weight of the waste includes the weight of excess water added for solidification purposes. The cement-solidified samples were allowed to cure 28 days prior to the initiation of leach-testing using the ANS 16.1 leach test procedure.

**Table 1. Summary of solidified resin waste leach studies**

Author	Reference Number	Sample Size	Leachant	Solidification Agent(s)	Resin Type
Piciulo	10	Small	Deionized water	Cement, vinyl ester-styrene	Simulated anion, mixed bed
Soo	11	Small to large	Deionized water	Cement, vinyl ester-styrene	Simulated anion, cation, mixed bed
Moriyama	12	Small	Deionized water	Cement, polyethylene	Simulated cation
Colombo	13	Small	Deionized water	Urea-formaldehyde	Simulated mixed bed
Dayal Arora	14 15	Small to full size	Deionized water	Cement	Simulated cation
Morcos	16	Small	Deionized water	Cement, bitumen	Simulated cation, mixed bed
Neilson	17	Full size	Deionized water	Cement	Plant waste, plant solid.; Na <sub>2</sub> SO <sub>4</sub> + mixed bed
Neilson	18	Small	Deionized water, simulated sea water	Cement, gypsum	Simulated cation, mixed bed
Barletta	19	Small	Deionized water	Cement	Simulated mixed bed
Swyler	20	Small	Deionized water	Cement	Simulated cation
Barletta	21	Small	Deionized water, sea water, ground water	Vinyl ester-styrene	Simulated NS-1 conc.
Arora	22	Small	Deionized water	Cement	Plant waste, lab solid.; mixed bed
Westsik	23	Small to large	Deionized water, simulated ground water	Cement, bitumen	Incin. ash, lab solid.; cation, anion
Neilson	24	Small	Deionized water, simulated sea water	Cement, vinyl ester-styrene	Plant waste, lab solid.; mixed bed

The results of these two studies indicated that the rates of release of organic acids from cement- and VES-solidified waste forms varied depending on the type of ion exchange resin used and the quantities of organic reagents they contained. It was concluded that it is difficult to generalize the leaching behavior of an organic acid in different solidification media. The rate of release of EDTA from cement samples was higher than that from VES samples, but oxalic acid showed the opposite behavior, being more readily released from the VES samples than from the cement samples. A finding common to both studies was that organic acid release may be governed by different mechanisms as the leaching time elapses.

Four of the studies summarized in Table 1 were conducted using actual plant resin wastes. The types of resin waste were: (a) boiling water reactor (BWR) sodium sulfate evaporator concentrate mixed with plant mixed bed resin;<sup>17</sup> (b) BWR resin waste;<sup>22</sup> (c) incinerator ash that prior to incineration consisted of 31 wt.% cation exchange resin, 11 wt.% anion exchange resin, and 58 wt.% general trash;<sup>23</sup> and (d) EPICOR-II prefilter mixed-bed resins.<sup>24</sup> None of these waste resins contained chelating or complexing agents, and therefore the results cannot be used to define the leaching behavior of chelated resin wastes.

To date, the few leaching studies that have been focused on assessing the stability and leachability of solidified decontamination resin wastes have been conducted using simulated decontamination resin wastes that have been solidified in the laboratory. None of the studies have investigated the leaching of radionuclides in the presence of high concentrations of chelating agents and/or stable transition metals. Application of these data to the performance of actual solidified decontamination waste in near-surface disposal sites is necessarily limited because of potential mechanistic and equilibrium influences of proprietary additives that are present in the decontamination and solidification formulas.

This program and work carried out in a related field-measurements program at the Idaho National Engineering Laboratory (INEL) (Effectiveness and Safety

Aspects of Selected Decontamination Processes,<sup>27</sup> are providing base-line data on the effectiveness of currently used decontamination processes and on the performance of the resulting waste-form products. A previous study at INEL determined the leachability indexes of radionuclides contained in boric acid and sulfate evaporator wastes solidified in cement at two operating LWRs.<sup>28</sup> Different waste-form sizes were tested to demonstrate that diffusion-based modeling techniques properly account for size effects on leaching. The current program is a continuation of that work with the focus shifted to the performance of specimens of cement-solidified decontamination resin wastes obtained from operating LWRs.

The major effort of this research is to establish a data base on the release of radionuclides, chelating and complexing agents, and stable transition metals from actual decontamination resin wastes solidified in cement at operating LWRs. Before the release rates of chelating and complexing agents could be determined, it was necessary to develop analytical procedures for measuring their concentrations in waste resins and in the leachate solutions obtained during leach testing. Methods were developed to concentrate leachate organic acids on ion-exchange columns and to analyze high-specific-activity waste resins using a gas-liquid chromatography technique.

In the following section of this report, the decontamination processes are described that were used at the five LWRs from which solidified resin waste samples were collected. Then, the cement-solidification system used at the plants sampled is described and an overview of cement-solidification chemistry is given. Next, descriptions of the leach test procedure used and associated data analysis methods are presented, together with descriptions of the gas-liquid and ion chromatography analysis methods that were developed for determining concentrations of chelating and complexing agents. This is followed by the results of the laboratory-scale leach tests of seven solidified decontamination resin waste specimens. Finally, a summary and conclusions derived from this work are given, with recommendations for future work.

## DECONTAMINATION PROCESSES

In most instances, chemical decontamination process formulas have been developed in a competitive environment; therefore, their exact chemical compositions are considered proprietary. The following descriptions of the decontamination processes used at the commercial LWRs from which solidified decontamination resin waste forms were collected were obtained from the literature and do not include information of a proprietary nature.

As was previously mentioned, 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 0.2 wt.% or less. Based on these concentrations, the quantity of reagents required for a concentrated process decontamination ranges from about 48 to 248 kg of reagent per cubic meter of primary system volume to be decontaminated. A similar estimate for a dilute process decontamination is less than 2 kg per cubic meter. 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- or single-step processes. Most of the processes were approximately 0.1 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 either high temperature and high pressure [i.e., 393 K (120°C) and  $2.4 \times 10^5$  Pa (35 psig)] or at low temperature [i.e., 333 to 363 K (60 to 90°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 added to reduce the attack on the base metal by the organic acids. Because of the higher chromium content of oxide films in reactor water cleanup systems, when decontaminating these systems an oxidizing pretreatment step is added that oxidizes chromium to a readily soluble state (i.e., +3 to +6 valence).

The reagents are slurried 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

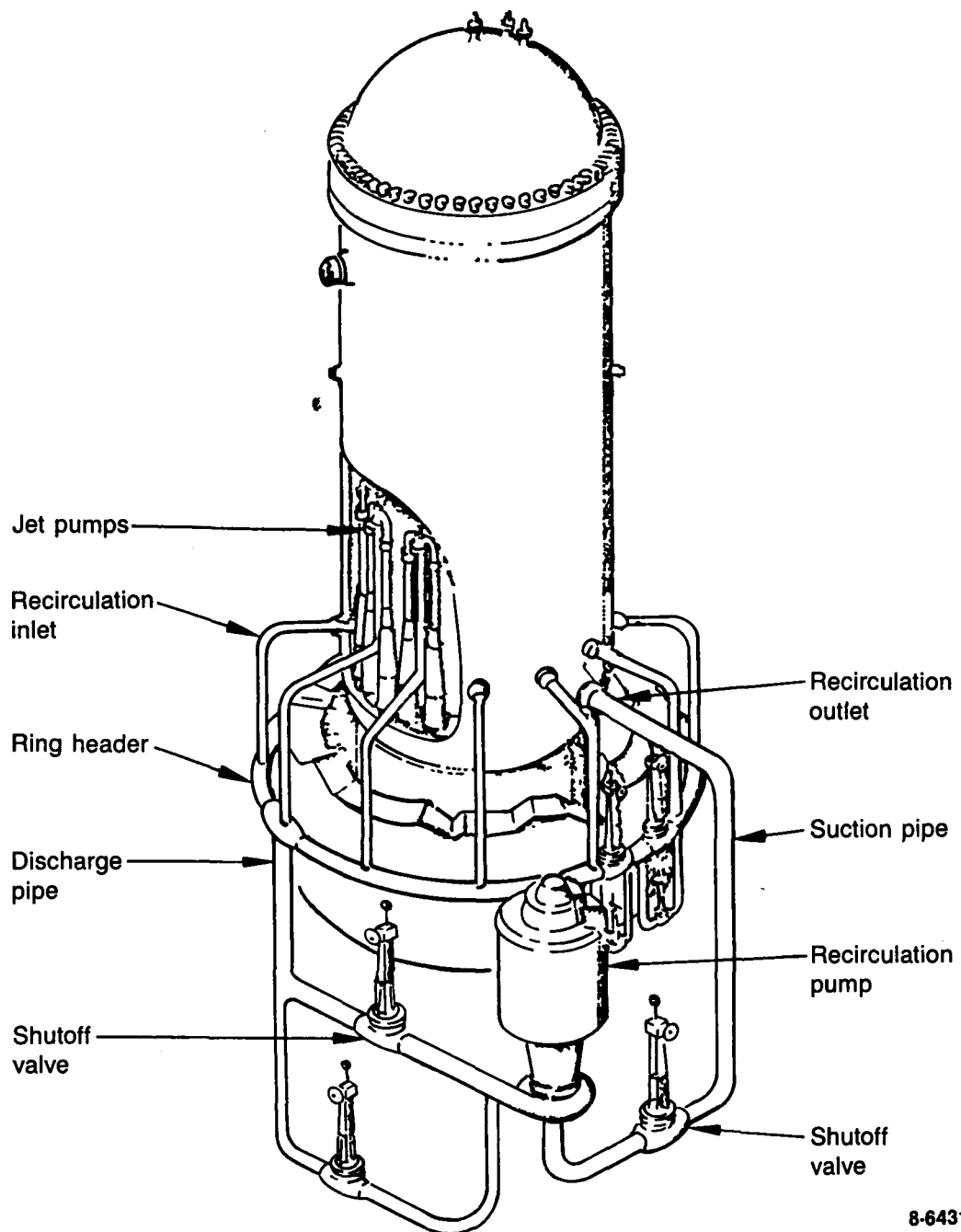
remove the corrosion and activation products and regenerate the reagents. Following completion of decontamination, mixed-bed resins are used to remove the residual metallic ions and the decontamination reagents.

During the course of this study, solidified resin waste specimens were collected from one pressurized water reactor (PWR), Indian Point-3, and four BWRs—Millstone-1, Peach Bottom-2, Pilgrim, and Cooper, which collectively had been decontaminated using four different decontamination processes. The Can-Decon, AP/Citrox, and Dow NS-1 processes were separately used to decontaminate the primary coolant recirculation systems at the four BWRs, and the LOMI process was used to decontaminate a steam generator channel head at the PWR. The dates of the decontaminations, the processes used, and the names of the companies that performed the decontaminations are summarized in Table 2. Specific details of the operational sequences used during the decontaminations are reported elsewhere.<sup>27</sup>

A schematic of a BWR reactor vessel and primary coolant recirculation system (PCRS) is shown in Figure 1. The PCRS provides the driving force for the movement of reactor coolant from the annulus area up through the reactor core region. The main components of the PCRS are the suction lines that allow water to be removed from the annulus area, the recirculation pumps, the discharge lines that carry water from the recirculation pump to the ring header, and the risers that direct the water to the jet pumps in the annulus. At a PWR, a steam generator channel head serves to direct coolant flow through the steam generator heat exchange tubes. Personnel access to perform much of the steam generator inspection, maintenance, and repair work is through the channel head manway openings.

### Can-Decon

During recent years, the Can-Decon process has been one of the most widely used decontamination processes. It was developed by Atomic Energy of Canada Limited for use in CANDU reactors and was subsequently modified for use in LWRs in the United States. Can-Decon is now marketed in the United States by London Nuclear Services Corporation, Inc., a subsidiary of London Nuclear Limited. It is a dilute chemical process that utilizes



8-6431

**Figure 1.** BWR primary coolant recirculation system.

**Table 2. Chemical decontamination summary**

Station Sampled	Date of Decon	System Deconned	Purpose	Process	Decontamination Contractor	Waste Volume (m <sup>3</sup> )
Millstone-1	04/84	PCRS	Pipe insp.	Can-Decon	London Nuclear Services Corp.	1.27
Peach Bottom-2	08/84	PCRS	Pipe repl.	Can-Decon	London Nuclear Services Corp.	1.47
Pilgrim	07/84	PCRS	Pipe insp.	Dilute NS-1	IT Nuclear Services Corp.	2.55
Cooper	11/84	PCRS	Pipe repl.	AP/Citrox	Pacific Nuclear Services	4.72
Indian Point-3	06/85	SGCH	SGCH cleanup	LOMI	Quadrex HPS	3.45

oxalic acid, citric acid, and EDTA.<sup>29</sup> Prior to the start of decontamination, the primary coolant is treated by ion exchange and chemical addition to ensure neutral pH, low conductivity, and low dissolved oxygen. Coolant temperature is maintained at about 393 K (120°C), and system pressure is kept at about  $2.4 \times 10^5$  Pa (35 psig). Once these conditions are met, a concentrated slurry of the reagent is injected into the primary coolant circulating in the system to be decontaminated.

The reagents dissolve the contaminated oxide layer by a reduction process that converts  $\text{Fe}^{+3}$  to  $\text{Fe}^{+2}$ . The dissolved corrosion and activation products are maintained in solution by the complexing agents. When used to decontaminate a BWR system, a single-step reducing solution is used; but when used on PWR systems, a dual-step process is utilized that alternates the reducing step with a dilute oxidizing step.<sup>30</sup> In order to prevent cross-contamination of the two solutions, free oxygen from the oxidizing step is scavenged with hydrazine before the reducing solution is introduced. During the time the reducing solution is used, the primary coolant is cycled through cation exchange resins to maintain reagent concentrations. The oxidizing solution is, on the other hand, not regenerated; but at the end of the process, which may cover a period of from 1 to 3 days, mixed-bed ion exchange resins are used to remove residual metallic ions and the decontamination reagents. These media—the cation and mixed-bed ion exchange resins—are the wastes that must be processed and disposed of.

## AP/Citrox

Pacific Nuclear Services, Inc. developed and licensed this proprietary process for application in the United States for both PWR and BWR systems. The AP/Citrox (alkaline permanganate)/(citric acid/oxalic acid) process is a dilute, low-temperature, low-pressure process.<sup>27,30</sup> For a PWR system, the AP/Citrox process is normally performed in the following steps: A heated slurry of AP is injected into the system being decontaminated; the primary coolant is circulated; then the coolant is drained, and the system is rinsed with reagent-free water. This step is followed by an optional dilute Citrox rinse that neutralizes any remaining AP solution. Following these initial steps, a more concentrated, heated Citrox solution is then circulated to accomplish the actual decontamination of the internal surfaces. This solution is then flushed from the system, and the system is rinsed with reagent-free water. These solutions are treated with cation and mixed-bed ion exchange resins to remove the soluble metallic ions and residual reagents.

For a BWR, system decontamination is performed as follows: a heated dilute Citrox solution is injected into the system and circulated for about 24 h. Oxalic and citric acids are then removed from the primary coolant by cycling it through mixed-bed ion exchange resins. Next, a heated alkaline permanganate solution is circulated in the system for about 8 h, after which time the primary coolant is again cycled through ion exchange resins. This chromium conditioning step is followed by a

second treatment with dilute CitroX solution. This last step is normally accomplished in 10 to 20 h. During this time, the primary coolant is cycled through cation exchange resins to remove soluble metallic ions and regenerate the reagents. The CitroX reagents and residual corrosion products are removed by mixed-bed ion exchange resins following completion of the decontamination operation.

## Dow NS-1

The Dow Chemical Company developed the NS-1 process and licensed IT Nuclear to act as the vendor of the process. The Dow NS-1 process is a high-temperature, high-pressure process that may be used in either a concentrated or dilute form.<sup>29</sup> For BWRs, it is usually a single-step process that requires about 100 h to be completed. The Dow NS-1 reagents are added to the primary coolant during several successive injections until the concentration is at the appropriate level. The primary coolant is circulated until no further increases in metallic ion concentrations are observed. After the decontamination is completed, the spent decontamination solution is drained from the system and the system is flushed with reagent-free water to remove any residual reagents. Solution reagents and soluble metallic ions are removed by passing the decontamination solution and flush water through a particulate filter and mixed-bed ion-exchange resins. A copper removal step is sometimes used at the end of the decontamination process.

## LOMI

The LOMI<sup>29,30,31</sup> process was developed by the Central Electricity Generating Board in Britain and is now being marketed in the United States by London Nuclear Technologies, Pacific Nuclear Services, and Quadrex Associates. 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 is a multi-step process that is applied differently to BWR and PWR systems. When LOMI is used to decontaminate a PWR, permanganate is injected under acidic conditions for nickel-based systems or under alkaline conditions for stainless steel systems. The permanganate solution is circulated in the system and then drained and normally destroyed using oxalic acid. Degassed picolinic acid (as sodium picolinate) is then injected into the coolant, and the coolant is circulated while being maintained at a temperature of between 353 and 363 K (80 and 90°C) under atmospheric pressure. Vanadous formate [ $V(HCOO)_2$ ] is then injected to reduce  $Fe^{+3}$  to  $Fe^{+2}$ . The picolinic acid maintains the  $Fe^{+2}$  and other corrosion products in solution. The resulting spent reagent solutions are drained from the system being decontaminated and are treated with mixed-bed ion-exchange resins. These resins constitute the decontamination waste product.

## RESIN WASTE SOLIDIFICATION

### Resin Waste Characteristics

The ion-exchange resins most frequently used to treat decontamination process solutions are organic bead resins of cation or mixed-bed form. Individual resin beads have diameters that are typically in the range of 0.35 to 1.15 mm in order to maximize effective surface area. The cation exchange resin most often encountered during the course of this study is Amberlite IRN-77, which is manufactured by Rohm and Haas Company, Philadelphia, Pa. This resin has a polystyrene-divinylbenzene matrix with sulfonic ( $\text{SO}_3^-$ ) exchange groups and is supplied in the  $\text{H}^+$  form (i.e.,  $\text{H}^+$  is the exchangeable counter ion). The mixed-bed resins generally consist of mixtures of Amberlite IRN-77 cation and either Amberlite IRN-78 anion or Ionac A-365 anion resins. IRN-78 is a strongly basic anion exchange resin having a polystyrene-divinylbenzene matrix that has a minimum of 85% of its exchange sites in the  $\text{OH}^-$  form. Ionac A-365 anion exchange resin, which is manufactured by Sybron, is a weak-base polyacrylate-based anion exchange resin that is supplied in the free base and  $\text{OH}^-$  forms. The latter resin is normally used, in mixed-bed form, to process LOMI decontamination solutions.

The minimum total exchange capacities of IRN-77 cation and INR-78 anion exchange resins are, respectively, 1.8 and 0.78 milliequivalents per cubic centimeter of wet resin. Piciulo et al.<sup>10</sup> calculated the approximate quantities of various reagents that are individually required to exhaust 50% of the exchange capacities of the previously mentioned ion-exchange resins. Extrapolating those results to 100% loading, the total exchange capacities of the IRN-77 anion exchange resin, expressed as milligrams of reagent per gram of dewatered resin, are: EDTA, 256; oxalic acid, 80; citric acid, 168; picolinic acid, 216; and formic acid, 84. Similar results for a 1:2 weight mixture of IRN-77 cation resin and IRN-78 anion resin are: EDTA, 172; oxalic acid, 52; citric acid, 112; picolinic acid, 144; and formic acid, 56. For a 1:2 weight mixture of IRN-77 cation and Ionac A-365 anion exchange resins, the total exchange capacities for picolinic and formic acids in a LOMI-type mixture are: picolinic acid, 216; and formic acid, 84, where, again, the capacities are expressed as milligrams of reagent per gram of resin. In general, the total exchange capacity weight loadings are proportional to reagent formula weight.

Decontamination ion-exchange resin wastes are normally maintained in a wet slurry condition to allow pumping them from ion-exchange columns to waste holding tanks and ultimately to the containers in which they are solidified. Ordinarily, a pumpable slurry will contain about 70% resin and 30% water by volume. Resin saturated with water to equilibrium will contain a substantial amount of water by weight. Most of this water is sorbed within the resin beads, and when resin waste is solidified in cement this water is unavailable for hydration of cement. Approximately 10 to 15 wt.% of the moisture content of dewatered resin is held in the interstitial spaces between resin beads, and it is this water that is consumed during the hydration of cement. Neilson et al.<sup>18</sup> measured the water content and densities of IRN-77 cation, IRN-78 anion, and IRN-77/IRN-78 mixed-bed resins under several different conditions, where the mixed-bed resin consisted of two parts cation to one part anion by weight. Their results are shown in Table 3.

Dewatered resin, as defined by Neilson, was resin saturated with water to equilibrium, with all free-standing water removed from above the surface of the settled resin. A resin slurry was defined as a mixture of 70 vol% settled, water-saturated resin and 30 vol% free-standing water. During a typical resin waste solidification process, resin waste is slurried into a large steel container, called a liner, that is the vessel used to perform the solidification. Prior to the addition of any solidification agents, free-standing water is pumped out of the liner until the free-standing water level is between about 2.54 and 5.08 cm above the surface of the resin beads. For an L6-80 liner, which has a usable volume of about 2.2 m<sup>3</sup>, that is filled with 1.5 m<sup>3</sup> of settled resin, 2.54 to 5.08 cm of free-standing water corresponds to only about 2.8 to 5.5 vol%. Since under normal solidification conditions only a small amount of free-standing water is left in the liner, the water contents of resin wastes are about the same as Neilson's dewatered resins.

A separate field-measurements study<sup>27</sup> conducted at INEL determined the effectiveness of chemical decontaminations performed at seven different BWRs between October 1983 and November 1984. The study collated data regarding the quantities of radionuclides and corrosion products removed from BWR primary system components as a result of chemical decontamination. Tables 4 and 5, respectively, show concentrations of activation products and stable transition



**Table 3. Densities and water contents of resins**

Resin	As Received		Dewatered		Slurry	
	Density (g/cm <sup>3</sup> )	Water (wt.%)	Density (g/cm <sup>3</sup> )	Water (wt.%)	Density (g/cm <sup>3</sup> )	Water (wt.%)
IRN-77	1.26	53.6	1.17	65.7	1.08	75.0
IRN-78	1.11	66.5	1.10	77.2	1.03	83.6
Mixed bed	1.21	57.9	1.15	72.0	1.06	79.6

**Table 4. Activation products present in decontamination resin wastes<sup>27</sup>**

System Deconned	Decon Process	Concentration in Wet Resin (μCi/cm <sup>3</sup> )					Total
		<sup>54</sup> Mn	<sup>55</sup> Fe	<sup>60</sup> Co	<sup>63</sup> Ni	Other	
PCRS	Dow NS-1	1.5 (4%) <sup>a</sup>	14.2 (39%)	17.9 (49%)	0.9 (3%)	2.0 (5%)	36.5
PCRS	Can-Decon	9.9 (23%)	— <sup>b</sup>	28.9 (67%)	— <sup>b</sup>	4.1 (10%)	42.9
RWCS	Can-Decon	— <sup>b</sup>	— <sup>b</sup>	7.1 (68%)	— <sup>b</sup>	3.3 (32%)	10.4
PCRS/RWCS	LOMI	0.2 (8%)	— <sup>b</sup>	1.5 (53%)	— <sup>b</sup>	1.1 (39%)	2.8
PCRS	Can-Decon	0.3 (1%)	— <sup>b</sup>	22.5 (70%)	— <sup>b</sup>	9.4 (29%)	32.2
PCRS	AP/Citrox	0.8 (12%)	— <sup>b</sup>	5.3 (78%)	0.05 (1%)	0.6 (9%)	6.8
PCRS	Can-Decon	— <sup>b</sup>	— <sup>b</sup>	95.2 (91%)	— <sup>b</sup>	8.9 (9%)	104.1
RWCS	Can-Decon	— <sup>b</sup>	— <sup>b</sup>	24.5 (85%)	— <sup>b</sup>	4.2 (15%)	28.7
PCRS	Can-Decon	4.8 (4%)	— <sup>b</sup>	106.4 (89%)	— <sup>b</sup>	8.3 (7%)	119.5

a. Percent of total measured activity.

b. Analysis was not performed or concentration was below detection limit.

**Table 5. Corrosion metals present in decontamination resin wastes<sup>27</sup>**

System Deconned	Decon Process	Concentration in Wet Resin ( $\mu\text{g} / \text{cm}^3$ )				
		Iron	Nickel	Zinc	Chromium	Total
PCRS	Can-Decon	8.8(+3) [77%] <sup>a</sup>	2.2(+3) [19%]	— <sup>b</sup>	5.3(+2) [5%]	1.2(+4)
PCRS/RWCS	LOMI	4.4(+2) [87%]	1.5(+1) [3%]	1.2(+1) [2%]	3.9(+1) [8%]	5.1(+2)
PCRS	Can-Decon	1.9(+4) [79%]	1.5(+3) [6%]	3.5(+2) [1%]	3.3(+3) [14%]	2.5(+4)
PCRS	Can-Decon	3.8(+3) [88%]	4.7(+2) [11%]	— <sup>b</sup>	1.7(+1) [0.4%]	4.2(+3)
RWCS	Can-Decon	2.3(+3) [84%]	2.7(+2) [10%]	— <sup>b</sup>	1.7(+2) [6%]	2.7(+3)
PCRS	Can-Decon	8.0(+3) [87%]	6.9(+2) [8%]	— <sup>b</sup>	4.9(+2) [5%]	9.1(+3)

a. Percent of total quantity of measured transition metals.

b. Analysis was not performed or concentration of metal was below detection limit.

metals in decontamination resin wastes which were calculated using data presented in Reference 27. The systems decontaminated at each plant were the PCRS and/or the reactor water clean-up system (RWCS). The decontamination processes used were either the Dow NS-1, Can-Decon, LOMI, or AP/Citrox processes. These BWRs are rated at between 536 and 1065 MWe and at the time of their decontamination had been in commercial operation between 10 and 13 years. The concentrations presented in Tables 4 and 5 were calculated using the results of analyses of reagent solutions performed by the licensees or decontamination vendors.

The results shown in Table 4 are incomplete because the concentrations of <sup>55</sup>Fe and <sup>63</sup>Ni in reagent solutions were seldom measured. Iron-55 decays by electron capture, and <sup>63</sup>Ni decays by beta emission; neither emits any gamma rays. Their analyses require the use of special radiochemical separation and measurement procedures, and for this reason they were not normally quantified. In the one case where both <sup>55</sup>Fe and <sup>63</sup>Ni concentrations were determined, <sup>60</sup>Co remained the predominant activity in the resin waste. It constituted 49% of the estimated total concentration of 36.5  $\mu\text{Ci}/\text{cm}^3$ , with <sup>55</sup>Fe, <sup>63</sup>Ni, and <sup>54</sup>Mn making up 39%, 4%, and 3%, respectively, of the total activity concentration. For the remaining resin waste

samples, <sup>60</sup>Co made up between 53% and 91% of the total activity estimated to be in the resin wastes. Other radionuclides that were occasionally quantified include <sup>51</sup>Cr, <sup>59</sup>Fe, <sup>65</sup>Zn, <sup>103</sup>Ru, and <sup>141</sup>Ce. The cumulative concentrations of these latter radionuclides ranged from 0.6 to 9.4  $\mu\text{Ci}/\text{cm}^3$  wet resin and collectively constituted between 5% and 39% of the total activities in the resin wastes. The total activity loadings of the resin wastes ranged from about 3  $\mu\text{Ci}/\text{cm}^3$  wet resin to about 120  $\mu\text{Ci}/\text{cm}^3$  wet resin and had an average value of about 43  $\mu\text{Ci}/\text{cm}^3$ .

The results presented in Table 5 show that iron constituted the major portion of the corrosion product waste generated during the decontamination of the BWRs. The concentrations of iron in the resin wastes ranged from  $4.4 \times 10^2$  to  $1.9 \times 10^4$   $\mu\text{g}/\text{cm}^3$  wet resin. On a case-by-case basis, iron made up from 77% to 88% of the total quantity of corrosion metals loaded on the resin wastes. The other significant corrosion metals were nickel, chromium, and zinc. The ranges of their concentrations in the resin wastes, expressed as  $\mu\text{g}/\text{cm}^3$  wet resin, were: nickel,  $1.5 \times 10^1$  to  $1.5 \times 10^3$ ; chromium,  $1.7 \times 10^1$  to  $3.3 \times 10^3$ ; and zinc,  $1.2 \times 10^1$  to  $3.5 \times 10^2$ . For individual decontaminations, nickel constituted from 3% to 19% and chromium from 0.4% to 14% of the total quantity of corrosion metals loaded on the resin wastes. The total

corrosion product loadings of the resin wastes ranged from  $5.1 \times 10^2$  to  $2.5 \times 10^4 \mu\text{g}/\text{cm}^3$  wet resin and had an average value of about  $9 \times 10^3 \mu\text{g}/\text{cm}^3$  wet resin.

## Cement Solidification Chemistry

The objective of solidification is to convert radioactive ion-exchange resin waste into a stable monolithic form in order to minimize the potential for radionuclide release to the environment during interim storage, transportation, and disposal. Hydraulic cement is more widely used than any other solidification agent to stabilize decontamination resin wastes. Portland cement is the primary hydraulic cement produced and is the most common one used for radioactive waste solidification. Interstitial and free-standing water in the resin waste reacts chemically with Portland cement to form hydrated silicate and aluminate compounds that interact to form a monolithic solid. Resin beads are physically entrapped and act as aggregate. The chemical and physical properties of cement-solidified decontamination resin waste forms are affected by the characteristics of the resin waste (e.g., pH, reagent loading), the waste-to-cement and water-to-cement ratios used during solidification, the type of Portland cement, the types of additives used to accelerate or inhibit cement hydration, and process variables such as temperature and cure time.

Portland cement is composed chiefly of the basic oxides of calcium, silicon, aluminum, and iron. Portland cements are not simple mixtures of these oxides but rather are mixtures of combinations of the basic oxides in the forms of tricalcium silicate, dicalcium silicate, tricalcium aluminate, and tetracalcium aluminoferrite. The formulas for these compounds and their commonly used abbreviations are shown in Table 6. All of the individual minerals present in Portland cement have different rates of hydration, strength development, and heat evolution; and, by varying the proportions of the constituents, the properties of the cement can be altered substantially. The chemical and mineralogical compositions<sup>13,32</sup> of four types of Portland cement are shown in Table 7. Type I is a general-purpose cement suitable for most uses and is the type normally used to solidify decontamination resin wastes. Type II is modified by changing the proportions of the principal oxides so as to increase the concentrations of  $\text{C}_2\text{S}$  and  $\text{C}_4\text{AF}$ . Type II has a lower heat of hydration than Type I and improved

resistance to sulfate attack. Type III has a lower  $\text{C}_2\text{S}$  content and higher  $\text{C}_3\text{S}$  content than Type I; and, as a result, it sets up faster than Type I cement. Compared to Type I, the relative concentrations of  $\text{C}_3\text{S}$  and  $\text{C}_2\text{S}$  are approximately reversed in Type IV cement. Type IV is used when the amount of heat generated during hydration must be kept to a minimum.

In general, the rate of hydration (i.e., the rate at which water chemically combines with cement) can be increased by increasing the tricalcium aluminate ( $\text{C}_3\text{A}$ ) and tricalcium silicate ( $\text{C}_3\text{S}$ ) concentrations in the cement formulation. The hydration of these compounds occurs over a relatively short period of time and liberates a substantial amount of heat. On the other hand, the heat liberated during the hydration of dicalcium silicate ( $\text{C}_2\text{S}$ ) and tetracalcium aluminoferrite ( $\text{C}_4\text{AF}$ ) is low. The primary hydration reactions of the principal compounds are shown in Table 8.

Irrespective of whether the original material is in the form of dicalcium or tricalcium silicates, the final product of cement hydration under normal conditions leads to the formation of the tricalcium disilicate hydrate ( $3 \text{CaO} \cdot 2 \text{SiO}_2 \cdot 3 \text{H}_2\text{O}$ ), which has been given the name of tobermorite. Because the two silicates differ in their lime ( $\text{CaO}$ ) contents, the reactions which lead to the formation of tobermorite are also different. The hydration of 100 parts of  $\text{C}_3\text{S}$  with 24 parts  $\text{H}_2\text{O}$ , by weight, results in the formation of about 75 parts of tobermorite and 49 parts of calcium hydroxide. This reaction is very rapid and is largely responsible for early strength. The hydration of 100 parts of  $\text{C}_2\text{S}$  with 21 parts  $\text{H}_2\text{O}$  yields about 100 parts of tobermorite and 21 parts by weight of calcium hydroxide. Hence, fully hydrated  $\text{C}_2\text{S}$  contains only about 18 wt. % calcium hydroxide. The rate of hydration of  $\text{C}_2\text{S}$  is much slower than that of  $\text{C}_3\text{S}$ , and therefore it contributes to the waste-form strength over a longer period of time. As shown in Table 8, the hydration reactions of  $\text{C}_3\text{A}$  and  $\text{C}_4\text{AF}$  consume substantially more water than the reactions of  $\text{C}_3\text{S}$  and  $\text{C}_2\text{S}$  and do not produce free calcium hydroxide.

The hydration of both tricalcium and dicalcium silicate internally produces the calcium hydroxide required for proper set. However, a problem encountered in solidifying decontamination resin wastes is that during the resin/cement mixing and early curing periods the cation ion-exchange sites rob the cement of cations (e.g.,  $\text{Ca}^{+2}$ ) and thereby interfere with the normal hydration process. It has also been demonstrated that as a result of ion-

**Table 6. Principal compounds present in Portland cements**

Compound	Abbreviation	Formula
Tricalcium silicate	C <sub>3</sub> S	3 CaO · SiO <sub>2</sub>
Dicalcium silicate	C <sub>2</sub> S	2 CaO · SiO <sub>2</sub>
Tricalcium aluminate	C <sub>3</sub> A	3 CaO · Al <sub>2</sub> O <sub>3</sub>
Tetracalcium aluminoferrite	C <sub>4</sub> AF	4 CaO · Al <sub>2</sub> O <sub>3</sub> · Fe <sub>2</sub> O <sub>3</sub>

**Table 7. Chemical and mineralogical composition of Portland cements**

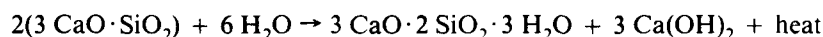
	Composition (wt. %)			
	Type I	Type II	Type III	Type IV
<u>Oxides:</u> <sup>a</sup>				
SiO <sub>2</sub>	20.67	22.44	19.83	27.51
CaO	63.03	63.81	63.41	63.73
Al <sub>2</sub> O <sub>3</sub>	6.13	3.88	5.21	1.95
Fe <sub>2</sub> O <sub>3</sub>	2.20	4.20	2.59	1.92
MgO	2.05	1.36	1.28	1.64
SO <sub>3</sub>	2.37	1.59	1.82	1.82
Balance	3.55	2.72	5.86	1.43
<u>Compounds:</u> <sup>b</sup>				
C <sub>3</sub> S	45	44	53	20
C <sub>2</sub> S	27	31	19	52
C <sub>3</sub> A	11	7	10	6
C <sub>4</sub> AF	8	13	7	14

a. D. L. Kantro and L. E. Copeland, "The Stoichiometry of the Hydration of Portland Cement," *National Bureau of Standards Monograph 43*, 1, 1962, p. 442.

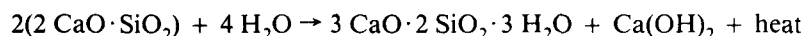
b. P. Colombo and R. M. Neilson, Jr., *Properties of Radioactive Wastes and Waste Containers*, NUREG/CR-0619, August 1979, p. 7.

**Table 8. Products of Portland cement hydration<sup>a</sup>**

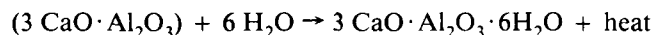
Tricalcium silicate:



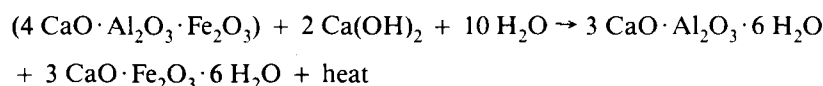
Dicalcium silicate:



Tricalcium aluminate:



Tetracalcium aluminoferrite:



a. Wolfgang Czernin, *Cement Chemistry and Physics for Civil Engineers*, London: Crosby Lockwood & Son Ltd., 1962, pp. 19-31.

exchange reactions, cation resin beads generally decrease in size prior to becoming encapsulated within the cement matrix.<sup>11</sup> When, following curing, the waste form is immersed in deionized water, wetting of the resin beads causes them to swell; and the swelling is often sufficient to crack the cement matrix. It has been shown<sup>33</sup> that volumetric changes occur when the resins are converted to ionic forms having varying degrees of hydration.

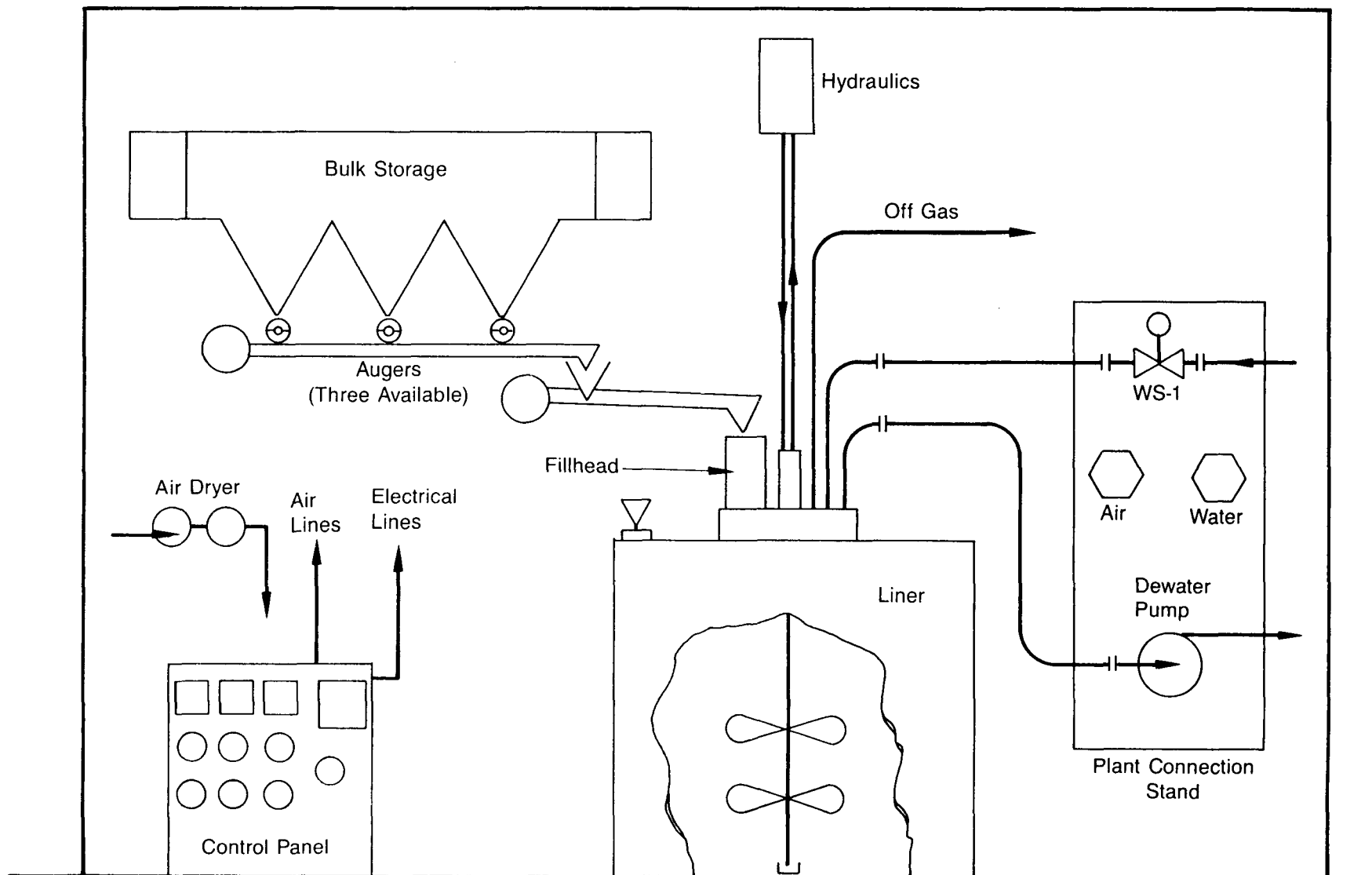
The deliberate addition of calcium hydroxide (slaked lime) to the resin waste prior to the addition of cement helps in minimizing the loss of calcium during cement hydration. The additional lime increases the pH of the waste and forms salts that have limited solubility. Sodium hydroxide is also sometimes used to increase waste pH. In addition to slaked lime and sodium hydroxide, resin wastes are sometimes treated with additives that help reduce the mobility of soluble radionuclides such as <sup>134</sup>Cs and <sup>137</sup>Cs. Additives which sorb cesium, such as zeolites and certain clays and silica, can reduce cesium leachability by factors of a hundred or more, depending upon the additive used.<sup>34</sup>

Successful solidification of resin wastes requires the proper mixture of waste, cement, and additives. The stability of a particular resin waste form is tested, qualified, and certified through a formal process control program (PCP). Each PCP describes the methodology that is used for solidifying a specific type of waste. Part of the PCP requires that, prior to solidifying waste contained in a liner, a small sample of the waste must be solidified to verify that the waste form will meet the sta-

bility requirements of the NRC Branch Technical Position.<sup>8</sup> The proportions of resin waste, cement, and additives anticipated to be used to solidify the waste are usually mixed in a beaker or other small container, and the sample is subsequently baked for about 12 h to simulate the exotherm that the resin/cement mixture in the liner experiences. If the sample sets up properly, the solidification proceeds using the tested recipe; otherwise, the proportions are changed until a satisfactory test sample is obtained.

## Chem-Nuclear Solidification System

Although several different vendors performed the decontaminations (see Table 2), all of the plants from which waste form specimens were collected contracted with Chem-Nuclear Services, Inc. to solidify the resin wastes. Solidifications were performed using Portland Type I cement. The solidification system, illustrated in Figure 2,<sup>28</sup> is portable and skid-mounted. It consists of a control panel, from which the mixing is controlled, a cement and waste distribution system, and a mixing apparatus that is installed in the solidification liner. The mixing apparatus consists of a motor, shaft, and multiple mixing blades. As shown in Figure 2, the blades are attached to the shaft at two or more levels and are oriented such that their movement creates a downward vortex in the center of the liner. The major components of the solidification system and their associated hydraulic pumps and air blowers are all mounted



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**Figure 2.** Chem-Nuclear Services, Inc. skid-mounted solidification system.

on skids which can be moved to appropriate locations and connected by hoses and electrical lines.

Solidifications were performed in either L6-80, L8-120, or L14-170 steel liners, which have usable capacities of 2.24, 3.28, or 4.79 m<sup>3</sup>, respectively. Waste resins were pumped by the plant to the Chem-Nuclear system, filling the liner to a predetermined volume. Excess free-standing water was pumped out of the liner, leaving between 2.54 and 5.08 cm of water above the surface of the settled resin beads. Slaked lime or sodium hydroxide was mixed with the resin waste prior to the introduction of Portland cement. The waste was continuously mixed while the cement was conveyed into the liner,

usually at a rate of about 900 kg/h. Mixing normally continued for about an hour after the total amount of cement had been added. When mixing was stopped, the liner fillhead was removed and the liner was vented to the plant ventilation system. The liners were usually equipped with thermocouples that monitored the hydration exotherm. The short-term hydration of tricalcium aluminate and tricalcium silicate produced peak temperatures of up to 343 K (70°C), and the observation of an exotherm peak was necessary to assure that proper solidification occurred. Solidified wastes were usually shipped to burial sites within a few days following solidification.

## EXPERIMENTAL PROCEDURES

Studies reported in the literature indicate that releases of radionuclides<sup>14,15,23,28</sup> and chelating agents<sup>11</sup> from cement-solidified waste forms closely follow predictions based on classical diffusion models. They show that for cylindrical samples the effective diffusivities derived by using the semi-infinite plane source diffusion model do not depend on sample size. In the majority of the leaching studies summarized in Table 1 (and Appendix A, Table A-1), the smallest samples tested were either approximately 5 x 5 cm or 5 x 10 cm (diameter x height). In order to allow for direct intercomparison of the results of this study with those of other studies and at the same time minimize exposure hazards and logistical requirements inherent in handling and testing radioactive samples, the majority of the waste form samples collected during this study were approximately 5 x 10 cm right-circular cylinders. The actual dimensions of the specimens collected, their volumes, surface areas, and other physical parameters are summarized in Table 9.

### Sample Collection

Unsolidified resin waste and cement-solidified resin waste specimens were collected from four BWRs and one PWR that collectively had been decontaminated using four different chemical decontamination processes. These decontaminations and subsequent sample collections took place between April 1984 and June 1985.

In each case, samples of decontamination resin waste were collected from the liner which was used to perform the solidification both prior to and following the addition of Portland cement and additives. Resin waste samples were obtained by dipping a 160-mL polyethylene wide-mouth bottle into the resin after it had been mixed a minimum of 15 min. The cement-solidified waste-form samples were collected during final mixing, or immediately upon completion of mixing. At the time of collection, the mixture was generally still a well-mixed thin paste, although occasionally the cement had hardened to the consistency of thick paste. In each case, a cylindrical polyethylene container was lowered into the liner to collect a sample of the cement/resin paste. The sample containers, which were fitted with air-tight, snap-on lids, were sealed following sample collection and were stored at least one month prior to being shipped to the INEL. Pre-

cautions were taken to assure that the samples were not subjected to temperature extremes during shipping and storage.

Millstone-1, which is a BWR rated at 654 MWe, began commercial operation in December 1970. The Can-Decon process was used to decontaminate the PCRS in April 1984. The PCRS decontamination generated 0.42 m<sup>3</sup> of mixed-bed resin waste and 0.85 m<sup>3</sup> of cation exchange resin waste. These wastes were solidified together in an L6-80 liner, which has a usable capacity of about 2.24 m<sup>3</sup>. A 160-mL sample of resin was dipped from the liner following 15 min of mixing. Two samples of cement/resin paste were obtained from the liner following the termination of mixing. Because the mixture had begun to set at the time of sampling, only two half-sized samples were collected. Both samples were allowed to cure for 684 days prior to the initiation of leach-testing. Following curing, these samples were 4.8 x 5.1 cm and 4.8 x 6.2 cm (diameter x height) rather than the nominal 5 x 10 cm. The surfaces of both samples were somewhat coarser than usual, and their upper surfaces were concave and irregular.

Peach Bottom-2 began commercial operation in July 1974. It is a BWR rated at 1065 MWe. The Can-Decon process was used to decontaminate the PCRS during August 1984. The decontamination generated 1.47 m<sup>3</sup> of cation and mixed-bed resin waste which were both slurried into the same L6-80 liner for solidification. A 160-mL sample of resin was collected from the liner after about 15 min of mixing. Two samples of cement/resin paste were obtained immediately after mixing was terminated. Following a 571-day curing period, the sample that was subjected to leach-testing was 4.8 x 8.9 cm. The surfaces of the waste-form specimen were smooth except for the upper surface which was rough and irregular.

Pilgrim, which is rated at 670 MWe, has been in commercial operation since December 1972. The PCRS was decontaminated in August 1984 using the Dow NS-1 process. A total of 2.55 m<sup>3</sup> of resin waste was produced during the decontamination; however, only 1.56 m<sup>3</sup> of this waste was solidified in the liner from which samples were obtained. Samples of resin were collected at the time the resins were transferred from a temporary holding tank to the solidification liner. A portion of the resin was used to prepare a solidified waste-form specimen, and the remainder was retained for analysis. The



**Table 9. Physical parameters of solidified decontamination resin waste samples**

Sample Identification	Date Collected	Cure Time (days)	Waste Type	Solidified Waste Form					Resin Loading		
				Diameter x Height (cm)	Surface Area (cm <sup>2</sup> )	Mass (g)	Volume (cm <sup>3</sup> )	Volume-to-Surface-Area Ratio (cm)	Wet (vol.%)	Wet (wt%)	Dry (wt%)
Millstone-1 F33	06/20/84	684	Rohm and Haas (R&H) IRN-77 and IRN-78	4.8 x 5.1	107	114.3	84.9	0.79	72	61	17
Millstone-1 F201	06/20/84	684	R&H IRN-77 & IRN-78	4.8 x 6.2	115	125.9	93.5	0.81	72	61	17
Peach Bottom-2	08/09/84	571	R&H IRN-77 & IRN-78	4.8 x 8.9	169	207.7	158.4	0.94	72	63	18
Pilgrim	08/17/84	654	NA	4.8 x 9.2	173	215.2	164.1	0.95	71	62	17
Cooper (mixed bed)	11/30/84	710	10% Purolite C-100-H, 90% Purolite A-600	4.4 x 9.5	164	205.2	147.8	0.90	72	60	17
Cooper (cation)	11/30/84	802	Purolite NRW-37	4.4 x 10.2	173	215.6	157.7	0.91	77	66	23
Indian Point-3	06/24/85	539	Ionac A-365, R&H IRN-77 + cation resin from radwaste	4.6 x 9.5	171	202.5	158.6	0.93	70	63	18

solidified waste-form specimen that was leach-tested was prepared by Chem-Nuclear Services, Inc. using a sample of the resin waste and the same mixing parameters used for the full-scale solidification.

Cooper, a BWR rated at 764 MWe, has been in commercial operation since July 1974. In November 1984, its PCRS was decontaminated using the AP/Citrox process. The decontamination generated 2.61 m<sup>3</sup> of cation exchange resin waste and 2.12 m<sup>3</sup> of mixed-bed resin waste. The cation and mixed-bed resins were solidified separately in L14-170 (4.79 m<sup>3</sup>) and L8-120 (3.28 m<sup>3</sup>) liners, respectively. A 160-mL sample of resin was dipped out of each liner after the resins had been mixed for about 30 min. One sample of cement/resin paste was removed from each liner immediately after mixing stopped. The volumes of resins solidified were determined by measurements made inside the liners, and Chem-Nuclear Services, Inc. supplied information on the masses of the cement and additives used for each liner. Following curing for 710 and 802 days, respectively, the mixed-bed and cation waste-form specimens had dimensions of 4.4 x 9.5 cm and 4.4 x 10.2 cm, respectively.

Prior to the initiation of leaching, the surfaces of the Cooper mixed-bed resin waste-form specimen were smooth and exhibited very little surface cracking. The only anomalous features the specimen exhibited were the numerous tiny chips that had broken off the circumference of one end of the waste form. The specimen appeared sound and well cured. Photographs of the Cooper cation resin waste form, taken prior to the initiation of leaching, are shown in Figure 3. A relatively large crack traversed one side of the waste form just below its mid-plane, and several small voids pitted the surface above the mid-plane. Partially encapsulated resin beads were exposed over about one-third of the surface of the top end of the waste form. Except for the features mentioned, the surfaces of the cation resin waste form were smooth and unpitted.

Indian Point-3 is a PWR rated at 965 MWe which began commercial operation in August 1976. A steam generator channel head was decontaminated during June 1985 using the LOMI process. The decontamination generated 2.27 m<sup>3</sup> of mixed-bed resin waste; but, for solidification, this waste was combined with 1.19 m<sup>3</sup> of resin waste from the plant radwaste system. A 160-mL sample of resin waste was collected after about 30 min of mixing in the liner. Samples of cement/resin paste were collected about 20 min after final mixing was terminated. Three resin waste-form specimens were

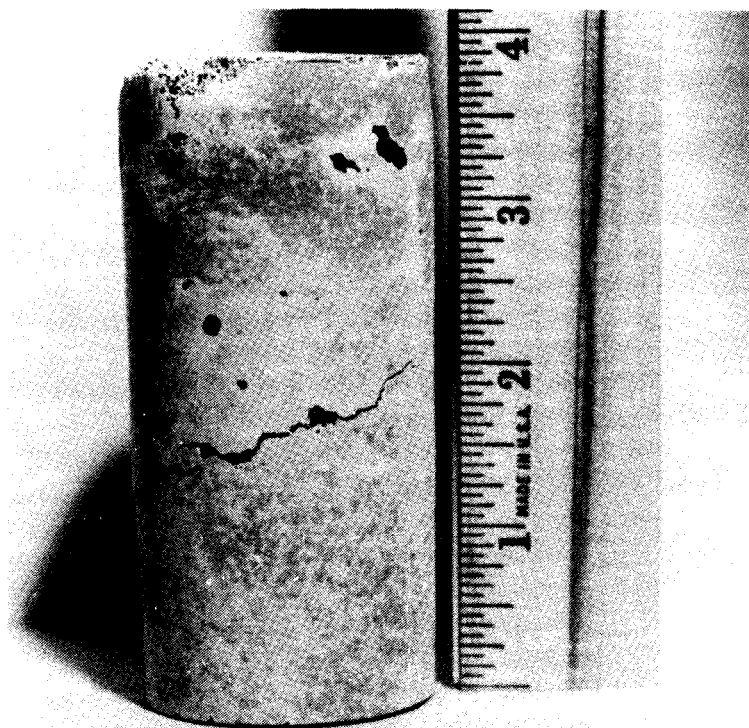
collected from the liner. Following curing for 539 days, the dimensions of the cylindrical waste form which was subjected to leach-testing were 4.6 x 9.5 cm. Photographs of the Indian Point-3 waste-form sample, taken prior to the start of leaching, are shown in Figure 4. As shown, the surface of the upper end of the specimen was not perpendicular to the specimen's longitudinal axis. Also, a region of the surface on the side of the sample having the shortest height was rough and exhibited small irregular voids. These features were both the result of the sample container being tilted during initial curing.

## Leach-Test Method

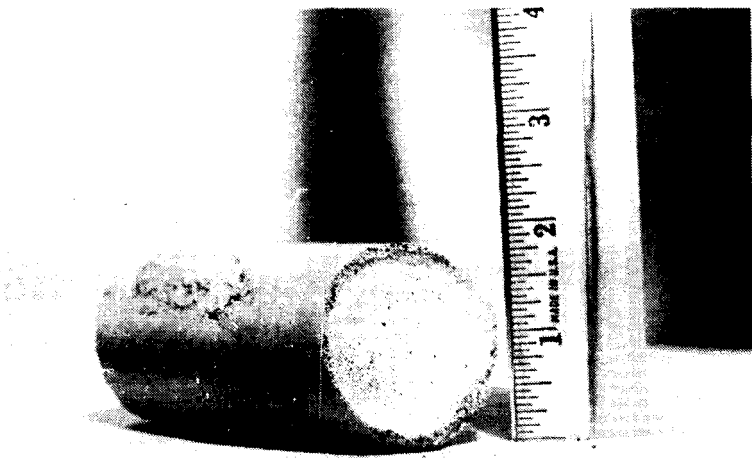
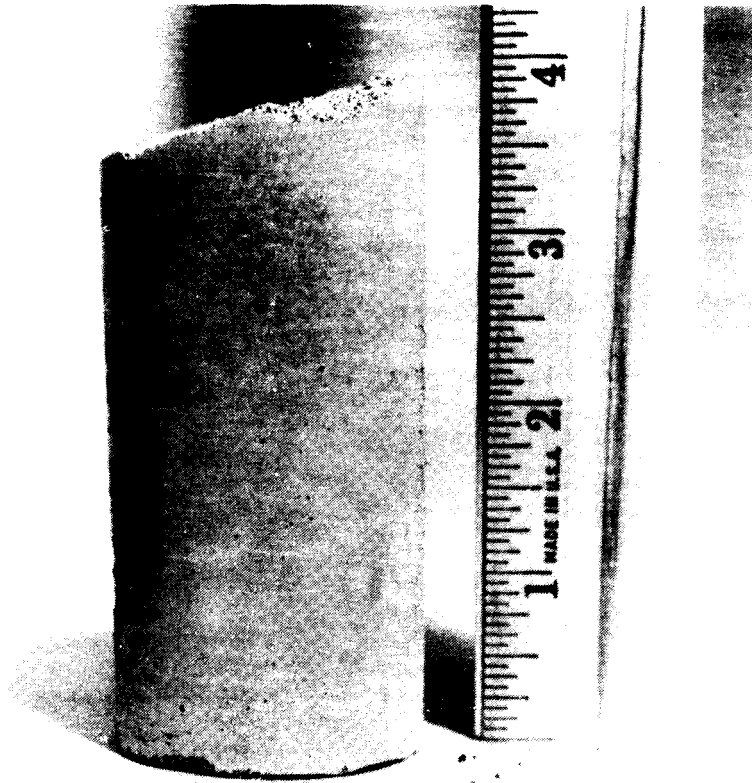
The test procedure used to measure the release of radionuclides, transition metals, and chelating agents from decontamination resin wastes solidified in Portland cement was American National Standard ANSI/ANS 16.1, "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure."<sup>26</sup> The initial version of the standard was issued during February 1984, and the final version was approved by the American National Standards Institute, Inc. in April 1986. Specific requirements of the April 1986 ANS 16.1 are provided in Appendix B.

The standard is recommended by the NRC in the Low-Level Waste Management Branch Technical Position on Waste Form<sup>8</sup> to measure the release of radionuclides from solidified low-level radioactive waste. The standard is intended to provide a means of indexing the release of radionuclides from waste forms using the results of relatively short-term tests performed in a laboratory. It is not intended to be used to define the long-term leaching behavior of waste forms under conditions representing actual burial environments. The method specified by the standard for analyzing leach-test data is based on the assumption that diffusion is the only significant 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 questioned<sup>34</sup>, the test procedure was, however, believed suitable 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 seven decontamination resin waste specimens that were leach-tested were



**Figure 3.** Cooper cation resin waste form prior to leaching.



**Figure 4.** Indian Point-3 resin waste form prior to leaching.

right-cylindrical solids. Five of the waste-form specimens had nominal dimensions of 5 x 10 cm (diameter x height), and two had nominal dimensions of 5 x 5 cm. The specimens were cured at ambient temperature between 539 and 802 days prior to the initiation of leach-testing. The cylindrical leach-test vessels were polyethylene containers having capacities of about 3 L. In each case, the waste-form sample was supported in the leach-test vessel by coarse mesh plastic netting that was suspended by a wire from the container lid. Deionized water having a conductivity of less than 3  $\mu$ mho/cm at 298 K (25°C) was used as the leachant after being allowed to come to ambient temperature. During leaching, leachant temperature was maintained at  $296 \pm 3$  K ( $23 \pm 3^\circ$ C). Leachant volumes ranged from 1070 mL for the Millstone-1 F33 specimen to 2030 mL for the Cooper cation resin waste form specimen. Values of the ratio of leachant volume to specimen external geometric surface area ranged from 7.7 for the Peach Bottom-2 sample to 11.7 for the Cooper cation resin sample. With few exceptions, leachates were changed out at intervals specified in ANS 16.1 (see Appendix B).

**Data Analysis.** Leaching occurs as the result of mass transport of species inside of as well as out of a waste form. Mass transport processes which have been identified as occurring in solidified waste forms include diffusion, dissolution, ion exchange, corrosion, and surface effects. A considerable amount of data obtained from samples which maintained their physical integrity during leaching indicate that internal bulk diffusion is the most likely rate-determining mechanism during the initial phases of the leaching process.<sup>35,36,37</sup>

The solution to the transport equation for diffusion in a homogeneous, semi-infinite medium has become the most widely used analytical expression to explain the leaching release of radionuclides and other diffusing species from solidified waste forms. This semi-infinite medium diffusion model is incorporated into ANS 16.1. One of the boundary conditions of this solution is that some portion of the waste form must retain its initial concentration of the diffusing species. Leaching of real waste forms leads to depletion, which must be accounted for in any realistic model of leaching behavior. This limitation is addressed in ANS 16.1 with the specification that the shape-specific solution of the mass transport equations be used for data analysis when depletion of a diffusant becomes greater than 20%.

For the case of a semi-infinite medium, the solution of the mass transport differential equation<sup>38</sup> is

$$\frac{\Sigma a_n}{A_0} = 2 \left( \frac{DT}{\pi} \right)^{1/2} \quad (1)$$

where

$a_n$  = quantity of species leached in interval  $n$

$A_0$  = total amount of species originally present in waste form

$\frac{\Sigma a_n}{A_0}$  = cumulative fraction leached through leach interval  $n$

$D$  = effective diffusivity ( $\text{cm}^2/\text{s}$ )

$T$  = cumulative leach time (s).

Limiting consideration to a one-dimensional, semi-infinite medium in which the diffusant is uniformly distributed prior to leaching and for which the surface concentration drops to zero upon initiation of leaching imposes certain initial and boundary conditions on Equation (1). Slight changes in this model reduce problems where initial boundary conditions are not met. In order to use Equation (1) to calculate cumulative release fractions for a real waste-form specimen, it must first be multiplied by a normalizing factor which is the ratio of the specimen's external geometric surface area to its volume. An actual sample will not have a zero surface concentration. This requires the addition of a constant term,  $\beta$ , which is the initial release from the surface. The semi-infinite medium diffusion equation then becomes

$$\frac{\Sigma a_n}{A_0} = 2 \left( \frac{S}{V} \right) \left( \frac{DT}{\pi} \right)^{1/2} + \beta \quad (2)$$

where

$S$  = external geometric surface area of specimen ( $\text{cm}^2$ )

$V$  = volume of sample ( $\text{cm}^3$ )

$\beta$  = fraction released from surface.

This equation is linear with the square root of cumulative leach time; consequently, the effective diffusivity can be calculated from Equation (2) by solving for the slope of the cumulative release fraction as a function of the square root of elapsed leach time. A disadvantage of this method of determining the effective diffusivity is that errors or bias in the data points are accumulated, affecting the calculated effective diffusivity. To eliminate this potential problem, the effective diffusivity can be calculated using measured incremental release fractions which effectively decouple data for individual leach intervals. Using this method, the equation for the effective diffusivity has the following form:

$$D_n = \pi \left( \frac{a_n/A_n}{(\Delta t)_n} \right)^2 \left( \frac{V}{S} \right)^2 T \quad (3)$$

where

$D_n$  = effective diffusivity for leach interval  $n$  ( $\text{cm}^2/\text{s}$ )

$(\Delta t)_n$  = duration of leach interval  $n$  (s)

$T = \left( \frac{t_n^2}{2} + \frac{t_{n+1}^2}{2} \right)^2$  = leaching time  
representing the "mean time" of the  $n$ th leaching interval (s)

$t_n$  = cumulative time through the  $n$ th leaching interval (s).

Equation (3) is the expression for effective diffusivity specified in ANS 16.1 when less than 20% of the diffusing species of interest has leached from the waste-form specimen. It is the equation that was used to calculate the majority of the effective diffusivities reported in this report.

If more than 20% of a diffusing species has been depleted from the waste-form specimen at the end of a leach interval, the effective diffusivity must be calculated from a shape-specific solution of the mass transport equations.<sup>37,39,40</sup> The equation for the cumulative release fraction for a finite cylinder is:

$$\frac{\sum a_n}{A_n} = 1 - \frac{32}{\pi^2} \sum_m \sum_n \frac{\exp \left[ - \left( \frac{J_{0m}}{r} \right)^2 Dt \right]}{J_{0m}^2}$$

$$\frac{\exp \left[ - \left( \frac{(2n-1)\pi}{h} \right)^2 Dt \right]}{(2n-1)^2} \quad (4)$$

where

$J_{0m}$  =  $m$ th positive root of the zeroth order Bessel function

$r$  = radius of cylinder (cm)

$h$  = height of cylinder (cm)

$t$  = elapsed leaching time (s).

The ANS 16.1 standard provides a convenient method for calculating the effective diffusivity given by Equation (4). For the case where more than 20% of a diffusant has leached from the specimen, the effective diffusivity is given by

$$D = \frac{Gd^2}{t} \quad (5)$$

where

$G$  = dimensionless time factor

$d$  = diameter of the cylinder (cm)

$t$  = cumulative leaching time since the beginning of the first leaching interval (s).

Values of  $G$  are tabulated in ANS 16.1 as a function of cumulative fraction leached for cylinders having various length-to-diameter ratios. Equation (5) was used to calculate effective diffusivities in those cases where the cumulative release fraction exceeded 20%.

The ANS 16.1 standard defines a material parameter of the leachability of diffusing species. This parameter is called the leachability index,  $L$ , and is defined as:

$$L_i = \frac{1}{k} \sum_{n=1}^k \left| \log(\beta/D_i) \right|_n \quad (6)$$

where

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

$D_i$  = effective diffusivity of nuclide  $i$  determined using either Equation (3) or Equation (5).

$k$  = number of leaching intervals.

As shown in Equation (6), the leachability index of a particular diffusing species is the average value of the incremental leachability indexes determined for  $k$  leaching intervals. The total number of leaching intervals specified to be used in ANS 16.1 is ten. Using Equation (6), average leachability indexes of selected radionuclides, transition metals, and chelating and complexing agents were calculated for each of the seven solidified resin waste specimens which were leach-tested.

## Analytical Methods

The data analysis methods of 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 corresponding to the resin wastes solidified were analyzed using several different analytical techniques. Leachate solutions were analyzed using the same techniques to determine the quantities of diffusing species that had leached from the waste-form specimens. Because of the differences in their physical characteristics and because the concentrations of radionuclides and chelating agents 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 quite different.

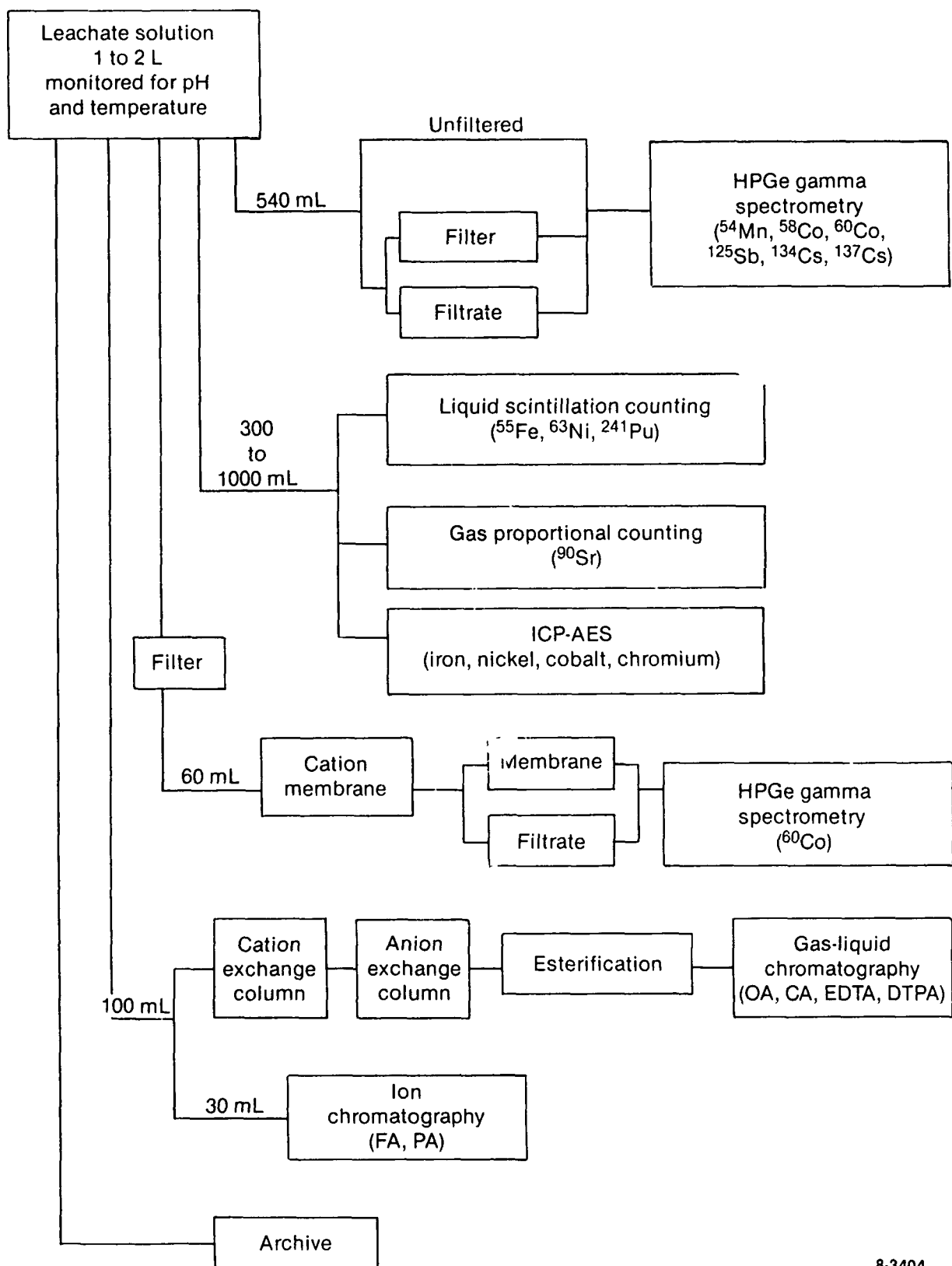
Prior to analysis, leachate solutions were partitioned into volumetric samples according to the analysis scheme shown in Figure 5. The analytical methods used to determine concentrations of radionuclides, transition metals, and chelating agents in leachate samples included high-resolution gamma ray spectrometry, liquid scintillation and gas proportional counting, inductively coupled plasma-atomic emission spectroscopy (ICP-AES), gas-liquid chromatography (GLC), and ion chromatography (IC). Because the cation and mixed-bed resin waste forms obtained from Cooper Station decomposed during leaching, leachate samples for these waste forms were filtered through 0.45- $\mu\text{m}$  filters before being analyzed for concentrations of gamma-emitting radionuclides. The resulting filters were subsequently analyzed to determine the concentrations of particulate species. In order to make a qualitative determination of what fraction of the  $^{60}\text{Co}$  in solution was com-

plexed, a 60-mL sample of each leachate was filtered through a 0.45- $\mu\text{m}$  filter and the filtrate was then passed through a cation exchange membrane. The membrane and membrane filtrate were then analyzed for concentrations of  $^{60}\text{Co}$ . Based on the assumption that uncomplexed  $^{60}\text{Co}$  was retained by the cation membrane, estimates were made of the fraction of  $^{60}\text{Co}$  that was complexed in each leachate. The detection of some of the chelating agents in leachate solutions required that the solutions be purified and concentrated prior to analysis.

Several analytical procedures for detecting and measuring organic acids in waste-form leachates have recently been examined by Brookhaven National Laboratory (BNL). Techniques such as high-performance liquid chromatography (HPLC) and IC are reviewed in Reference 41; and the results of measurements of organic acids performed using spectrophotometric, potentiometric, and IC techniques are discussed in Reference 10. Regular spectrophotometric and potentiometric procedures were determined to be of limited usefulness in determining the concentrations of organic acids in leachates and have not been employed by BNL in more recent studies.<sup>11</sup> Ion chromatography techniques were usually employed by BNL to determine the concentrations of organic acids in leachate solutions. The IC detection limits for organic acids reported by Piciulo et al.<sup>10</sup> were  $10^{-7}$  M EDTA;  $10^{-5}$  M formic acid; and  $10^{-6}$  M citric acid, oxalic acid, or picolinic acid.

The following subsections 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 10 CFR 61 radionuclides (i.e.,  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{242}\text{Cm}$ , and  $^{244}\text{Cm}$ ) at INEL by Westinghouse Idaho Nuclear Company (WINCO) using standard environmental analysis procedures.<sup>42</sup> In each case, samples of resin were removed from the resin waste after it had been homogenized using a stirrer set at low speed. The vials containing the extracted resin samples were intermittently tapped until the resin beads had completely settled into the bottoms of the vials. All free-standing water was then removed from the vials, and the volumes and masses of the tap-settled, wet resin samples were recorded. These resin samples were then ashed and dissolved in concentrated acids following standard



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Figure 5. Leachate solution analysis scheme.



procedures.<sup>42</sup> Because the dissolutions had relatively high specific activities, aliquots of the dissolutions were normally diluted prior to being analyzed. Measured concentrations are expressed in units of  $\mu\text{Ci/mL}$  of tap-settled, wet resin.

As shown in Figure 5, the concentrations of gamma-emitting radionuclides (i.e.,  $^{54}\text{Mn}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$ ) in leachates were determined using hyper-pure germanium (HPGe) spectrometers. In each case, a 540-mL sample of leachate was collected after the leachate had been gently stirred using a stirring rod. This sample was usually filtered through a  $0.45\text{-}\mu\text{m}$  filter, and the filter and filtrate were subsequently analyzed using HPGe spectrometers which had been calibrated using reference sources traceable to the National Bureau of Standards. Both volumetric and filter samples were analyzed using counting geometries for which absolute counting efficiencies had been determined. The resulting spectra were analyzed using the GAUSS VIII gamma-ray spectra analysis code developed at the INEL.

The concentrations of beta-emitting radionuclides (i.e.,  $^{55}\text{Fe}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ , and  $^{241}\text{Pu}$ ) in leachates were measured by WINCO according to standard environmental analysis procedures. Chemical precipitation and extraction techniques were employed to separate the radionuclides of interest. Quantitative analyses were then performed using liquid scintillation and gas proportional counting techniques. In those cases where the leachate had been filtered, the filters were dissolved and added to the corresponding filtrates prior to analysis for beta-emitting radionuclides.

**Elemental Analysis.** Samples of dissolved resin wastes and leachate solutions were analyzed for concentrations of chromium, iron, cobalt, and nickel using ICP-AES. 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. Analyses were performed by WINCO according to standard operating procedures. These procedures included optimizing the spectrometer's optics by peaking on a strong transition metal solution, aligning the aperture plate based on the mercury line position, and calculating metal concentrations using four-point calibration curves. Potential analytical background interferences introduced by the unique sample matrix (e.g., high  $\text{Ca}^{+2}$  concentrations) were evaluated using prepared standards. Only lot-analyzed, trace

metal analysis grade reagents were used to prepare standards. Duplicate samples were analyzed whenever possible.

**Gas-Liquid Chromatography Method.** A GLC method adapted from the techniques reported by Rudling<sup>43</sup> and Eskall and Pick<sup>44</sup> was developed to determine the concentrations of decontamination reagents in resin waste and leachate samples. The GLC technique provides a means of simultaneously determining the concentrations of oxalic acid, citric acid, EDTA, and DTPA in both resin waste and leachate samples. Prior to analysis, the involatile acids are converted to their volatile methyl ester derivatives by reaction with borontrifluoride in methanol. The esterification reaction has been shown<sup>44</sup> not to be affected by the presence of transition metal ions (e.g.,  $\text{Fe}^{+3}$ ,  $\text{Cr}^{+3}$ , and  $\text{Ni}^{+2}$ ) which are present at relatively high concentrations in decontamination resin wastes. The GLC method was used to analyze resin waste and leachate samples representing the Can-Decon, AP/Citrox, and Dow NS-1 decontamination processes, all of which use combinations of the previously mentioned organic acids. The same method was also used to analyze samples of LOMI decontamination resin waste for concentrations of picolinic acid. (IC analysis methods were used to determine the concentrations of formic and picolinic acids in LOMI leachate samples.) Because the concentrations of organic acids in leachate samples were expected to be several orders of magnitude lower than their corresponding concentrations in resin waste samples, methods were developed to concentrate the organic acids in leachate samples prior to their esterification and analysis.

The preconcentration method that was initially tested was freeze-drying relatively large volumes of leachate in volumetric flasks. However, this method was eventually discarded because of the long times required to dry the samples and because of the problems associated with consolidating the dried acids following freeze-drying. In order to eliminate these difficulties, anion exchange resin columns were used to concentrate the organic acids in leachate samples. Preliminary analyses showed that a 100-mL leachate sample volume was normally sufficient to provide adequate analysis sensitivity. The first step in the preparation of a leachate sample involved passing a 100-mL sample through a 6.4-mm diameter by 76-mm long column packed with AG MP-50, 20/50 mesh hydrogen form cation resin. This procedure removed dissolved activity (e.g.,  $^{60}\text{Co}$ ) and also removed cations such as  $\text{Ca}^{+2}$

that might interfere with the GLC analysis. It also served to convert complexed organic acids to uncomplexed forms which could be trapped on the anion resin column. The eluate of the cation exchange column was then passed through a similarly sized column packed with AG 1-X8, 100/200 mesh chloride form anion resin. Uncomplexed organic acids are retained on the anion exchange resin. The anion resin was then flushed from the column into a 5-mL reacti-vial using deionized water. The sample was placed in a freezer overnight, and then it was freeze-dried under reduced pressure.

Following freeze-drying, the anion resin was esterified directly so as to achieve a maximum concentration factor. A triangular stirring bar and 1 mL of boron trifluoride in 14% (w/v) methanol were added to the reacti-vial. The vial was sealed and placed in a reacti-therm heating/stirring block where it was stirred continuously for 2 h while heated to 348 K (75°C). Following esterification, the sample was allowed to cool to room temperature; and then 1 mL of HPLC/Spectro-grade chloroform was injected into the vial using a gas-tight syringe. The vial was then shaken vigorously to mix the solutions; then 3 mL of  $\text{KH}_2\text{PO}_4$  buffer and 100  $\mu\text{L}$  of a solution of ethyl benzoate and chloroform, which was used as an internal standard, were added. The sample was next shaken vigorously for 2 min using an electric vibrator/mixer and then centrifuged for 15 min at 4,000 rpm to separate the aqueous and chloroform components. The aqueous layer was removed and discarded using a Pasteur pipette, and the chloroform layer containing the methyl esters of the organic acids was pipetted into a 1-mL serum vial which was sealed as soon as the transfer was completed. It was this chloroform component that was analyzed by GLC.

Waste resin samples were esterified using the same procedure employed to esterify the anion resins used to concentrate leachate organic acids. Waste resin samples were removed from the original waste resin containers after the resins had been homogenized using a mixer set at low speed. The masses of the waste resin samples that were esterified and analyzed by GLC ranged from 0.34 to 0.50 g (dry mass). These samples, in their original states, exhibited relatively high specific activities; however, in each case, following esterification, gamma-emitting radionuclides (i.e.,  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ) were segregated exclusively to the aqueous layer. No detectable activity was measured in the chloroform components of the esterified resin waste samples.

A Perkin Elmer model Sigma 300 gas chromatograph equipped with a flame ionization detector (FID) was used to measure the concentration of the methyl ester derivatives. The chromatograph column was contained in an oven which could be operated at temperatures from 308 K (35°C) up to 773 K (500°C) and was fitted with a single-channel temperature controller that provided linear temperature programming for the oven. Separation of the esters was achieved using a 6.4-mm outside diameter, 1.83-m-long glass column packed with 5% SP-2250 on 100/120 mesh Supelcoport. Nitrogen flow through the column was kept constant at 30  $\text{cm}^3/\text{min}$ . The column injector temperature was set to 558 K (285°C), and the FID temperature was kept constant at 583 K (310°C). The volume of sample used for analysis was 5  $\mu\text{L}$ . Following sample injection, the column oven temperature was maintained at 323 K (50°C) for 4 min and was then raised at 10 K/min (10°C/min) to 583 K (310°C). The lower temperature was found to be optimum for the elution of the ester of oxalic acid, while the optimum temperatures for the elution of the esters of the other acids were above 423 K (150°C). For each GLC analysis, chromatograms and areas of individual peaks were recorded on a Hewlett-Packard Model 3390A integrator. The integrator recorded peak retention times and peak fit parameters. Results were recorded as the ratio of the area of the ester peak to the area of the internal standard peak. Ethyl benzoate was chosen as the internal standard because it yielded a sharp symmetrical peak which did not interfere with the peaks of any of the other esters.

To assess whether or not the esterification of the chloroform anion resin produced any peaks that interfered with the internal standard peak or the peaks of the esters of the organic acids of interest, two samples of the anion resin that contained no acids were esterified following the standard procedure. To one of the two samples, 100  $\mu\text{L}$  of ethyl benzoate internal standard solution was added; but no internal standard was added to the second sample. Five- $\mu\text{L}$  samples of the chloroform layers of both samples were analyzed using the standard gas chromatograph operating parameters. Neither of the chromatograms of these samples exhibited any interfering peaks.

In order to determine calibration functions for the gas chromatograph, standards were prepared that contained known amounts of oxalic acid, citric acid, EDTA, and DTPA. Calibration standards were prepared by either freeze-drying aliquots of a standard solution containing known quantities of

the four acids or by directly measuring the individual masses of the acids added to a mixture of the four acids. The former method was used to prepare the standards having the smaller masses, while the latter technique was used to make the larger mass standards. Duplicate sets of six standards were prepared that contained the following masses of each of the four acids: 0.1 g, 0.2 g, 0.5 g, 1.0 g, 3.0 g, and 5.0 g. (The exact masses of each acid were close to these values.) Following drying, the standards were esterified following the procedure previously described. Each of the twelve standards was analyzed five or more times to determine a response curve for each of the four acids. In each case, the response, expressed as the ratio of the peak area of the methyl ester of the acid to the peak area of the ethyl benzoate internal standard, was a linear function of acid mass. For each acid, the calibration data were fit using linear regression methods to determine the slope and intercept of the linear calibration function and the error in the estimate of measured concentrations. Correlation coefficients of these fits were equal to or better than 0.9999, indicating that the response functions closely approximated straight lines.

Picolinic acid calibration standards were also prepared following the same procedures used to prepare the standards containing oxalic acid, citric acid, EDTA, and DTPA. Four standards were prepared that contained about 1.4 g, 4.5 g, 10.9 g, and 33.0 g of picolinic acid. Following freeze-drying, these standards were esterified according to the usual procedure. The peak areas of the methyl ester of picolinic acid were divided by the corresponding peak areas of the ethyl benzoate internal standard, and the data were fit using linear regression methods to determine the slope and intercept of the linear calibration function and the error in the estimate of measured concentrations. The correlation coefficient of the fit was 0.99994.

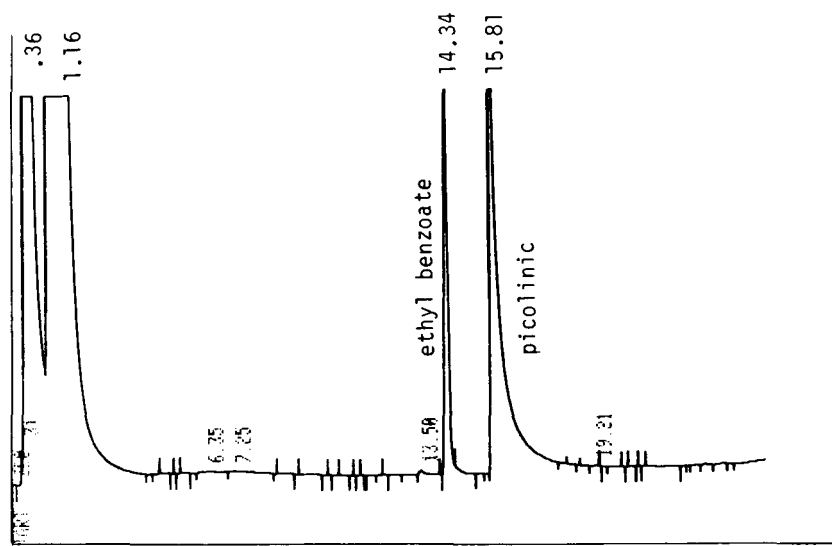
The chromatogram in Figure 6 shows the separation of ethyl benzoate and the ester of picolinic acid where the analysis was performed on the 4.5-mg picolinic acid calibration standard. Retention times were, respectively, 14.3 and 15.8 min. The broad peaks having retention times less than 2 min are chloroform peaks. The picolinic acid standard was diluted by a factor of five prior to the analysis. Figure 7 shows the separation of the esters of oxalic acid, citric acid, EDTA, and DTPA where the analysis was performed on a calibration standard containing about 3 mg of each of the four acids. Retention times were 8.5, 19.5, 27.3, and 35.4 min, respectively. The chloroform extract of this stand-

ard was also diluted by a factor of five prior to analysis to prevent the integrator from overranging at peak locations.

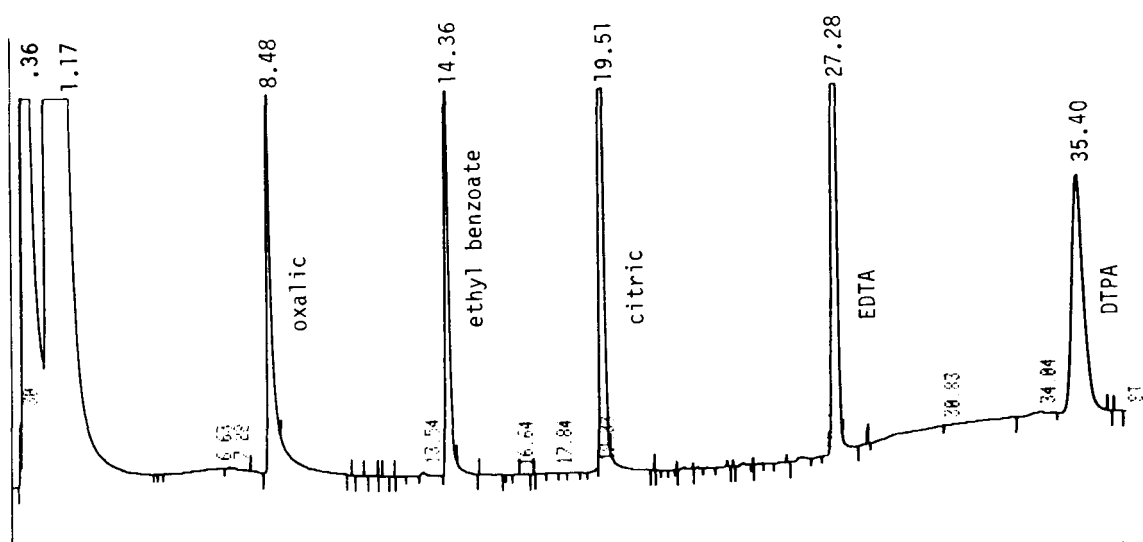
In order to confirm that transition metal ions did not interfere with the GLC analysis of leachate samples, a standard solution containing oxalic acid, citric acid, and EDTA was analyzed with and without a metal ion component. Following an initial analysis, quantities of chromium (as chromium chloride) and iron (as ferrous chloride) were added to the standard solution such that their concentrations were each about 100  $\mu\text{g/mL}$ . Concentrations of the three acids were each about 10  $\mu\text{g/mL}$ . Samples of the spiked solution were freeze-dried, esterified, and analyzed using standard procedures. The analytical response of the GLC for oxalic acid, citric acid, and EDTA was not affected by the presence of these metal ions.

Measurements were also performed to determine the retention efficiency of the anion exchange resin column used to preconcentrate leachate organic acids. Standard solutions, each having a volume of 100 mL, were prepared which contained approximately equal quantities of oxalic acid, citric acid, EDTA, and DTPA. In order to simulate the composition of typical leachate samples,  $\text{Ca}(\text{OH})_2$  was added to the solutions such that its concentration was about 200  $\mu\text{g/mL}$ . The 100-mL solutions were then processed through the cation and anion exchange resin columns using the same procedure followed when preconcentrating leachate samples. The anion resins were then esterified and analyzed according to the usual procedures. At acid concentrations of 10  $\mu\text{g/mL}$ , the retention efficiencies were: oxalic acid, 87%; citric acid, 96%; EDTA, 94%; and DTPA, 94%. At concentrations of 30  $\mu\text{g/mL}$ , the results were: oxalic acid, 109%; citric acid, 116%; EDTA, 116%; and DTPA, 88%. The ranges of the efficiencies reflect the cumulative uncertainties associated with preparation of the standards, esterification, and GLC analysis. The results indicate that the organic acids were very efficiently removed by the anion resin exchange column in the 10- to 30- $\mu\text{g/mL}$  concentration range.

**Ion Chromatography Methods.** The leachate samples resulting from the leach testing of the Indian Point-3 waste-form specimen were analyzed for picolinic and formic acids using IC methods. Picolinic acid has a distinctive absorption spectrum in the UV region with peaks at 264, 267, 271, and 275 nm. The concentration of picolinic acid can, therefore, be determined by direct measurement.



**Figure 6.** Chromatogram of picolinic acid calibration standard.



**Figure 7.** Chromatogram of a calibration standard containing oxalic acid, citric acid, EDTA, and DTPA.



## EXPERIMENTAL RESULTS

A primary objective of this leach study was to determine effective diffusivities and leachability indexes of diffusing species using diffusion models described previously. Using the models, measured effective diffusivities can be used to predict long-term leaching behavior, assuming that the release mechanisms do not change with age or degradation of the samples tested. Approximate and exact solutions of the general differential equation describing diffusion-controlled releases from the waste-form specimens were used. The approximate solution, the semi-infinite medium diffusion model, was used to calculate effective diffusivities by applying Equation (3) to incremental release data; and the exact solution, the finite-cylinder diffusion model, was used to calculate effective diffusivities by applying Equation (5) to cumulative release data. The finite-cylinder model was used to analyze leach data when the cumulative release of a species equaled or exceeded 20% of its initial inventory in the waste-form specimen. Leachability indexes were calculated for each leaching interval using the definition of leachability index given by Equation (6), and the average value of the leachability index of a species was calculated as the simple arithmetic average of the leachability indexes determined for all leaching intervals. Standard deviations of the mean were calculated for all average leachability indexes.

This section presents the results of measurements of the concentrations of radionuclides, transition metals, and decontamination reagents in samples of resin waste collected from five operating LWRs and also presents the results of measurements of the release of these species from the same resin wastes solidified in Portland Type I or Type II cement. Release data are shown graphically as plots of cumulative fraction release (CFR) 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, according to Equation (2), have slopes that are proportional to the corresponding effective diffusivities. The results of data analysis performed according to ANS 16.1 are given in tabular form for each radionuclide, transition metal, and organic acid. Release rates for these species are also tabulated and are expressed both as quantity released per second per unit sample surface area ( $\mu\text{Ci}/\text{cm}^2/\text{s}$  or  $\mu\text{g}/\text{cm}^2/\text{s}$ ) and as fractional release per second divided by the

sample's surface-area-to-volume ratio ( $\text{cm}/\text{s}$ ). Release rates were also calculated expressed as fractional release per second per square centimeter of sample external surface area ( $\text{F}/\text{cm}^2/\text{s}$ ).

Release rates, incremental and cumulative release fractions, effective diffusivities, and leachability indexes of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ ,  $^{137}\text{Cs}$ , and  $^{241}\text{Pu}$  are presented in Appendix E. Similar leach data for the transition metals iron, nickel, cobalt, and chromium are also presented in Appendix E. Leaching results for chelating or complexing agents—oxalic acid, citric acid, EDTA, DTPA, picolinic acid, and formic acid—are given in Appendix F. Radionuclide and stable metal concentrations in waste-form leachates are presented in Appendix G, and the concentrations of chelating and complexing agents in the leachates are tabulated in Appendix H.

### Concentrations in Resin Wastes

The concentrations of 10 CFR 61 radionuclides, selected transition metals, and decontamination reagents in the resin waste samples obtained from Indian Point-3, Cooper, Millstone-1, Pilgrim, and Peach Bottom-2 were measured in order to determine the quantities of these species present in the solidified waste-form specimens prior to the initiation of leach-testing. The initial quantity of a radionuclide or other species of interest in a waste-form specimen was estimated by multiplying its measured concentration in the resin waste sample, expressed as  $\mu\text{Ci}/\text{cm}^3$  or  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin, times the volume of wet resin in the solidified waste-form sample. The waste loadings used to estimate initial inventories are itemized in Table 9, where they are expressed in units of volume percent. The waste loadings of the two Cooper waste-form samples were determined using the results of measurements of the volumes and/or masses of resin, cement, slaked lime, and NaOH that were added to the liners from which waste-form samples were obtained. All other resin waste loadings were assumed to be the same as those specified by Chem-Nuclear Services, Inc., in Reference 30. The waste loadings in Table 9 correspond to wet resin volumes in the solidified samples that range from about  $61\text{ cm}^3$  for the smaller Millstone-1 sample to about  $116\text{ cm}^3$  for the Pilgrim sample. Using the densities and water contents of "dewatered" resins presented in Table 3, the wet and dry resin loadings of the

waste-form specimens were calculated. The results are presented in Table 9. These data indicate that the wet resin loadings of the samples ranged from 60 to 66 wt. % and the dry resin loadings ranged from 17 to 23 wt. %.

The concentrations of radionuclides in the resin wastes are presented in Table 10, where the results are expressed as  $\mu\text{Ci}/\text{cm}^3$  of tap-settled, wet resin decay-corrected to the sample collection dates. The reported uncertainties are one sigma values. The concentrations of  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ , and  $^{63}\text{Ni}$  in the resins are plotted in Figure 8, while similar results for  $^{54}\text{Mn}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  are plotted in Figure 9. Results for  $^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ , and  $^{241}\text{Pu}$  are shown in Figure 10.

The data presented in Table 10 and Figures 8 through 10 show that  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{63}\text{Ni}$  were the major activities in the resin wastes. The concentrations of these radionuclides ranged from about 0.1 to 20  $\mu\text{Ci}/\text{cm}^3$ . Cobalt-60 made up from 24% to 59%,  $^{55}\text{Fe}$  from 15% to 62%,  $^{63}\text{Ni}$  from 3% to 43%, and  $^{54}\text{Mn}$  from 4% to 8% of the total radionuclide inventories of the resins. The maximum concentrations of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  were, on the other hand, only 0.13 and  $5.9 \times 10^{-4}$   $\mu\text{Ci}/\text{cm}^3$ , respectively. These two radionuclides generally constituted less than 1% of the total radionuclide inventories of the resins. Among the transuranics,  $^{241}\text{Pu}$  exhibited the highest activity;  $^{241}\text{Pu}$  concentrations ranged from 0.35 to  $5 \times 10^{-3}$   $\mu\text{Ci}/\text{cm}^3$ . The concentrations of transuranics in the Millstone-1 resin waste were typically an order of magnitude higher than corresponding concentrations in other resin wastes which, excluding  $^{241}\text{Pu}$ , normally ranged from  $1 \times 10^{-6}$  to  $1 \times 10^{-4}$   $\mu\text{Ci}/\text{cm}^3$  (see Figure 10). Omitting  $^{58}\text{Co}$ , which has a half-life of only 70.9 days, total radionuclide concentrations varied from about 1.7  $\mu\text{Ci}/\text{cm}^3$  for the mixed-bed resin sample from Cooper to 31.7  $\mu\text{Ci}/\text{cm}^3$  for the mixed-bed resin sample from Millstone-1. Excluding the result for the Cooper mixed-bed resin, the average loading on the remaining five resin waste samples was  $17 \pm 4$   $\mu\text{Ci}/\text{cm}^3$ . Differences in radionuclide concentrations should not be interpreted as reflecting differences in the effectiveness of one decontamination process over another, since the surface areas decontaminated and the volumes of resin used to process the decontamination solutions were different.

The concentrations of chromium, iron, cobalt, and nickel in the resin wastes are presented in Table 11 and Figure 11, where the results are expressed as  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin. In general, the spectrochemical analysis results were

accurate only to  $\pm 5\%$ . In those cases where duplicate analyses were performed, the reported uncertainties are the standard deviations of the mean values. Standard deviations of duplicate measurements ranged from about  $\pm 8\%$  to  $\pm 40\%$ . A large deviation can be attributed to a nonhomogeneous distribution of the metal in the resin sample. Iron and nickel were the major transition metals in the resins, while chromium and cobalt were present at trace levels. The concentration of iron ranged from about  $5 \times 10^2$   $\mu\text{g}/\text{cm}^3$  in the Cooper mixed-bed resin to  $1 \times 10^4$   $\mu\text{g}/\text{cm}^3$  in the Cooper cation resin. The concentrations of the other transition metals were also significantly higher in the cation resin as compared to the mixed-bed resin. Because the Cooper cation resin was used to regenerate decontamination reagents during the decontamination of the PCRS and the mixed-bed resin was used to remove decontamination reagents and residual soluble corrosion products following completion of decontamination, it is not surprising that the concentrations of cationic species in the two resins are very different. Concentrations of nickel varied from 74  $\mu\text{g}/\text{cm}^3$  in the Cooper mixed-bed resin to about  $2 \times 10^3$   $\mu\text{g}/\text{cm}^3$  in the Millstone-1 resin waste.

Examples of chromatograms of the resin wastes obtained by the GLC method are presented in Appendix C, Figures C-1 through C-6. Some of the chromatograms exhibited peaks that have not yet been identified. It is possible that some of these peaks are organo-metal complexes (see Figures C-2 and C-5). Concentrations of organic acids in the resin wastes measured using the GLC method are presented in Table 12 and Figure 12, where the results are expressed as  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin. Uncertainties are the quadrature sums of one sigma systematic and random errors. Systematic errors were calculated using the uncertainties in the slopes and intercepts of the gas chromatograph calibration functions, and random errors were determined from the results of multiple analyses of individual resin samples. The ranges of reagent loadings on the resin wastes, expressed as  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin, were: oxalic acid,  $2.7 \times 10^2$  to  $1.6 \times 10^4$ ; citric acid,  $9.6 \times 10^1$  to  $2.5 \times 10^4$ ; and EDTA,  $3.7 \times 10^3$  to  $6.1 \times 10^3$ . The concentration of DTPA in the Pilgrim resin waste was  $3.6 \times 10^2$   $\mu\text{g}/\text{cm}^3$ , and the concentration of picolinic acid in the Indian Point-3 resin waste was  $4.1 \times 10^3$   $\mu\text{g}/\text{cm}^3$ . The concentration of formic acid listed in Table 12 is not a measured value; it is simply an estimate based on the concentration of picolinic acid in the resin. With the 5% SP-2250

**Table 10. Concentrations of radionuclides in resin wastes ( $\mu\text{Ci}/\text{cm}^3$ )<sup>a</sup>**

Nuclide	Indian Point-3 (06/24/85)	Cooper (MBR) (11/30/84)	Cooper (CR) (11/30/84)	Millstone-1 (06/20/84)	Pilgrim (08/17/84)	Peach Bottom-2 (08/09/84)
<sup>3</sup> H	5.1 ± 0.5 (−4)	3.8 ± 0.1 (−4)	3.9 ± 0.2 (−6)	1.14 ± 0.04(−3)	2.13 ± 0.09(−5)	2.6 ± 0.1 (−2)
<sup>14</sup> C <sup>b</sup>	4.8 ± 0.8 (−5)	< 1 (−5)	< 1 (−5)	3.6 ± 0.2 (−5)	5 ± 1 (−5)	— <sup>c</sup>
<sup>54</sup> Mn	4.81 ± 0.07(−1)	1.45 ± 0.07(−1)	1.04 ± 0.03(+0)	2.44 ± 0.04(+0)	3.8 ± 0.2 (−1)	1.68 ± 0.01(−1)
<sup>55</sup> Fe	1.69 ± 0.08(+0)	4.0 ± 0.2 (−1)	5.7 ± 0.3 (+0)	2.0 ± 0.1 (+1)	2.7 ± 0.1 (+0)	— <sup>c</sup>
<sup>57</sup> Co	4.4 ± 0.4 (−2)	— <sup>d</sup>	— <sup>d</sup>	— <sup>d</sup>	— <sup>d</sup>	— <sup>d</sup>
<sup>58</sup> Co	2.25 ± 0.01(+1)	3.9 ± 0.6 (−2)	7.1 ± 0.6 (−1)	5 ± 2 (−1)	9 ± 9 (−2)	— <sup>d</sup>
<sup>60</sup> Co	3.93 ± 0.01(+0)	7.31 ± 0.08(−1)	6.69 ± 0.03(+0)	7.81 ± 0.03(+0)	5.19 ± 0.02(+0)	1.18 ± 0.01(+1)
<sup>63</sup> Ni	4.8 ± 0.2 (+0)	3.6 ± 0.2 (−1)	1.03 ± 0.05(+1)	8.7 ± 0.4 (−1)	2.8 ± 0.1 (−1)	— <sup>c</sup>
<sup>89</sup> Sr	5 ± 1 (−3)	< 2 (−5)	< 9 (−6)	1.3 ± 0.4 (−2)	1.0 ± 0.6 (−3)	— <sup>c</sup>
<sup>90</sup> Sr	1.3 ± 0.2 (−4)	2.0 ± 0.3 (−4)	7 ± 1 (−5)	5.9 ± 0.4 (−4)	1.5 ± 0.2 (−4)	1.6 ± 0.3 (−5)
<sup>95</sup> Nb	— <sup>d</sup>	9 ± 3 (−3)	— <sup>d</sup>	— <sup>d</sup>	— <sup>d</sup>	— <sup>d</sup>
<sup>99</sup> Tc	— <sup>c</sup>	1.1 ± 0.3 (−4)	3 ± 3 (−5)	— <sup>d</sup>	2.2 ± 0.6 (−5)	— <sup>c</sup>
<sup>129</sup> I	< 1 (−5)	< 3 (−5)	< 4 (−5)	< 2 (−6)	< 2 (−6)	— <sup>c</sup>
<sup>125</sup> Sb	9.3 ± 0.7 (−4)	4.2 ± 0.5 (−2)	< 2 (−4)	1.6 ± 0.4 (−1)	8.4 ± 0.4 (−3)	< 6 (−5)
<sup>137</sup> Cs	1.34 ± 0.04(−1)	2.1 ± 0.2 (−4)	< 2 (−4)	2.1 ± 0.1 (−3)	3 ± 1 (−2)	2.83 ± 0.02(−3)
<sup>238</sup> Pu	1.16 ± 0.02(−5)	3.1 ± 0.2 (−5)	7.7 ± 0.3 (−5)	4.9 ± 0.1 (−3)	2.2 ± 0.1 (−4)	— <sup>c</sup>
<sup>239</sup> <sup>240</sup> Pu	1.30 ± 0.07(−5)	1.50 ± 0.08(−5)	3 ± 2 (−5)	2.60 ± 0.05(−3)	3.1 ± 0.1 (−4)	— <sup>c</sup>
<sup>241</sup> Pu	4.9 ± 0.6 (−2)	2.2 ± 0.1 (−2)	1.01 ± 0.04(−2)	3.5 ± 0.1 (−1)	3.5 ± 0.1 (−2)	5.1 ± 0.6 (−3)
<sup>241</sup> Am	9.4 ± 0.4 (−6)	2.35 ± 0.03(−5)	1.05 ± 0.03(−4)	3.99 ± 0.04(−3)	5.3 ± 0.2 (−6)	— <sup>c</sup>
<sup>242</sup> Cm	1.9 ± 0.1 (−4)	6.35 ± 0.06(−5)	6.8 ± 0.2 (−5)	4.1 ± 0.2 (−4)	1.74 ± 0.06(−5)	— <sup>c</sup>
<sup>244</sup> Cm	1.07 ± 0.09(−5)	6.64 ± 0.09(−5)	8.1 ± 0.2 (−5)	3.40 ± 0.04(−3)	1.38 ± 0.02(−5)	— <sup>c</sup>

a. Activities are decay-corrected to sample collection dates and are expressed as  $\mu\text{Ci}/\text{cm}^3$  of tap-settled, wet resin.

b. Activity expressed as  $\mu\text{Ci}/\text{cm}^3$  of dry resin.

c. Analysis not performed.

d. Not detected.



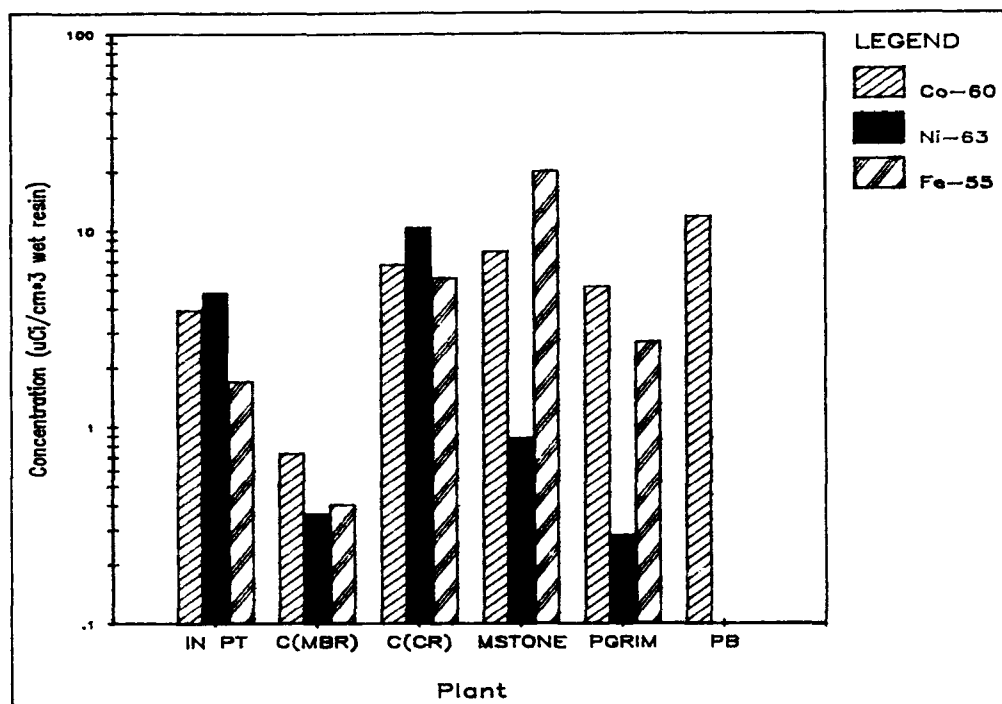


Figure 8. Concentrations of  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ , and  $^{63}\text{Ni}$  in resin wastes.

column, the GLC technique was not successful when applied to formic acid because of the low molecular weight and boiling point of methyl formate, which are 60.05 and 305 K (31.5°C), respectively. The boiling point of chloroform is 334 K (61.3°C), and, consequently, the chloroform solvent peak masked the methyl formate peak, making its detection impossible.

The concentrations of oxalic and citric acids in the Cooper cation resin waste were, as expected, substantially lower than their corresponding concentrations in the Cooper mixed-bed resin. The total reagent loadings on the resin wastes, expressed as weight percents, were: Cooper mixed-bed resin, 4.1; Millstone-1, 2.0; Peach Bottom-2, 1.6; Indian Point-3, 0.8; Pilgrim, 0.7; and Cooper cation resin, 0.04. The relatively low loading on the Indian Point-3 resin can be explained by the fact that resins from the plant radwaste system were mixed with the decontamination resins prior to solidification. Specific information regarding the types of resins that were solidified at Pilgrim is not available (see Table 9 for resin types); however, the fact that a larger-than-usual volume of resin (i.e., 2.55 m<sup>3</sup>) was used to process the decontamination solution might explain the low loading.

## Release of Radionuclides from Waste Forms

This section presents the results of measurements of the release of radionuclides from the seven solidified decontamination resin waste specimens which were leach-tested according to the ANS 16.1 procedure. The physical parameters of the specimens and the types of resins they contained are given in Table 9. The resins in the solidified specimens had been used to process Can-Decon, Citrox, Dow NS-1, and LOMI decontamination solutions following chemical decontaminations of primary coolant systems. Consequently, they contained significant quantities of transition metals and decontamination reagents in addition to the activity removed as a result of decontamination.

Leachate samples were not analyzed for all of the radionuclides listed in Table 10. A limited number were selected based on the likelihood of their being detected in the leachates. The radionuclides having the highest concentrations in the resin wastes and those that were thought to have relatively high release rates were selected for analysis. Using these criteria, the following radionuclides were selected for analysis:  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,

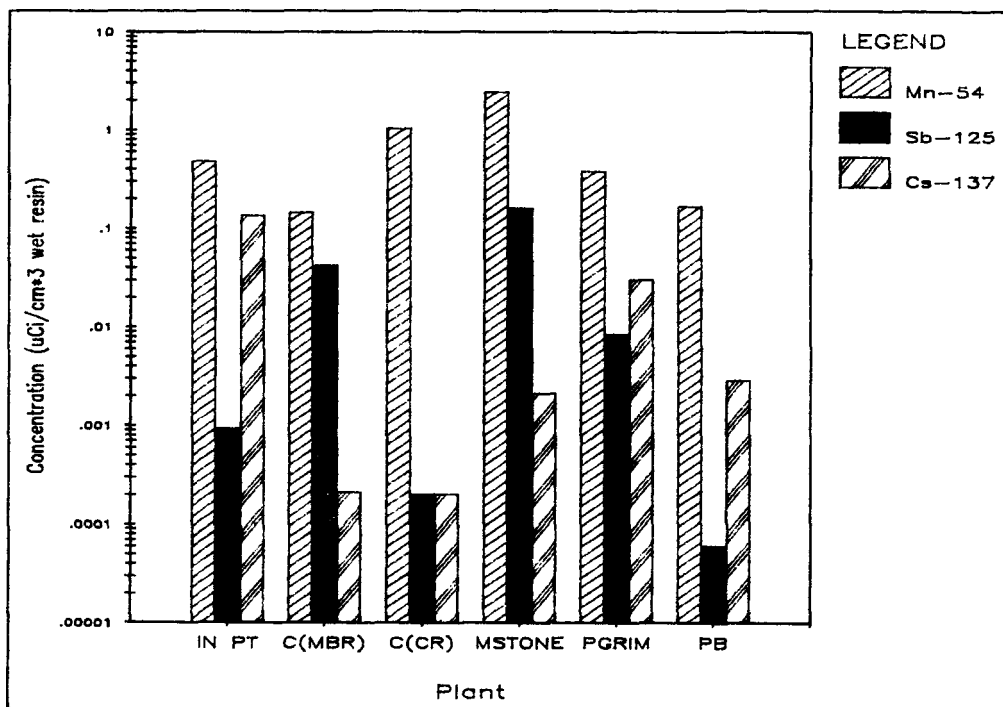


Figure 9. Concentrations of  $^{54}\text{Mn}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  in resin wastes.

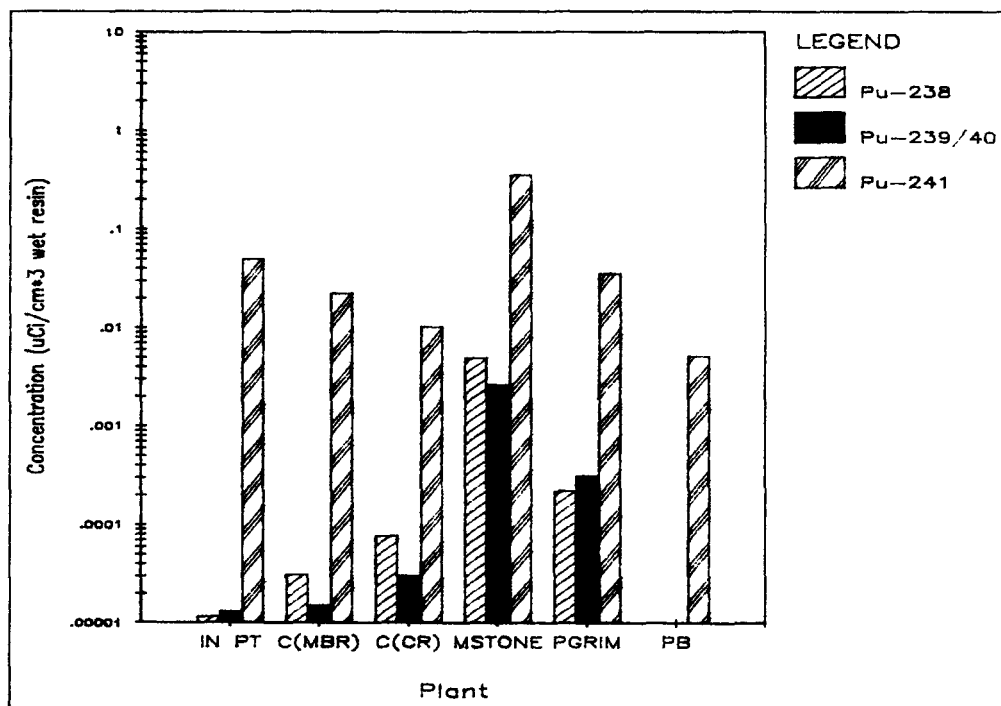


Figure 10. Concentrations of  $^{238}\text{Pu}$ ,  $^{239-240}\text{Pu}$ , and  $^{241}\text{Pu}$  in resin wastes.

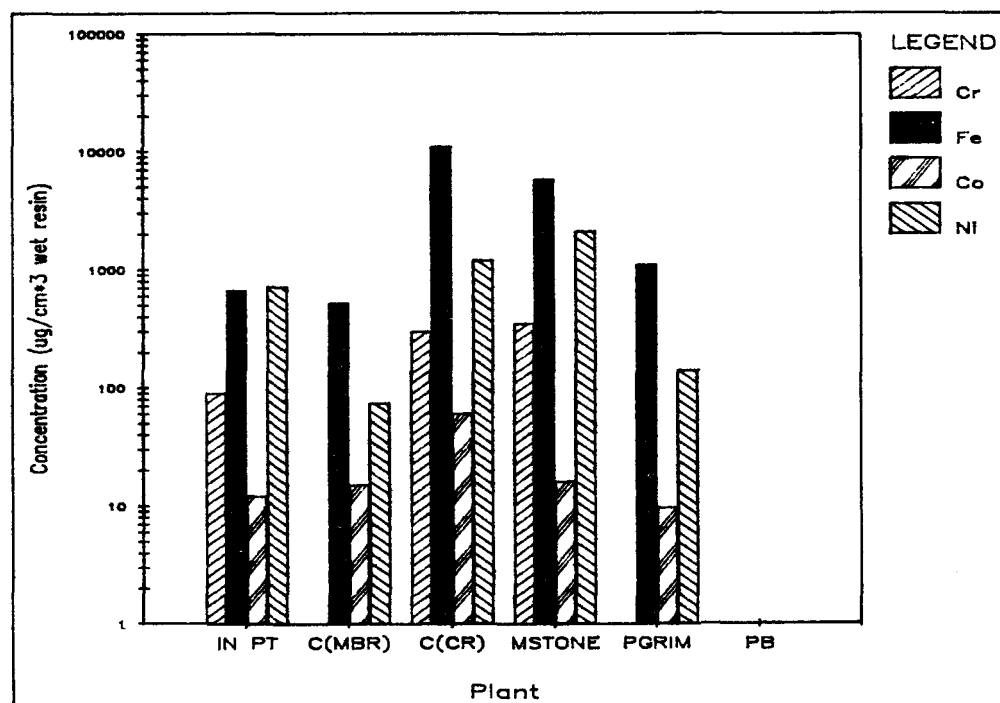
**Table 11. Concentrations of stable metals in resin wastes ( $\mu\text{g}/\text{cm}^3$ )<sup>a</sup>**

Plant	Chromium	Iron	Cobalt	Nickel
Indian Point-3	9.0 $\pm$ 0.5 (+1)	6.6 $\pm$ 0.3 (+2)	< 1.2 (+1)	7.1 $\pm$ 0.4 (+2)
Cooper (MBR)	— <sup>b</sup>	5.2 $\pm$ 0.6 (+2) <sup>c</sup>	< 1.5 (+1)	7.4 $\pm$ 0.6 (+1) <sup>c</sup>
Cooper (CR)	3.0 $\pm$ 1 (+2)	1.1 $\pm$ 0.4 (+4) <sup>c</sup>	6.0 $\pm$ 2 (+1)	1.2 $\pm$ 0.5 (+3) <sup>c</sup>
Millstone-1	3.5 $\pm$ 0.2 (+2)	5.8 $\pm$ 0.3 (+3)	1.60 $\pm$ 0.08(+1)	2.1 $\pm$ 0.1 (+3)
Pilgrim	— <sup>b</sup>	1.10 $\pm$ 0.05(+3)	9.6 $\pm$ 0.5 (+0)	1.40 $\pm$ 0.07 (+2)
Peach Bottom-2	— <sup>b</sup>	— <sup>b</sup>	— <sup>b</sup>	— <sup>b</sup>

a. Concentrations are expressed as  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin.

b. Analysis not performed.

c. Standard deviation of results for duplicate samples.



**Figure 11. Concentrations of chromium, iron, cobalt, and nickel in resin wastes.**

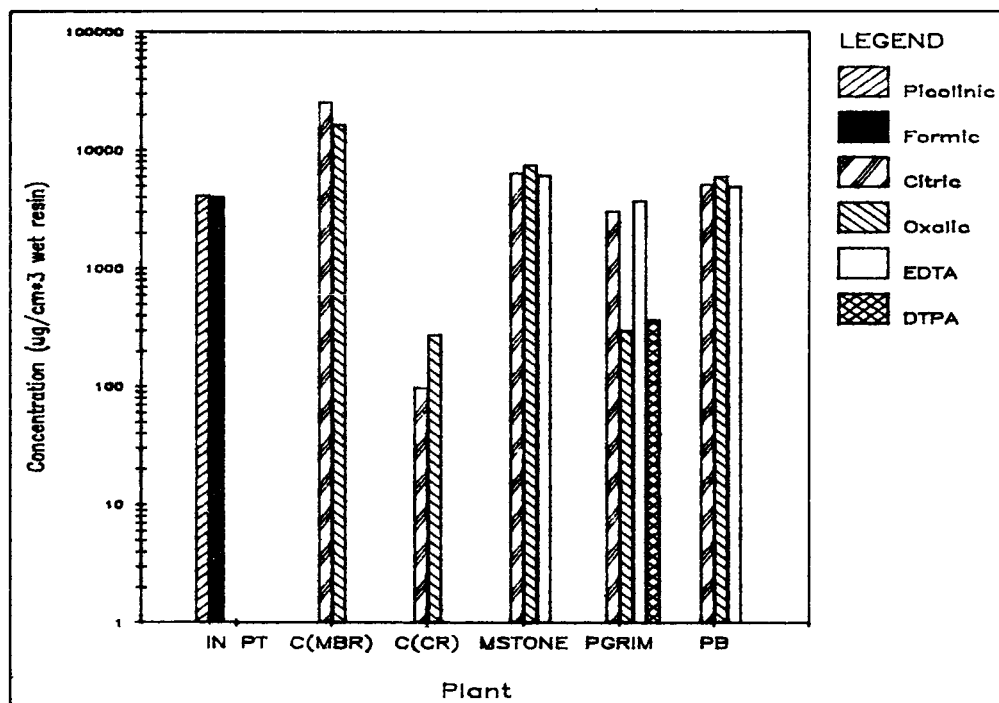
**Table 12. Concentrations of organic acids in resin wastes ( $\mu\text{g}/\text{cm}^3$ )<sup>a</sup>**

Plant	Oxalic	Citric	EDTA	DTPA
Cooper (MBR)	1.62 $\pm$ 0.03 (+4)	2.5 $\pm$ 0.1 (+4)	— <sup>b</sup>	— <sup>b</sup>
Cooper (CR)	2.70 $\pm$ 0.01 (+2)	9.6 $\pm$ 1.1 (+1)	— <sup>b</sup>	— <sup>b</sup>
Millstone-1	7.4 $\pm$ 0.2 (+3)	6.3 $\pm$ 0.3 (+3)	6.1 $\pm$ 0.4 (+3)	— <sup>b</sup>
Pilgrim	2.93 $\pm$ 0.08 (+2)	3.0 $\pm$ 0.1 (+3)	3.7 $\pm$ 0.3 (+3)	3.62 $\pm$ 0.09 (+2)
Peach Bottom-2	5.91 $\pm$ 0.07 (+3)	5.08 $\pm$ 0.04 (+3)	4.9 $\pm$ 0.3 (+3)	— <sup>b</sup>
	Formic	Picolinic		
Indian Point-3	4.0 $\pm$ est. (+3) <sup>c</sup>	4.1 $\pm$ 0.2 (+3)		

a. Concentrations expressed as  $\mu\text{g}/\text{cm}^3$  tap-settled, wet resin.

b. Not detected.

c. Concentration estimated based on measured concentration of picolinic acid.



**Figure 12.** Concentrations of organic acids in resin wastes.

$^{125}\text{Sb}$ ,  $^{137}\text{Cs}$ , and  $^{241}\text{Pu}$ . All leachate samples were analyzed by gamma ray spectrometry; and, therefore, the concentrations of the gamma-emitting radionuclides (i.e.,  $^{54}\text{Mn}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$ ) were normally determined by a single measurement. Because specific radiochemical separations were required for the determinations of the concentrations of  $^{55}\text{Fe}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ , and  $^{241}\text{Pu}$ , budgetary restraints limited the number of leachate samples that could be analyzed for these beta-emitting radionuclides. Approximately one-half of the total number of leachate samples were analyzed for these beta emitters. Consequently, in most cases, it was not possible to determine CFR values for these four radionuclides. Also, for some analyses, detection limits were not available for certain gamma-emitting radionuclides. Radionuclides such as  $^{54}\text{Mn}$  and  $^{125}\text{Sb}$  were frequently not detected; and, in some cases, the lack of detection limits made it impossible to calculate the CFR values of these radionuclides.

**Can-Decon Waste Forms.** Three Can-Decon waste-form samples were leach-tested. Two of these samples were collected from the same liner at Millstone-1, and the third was obtained from Peach Bottom-2. All three samples contained mixed-bed resins consisting of Rohm and Haas IRN-77 cation and IRN-78 anion exchange resins. The Millstone-1 samples, designated samples F33 and F201, were the same diameter but only about one-half the length of the Peach Bottom-2 sample (see Table 9). Because of their shorter lengths, the Millstone-1 samples had volume-to-surface-area ratios of about 0.8, whereas the corresponding value for the Peach Bottom-2 sample was about 0.9. The surfaces of the Millstone-1 samples had a similar texture, which was somewhat coarser than the surface of the Peach Bottom-2 sample. The surface of the latter sample was smooth except for the top surface, which was rough and irregular.

During the prerinse and subsequent leaching intervals, a slow deterioration of both Millstone-1 samples was observed. The samples appeared to remain intact during the leach-testing, but as much as 3% to 5% of the mass of each sample had sloughed off the ends of the samples after 3 months of continuous leaching. The F201 sample, which exhibited a superficial crack prior to the start of leaching, did show noticeably greater deterioration compared to the F33 sample, and the crack did widen and deepen slightly. The Peach Bottom-2 sample, when initially immersed in the leachant, bubbled for a short time, but much less

vigorously than most of the other waste-form samples that were tested. This sample did not exhibit any observable deterioration during 90 days of continuous leaching, other than a few small chips flaking off the ends of the cylinder.

A summary of the radionuclide leach-test results for the Can-Decon waste-form samples is given in Table 13. For each sample, the cumulative quantity of each radionuclide released during leaching, expressed as fraction of initial inventory, is presented in Table 13, along with the ranges of the measured effective diffusivities and leachability indexes. Because of budgetary restraints, the Millstone-1 F33 leachates were not analyzed for beta-emitting radionuclides; therefore, results for  $^{55}\text{Fe}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ , and  $^{241}\text{Pu}$  are not presented for this sample. Radionuclide CFR values for the three Can-Decon waste forms are presented graphically in Figures 13 through 18, where CFR values are plotted against the square root of elapsed leaching time. According to Equation (2), the CFR data presented in this fashion should approximate straight lines if diffusion is the controlling release mechanism. Average radionuclide release rates during individual leaching intervals are presented in Appendix E, and radionuclide concentrations in the leachate samples are tabulated in Appendix G.

The results presented in Table 13 show that the effective diffusivities determined for a given radionuclide varied by as much as three orders of magnitude. Of the radionuclides that were detected,  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ , and  $^{125}\text{Sb}$  exhibited the largest ranges in effective diffusivities. The effective diffusivities of  $^{137}\text{Cs}$  for any given waste-form sample, were, on the other hand, relatively constant, varying by no more than a factor of five. The ranges of effective diffusivities for the Peach Bottom-2 sample were generally significantly smaller than corresponding ranges for the other two Can-Decon waste forms. The relatively large ranges of effective diffusivities exhibited by many of the radionuclides reflects the fact that radionuclide release rates generally decreased as cumulative leaching time increased. Decreases in release rates were more pronounced when longer leaching intervals were used.

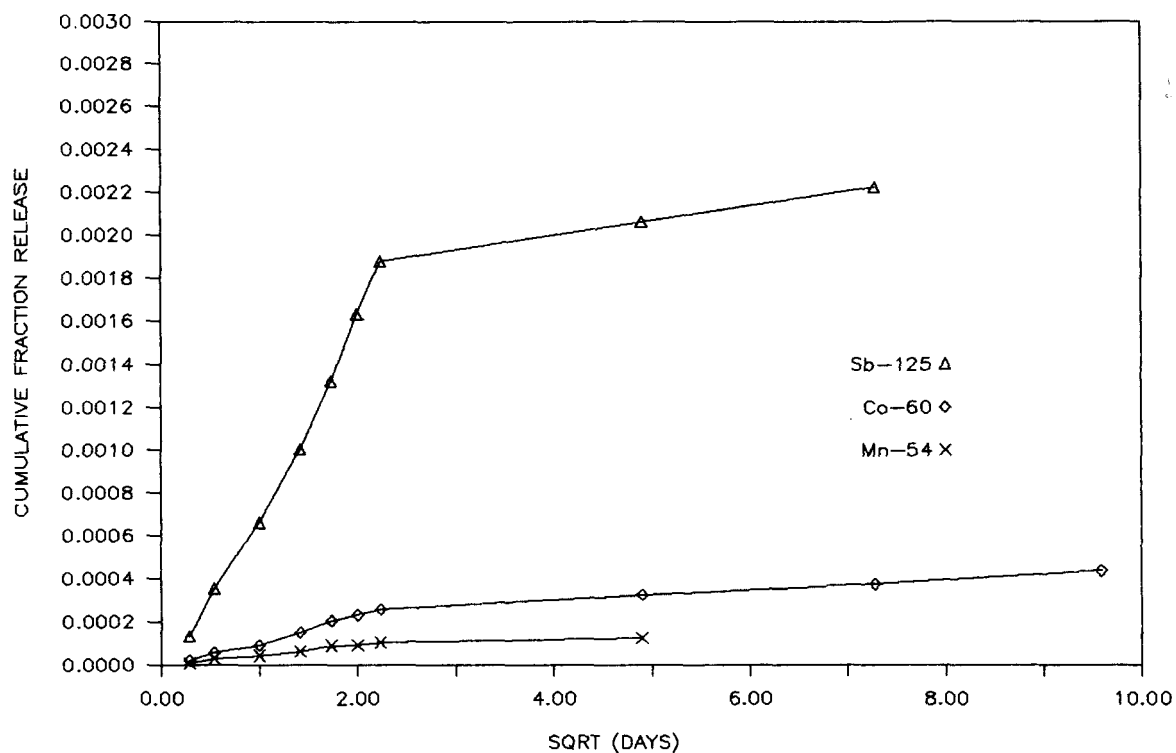
The ranges of radionuclide release rates for the Can-Decon waste forms are presented in Table 14, where they are expressed as fraction of initial inventory,  $F$ , released per second per square centimeter of sample external surface area ( $F/\text{cm}^2/\text{s}$ ). In all cases, maximum release rates occurred during the first two leaching intervals, which were, respectively, 2 and 5 h long; and minimum release rates occurred during the last three leaching intervals,

**Table 13. Radionuclide leach-test results for Can-Decon waste forms**

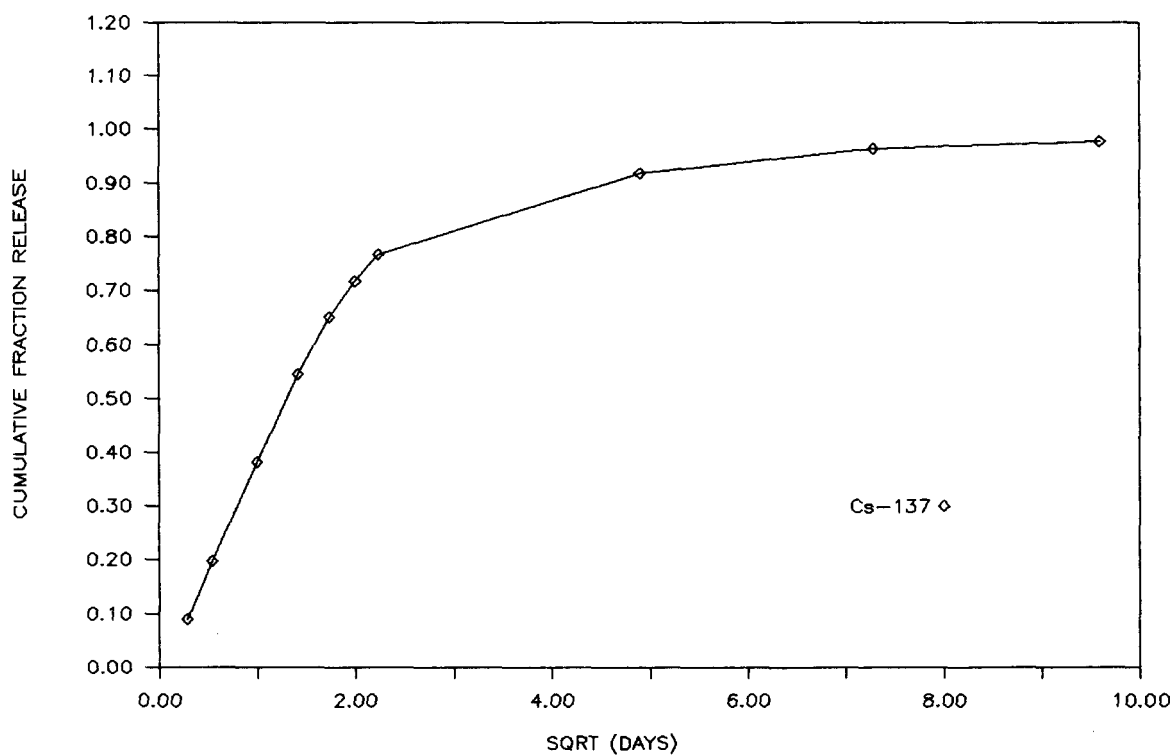
Nuclide	Maximum CFR	Range Effective Diffusivity (cm <sup>2</sup> /s)	Range Leachability Index
<u>Millstone-1 F33:</u>			
<sup>54</sup> Mn	<1.2 (–4)	3.6 (–16) to 4.0 (–14)	13.4 to 15.4
<sup>60</sup> Co	4.4 (–4)	2.4 (–15) to 1.8 (–13)	12.7 to 14.6
<sup>125</sup> Sb	2.2 (–3)	2.7 (–14) to 7.5 (–12)	11.1 to 13.6
<sup>137</sup> Cs	9.8 (–1)	2.9 (–7) to 1.5 (–6)	5.8 to 6.5
<u>Millstone-1 F201:</u>			
<sup>54</sup> Mn	<5.3 (–5)	6.0 (–17) to 5.6 (–14)	13.3 to 16.2
<sup>55</sup> Fe	— <sup>a</sup>	1.3 (–17) to 2.5 (–14)	13.6 to 16.9
<sup>60</sup> Co	1.5 (–3)	9.3 (–15) to 6.4 (–13)	12.2 to 14.0
<sup>63</sup> Ni	— <sup>a</sup>	1.6 (–12) to 3.5 (–11)	10.5 to 11.8
<sup>90</sup> Sr	— <sup>a</sup>	3.1 (–9) to 3.0 (–8)	7.5 to 8.5
<sup>125</sup> Sb	3.3 (–3)	3.6 (–14) to 1.7 (–11)	10.8 to 13.4
<sup>137</sup> Cs	9.6 (–1)	2.5 (–7) to 1.2 (–6)	5.9 to 6.6
<sup>241</sup> Pu	— <sup>a</sup>	<4.1 (–17) to <1.4 (–13)	>12.9 to >16.4
<u>Peach Bottom-2:</u>			
<sup>54</sup> Mn	<1.3 (–3)	<4.0 (–14) to 1.3 (–12)	11.9 to >13.4
<sup>60</sup> Co	1.3 (–3)	7.7 (–14) to 2.6 (–13)	12.6 to 13.1
<sup>90</sup> Sr	— <sup>a</sup>	1.6 (–10) to <2.5 (–9)	> 8.6 to 9.8
<sup>125</sup> Sb	— <sup>b</sup>	— <sup>b</sup>	— <sup>b</sup>
<sup>137</sup> Cs	3.6 (–1)	1.1 (–8) to 2.5 (–8)	7.6 to 7.9
<sup>241</sup> Pu	— <sup>a</sup>	<7.9 (–12) to <2.8 (–10)	> 9.5 to >11.1

a. Not all leachate samples were analyzed for this radionuclide.

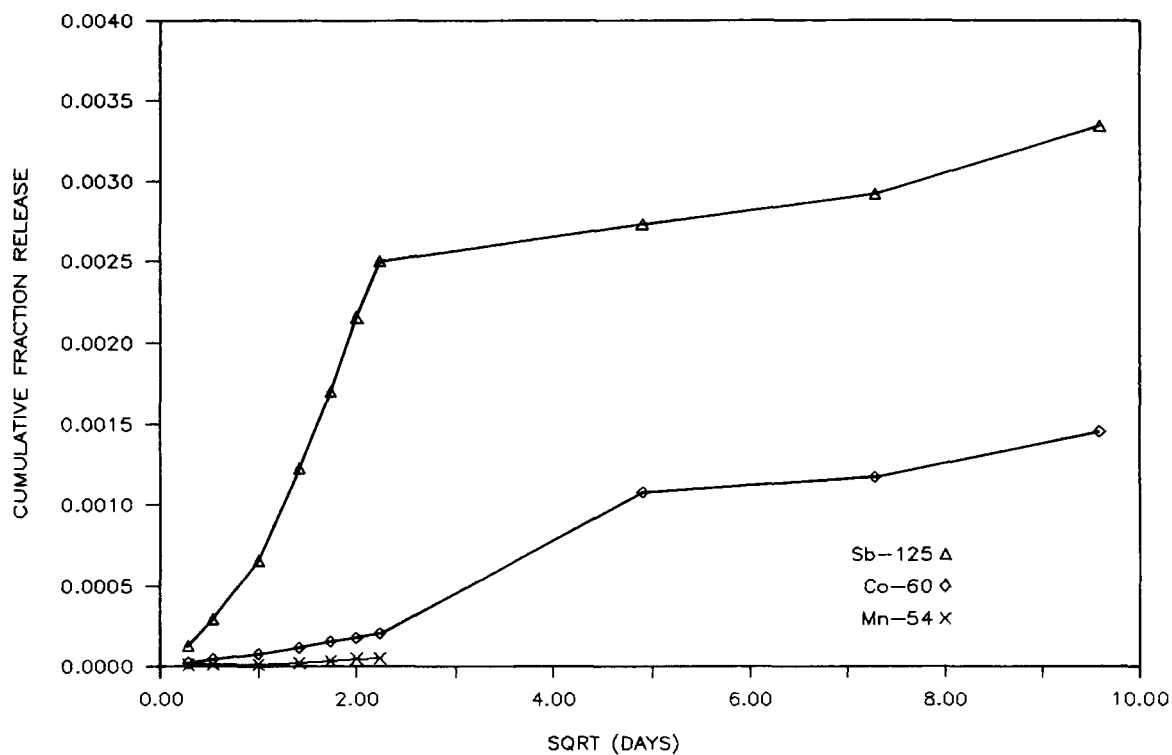
b. Concentrations in both the resin waste and the leachates were less than the detection limit.



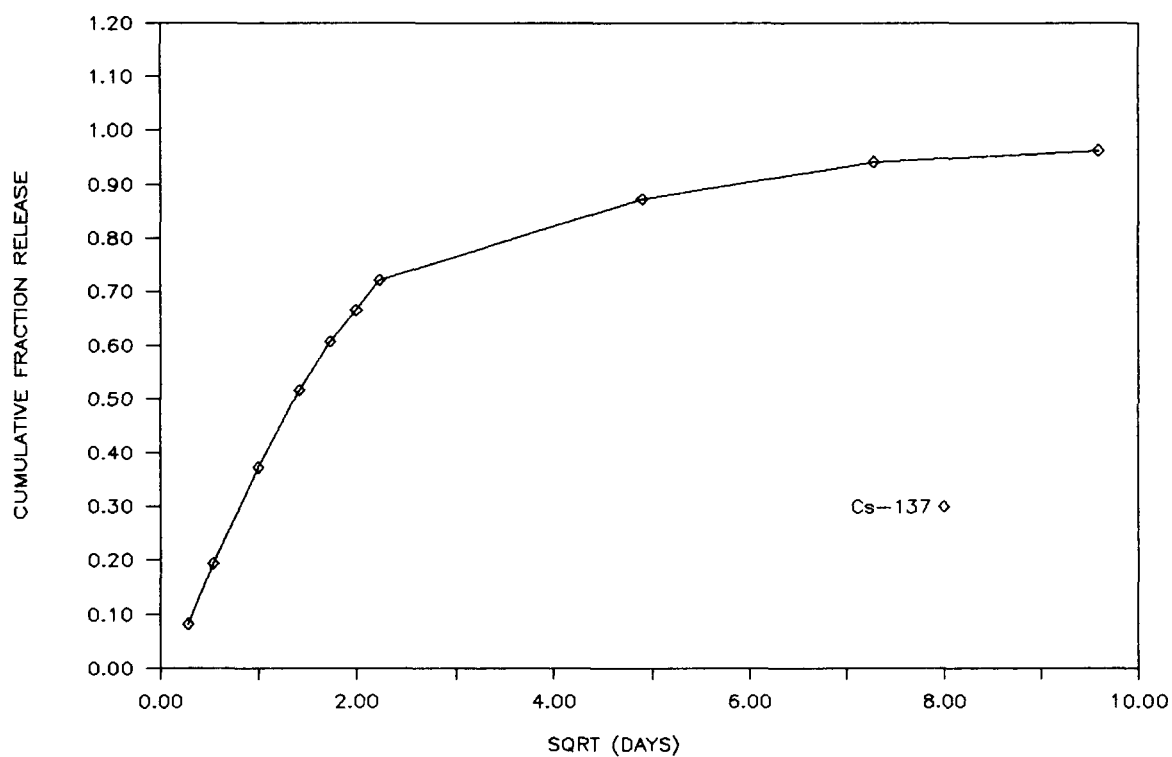
**Figure 13.** Cumulative fraction release of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ , and  $^{125}\text{Sb}$  from the Millstone-1 F33 Can-Decon waste form.



**Figure 14.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Millstone-1 F33 Can-Decon waste form.

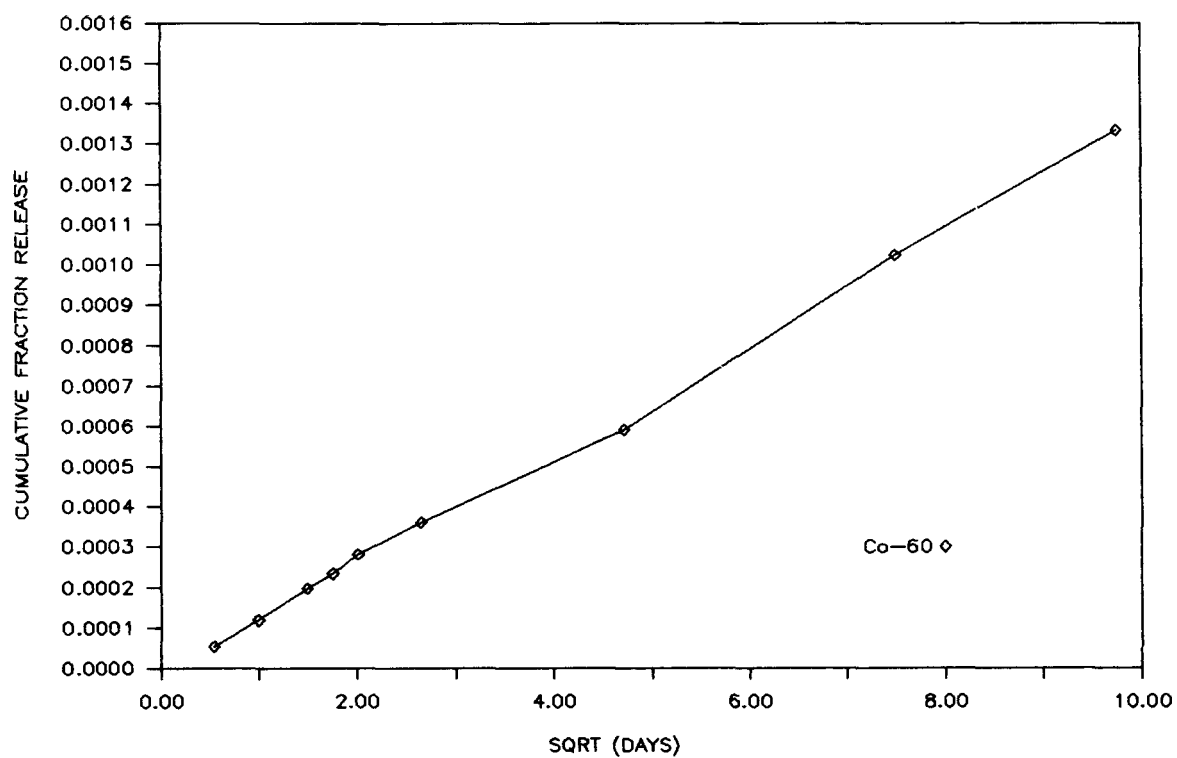


**Figure 15.** Cumulative fraction release of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ , and  $^{125}\text{Sb}$  from the Millstone-1 F201 Can-Decon waste form.

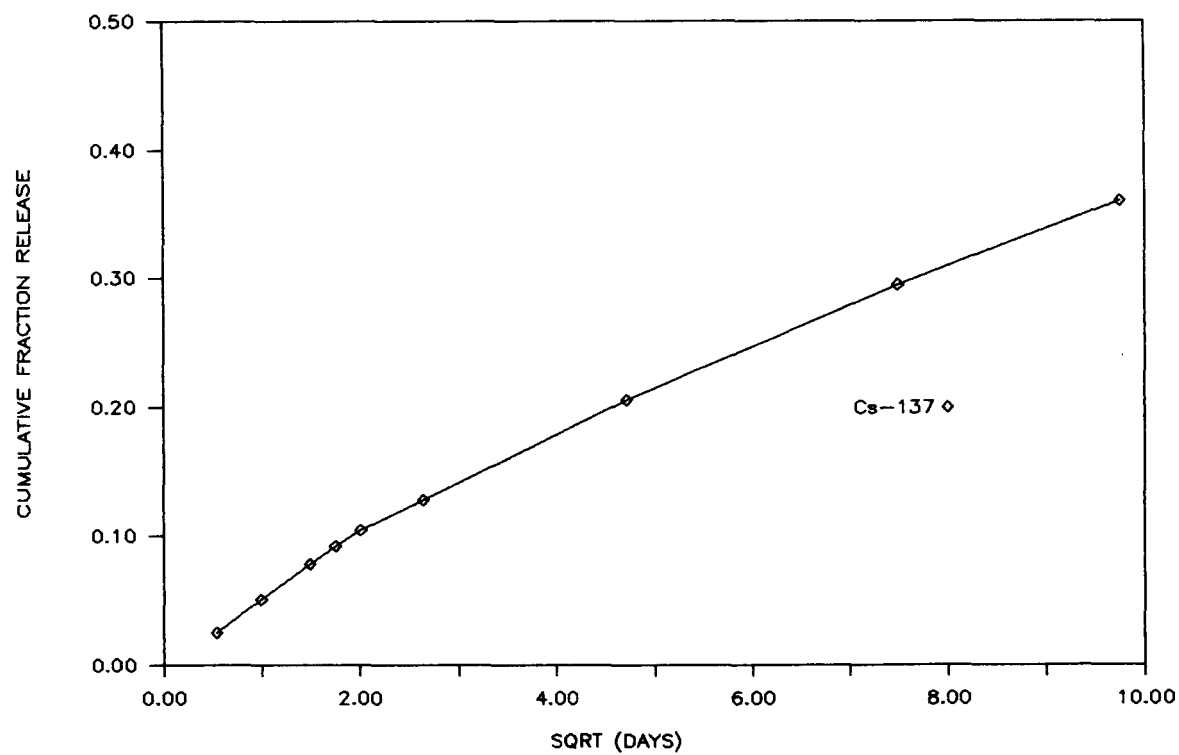


**Figure 16.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Millstone-1 F201 Can-Decon waste form.





**Figure 17.** Cumulative fraction release of  $^{60}\text{Co}$  from the Peach Bottom-2 Can-Decon waste form.



**Figure 18.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Peach Bottom-2 Can-Decon waste form.

**Table 14. Radionuclide release rates for Can-Decon waste forms (F/cm<sup>2</sup>/s)<sup>a</sup>**

Nuclide	Millstone-1 F33	Millstone-1 F201	Peach Bottom-2
<sup>54</sup> Mn	1.09 (– 11) to 1.21 (– 13)	1.12 (– 11) to 3.96 (– 14)	< 1.0 (– 11) to < 3.9 (– 13)
<sup>55</sup> Fe	— <sup>b</sup>	1.40 (– 11) to 8.90 (– 15)	— <sup>b</sup>
<sup>60</sup> Co	2.55 (– 11) to 1.71 (– 13)	2.43 (– 11) to 3.25 (– 13)	1.24 (– 11) to 5.46 (– 13)
<sup>63</sup> Ni	— <sup>b</sup>	2.32 (– 10) to 3.08 (– 12)	— <sup>b</sup>
<sup>90</sup> Sr	— <sup>b</sup>	7.98 (– 9) to 2.13 (– 10)	— <sup>b</sup>
<sup>125</sup> Sb	1.74 (– 10) to 6.07 (– 13)	1.52 (– 10) to 6.39 (– 13)	— <sup>c</sup>
<sup>137</sup> Cs	1.16 (– 7) to 3.82 (– 11)	9.93 (– 8) to 5.52 (– 11)	5.85 (– 9) to 1.15 (– 10)

a. Fraction of initial inventory released per second per cm<sup>2</sup> of sample surface area.

b. Leachates not analyzed for this radionuclide.

c. Radionuclide not detected.

which were nominally 14, 28, and 43 days long. As expected, <sup>137</sup>Cs had the highest release rate for each sample, with maximum release rates of  $1.16 \times 10^{-7}$ ,  $9.93 \times 10^{-8}$ , and  $5.85 \times 10^{-9}$  F/cm<sup>2</sup>/s for the Millstone-1 F33, Millstone-1 F201, and Peach Bottom-2 waste forms, respectively. The radionuclides exhibiting the smallest release rates were <sup>54</sup>Mn, <sup>55</sup>Fe, and <sup>60</sup>Co. The decreases in release rates observed as cumulative leaching time increased are similar to those observed for radionuclides released from cement-solidified boric acid and sulfate waste forms.<sup>27</sup>

The CFR data for <sup>54</sup>Mn, <sup>60</sup>Co, <sup>125</sup>Sb, and <sup>137</sup>Cs for the Millstone-1 F33 sample are plotted in Figures 13 and 14, and similar results for the Millstone-1 F201 sample are plotted in Figures 15 and 16. When plotted against the square root of elapsed leaching time, the CFR values of these four radionuclides are approximately linear over two different time regions. Through the first 5 days of leaching, CFR values generally increased much more rapidly than they did during later leaching intervals. A change in slope indicates a change in the predominant release mechanism. The predominant release mechanism during the second period may be present during the first period, but may not noticeably affect releases. The initial fast releases are generally classified as surface effect releases,

which do not involve diffusion through the solid over any significant distance. After 5 days, the measured CFR values of all radionuclides were approximately proportional to the square root of elapsed leaching time, indicating that the releases were controlled by diffusion.

As shown in Table 13 and Figures 14 and 16, essentially the entire inventory of <sup>137</sup>Cs was released from the Millstone-1 samples after 3 months of continuous leaching. In the case of the F33 sample, 77% of its initial inventory of <sup>137</sup>Cs was released from the sample during the first 5 days of leaching. The fractional release of <sup>137</sup>Cs from the F201 sample was similar, being 72% after 5 days. Figures 14 and 16 indicate that the releases of <sup>137</sup>Cs from the two Millstone-1 samples were very similar over the 3-month-long leach-test. Although the cumulative releases of <sup>54</sup>Mn, <sup>60</sup>Co, and <sup>125</sup>Sb from the Millstone-1 samples were quite small compared to the <sup>137</sup>Cs releases, the general behavior of their releases closely duplicated the trends in the releases of <sup>137</sup>Cs. Releases occurring during the initial fast release period (0 to 5 days) accounted for 86% of the total quantity of <sup>125</sup>Sb released from the F33 sample at the end of 53 days of leaching and 76% of the total quantity of <sup>125</sup>Sb released from the F201 sample at the end of 92 days of leaching. Similarly, 59% of the total quantity of <sup>60</sup>Co released from the

F33 sample was released during the first 5 days of leaching. Apparently, the observed deteriorations of the Millstone-1 samples were sufficient to cause significant surface effect releases. As shown in Table 13, the maximum CFRs of  $^{125}\text{Sb}$  for the F33 and F201 samples were  $4.4 \times 10^{-4}$  and  $3.3 \times 10^{-3}$ , respectively. Cumulative releases of  $^{60}\text{Co}$  and  $^{54}\text{Mn}$  were factors of between 2 and 60 lower than the total releases of  $^{125}\text{Sb}$ .

The CFR data for the Peach Bottom-2 sample are plotted in Figures 17 and 18. These data are also approximately linear over two different time regions, but the slopes of the CFR data in the two regions do not differ as much as the slopes of the Millstone-1 CFR data. As was previously mentioned, the Peach Bottom-2 sample showed no signs of deterioration during leaching. Figures 17 and 18 indicate that the surface effect release of  $^{137}\text{Cs}$  from the Peach Bottom-2 waste form was less than 5% of the  $^{137}\text{Cs}$  inventory and that of  $^{60}\text{Co}$  was essentially zero, indicating that diffusion was the dominant release mechanism throughout the leach test. The transition between the two linear regions occurs at 5 days, as was the case with the Millstone-1 CFR data. While nearly the entire inventory of  $^{137}\text{Cs}$  was released from the Millstone-1 samples, only 36% of the  $^{137}\text{Cs}$  initially in the Peach Bottom-2 sample was released after 95 days of continuous leaching. The CFR of  $^{60}\text{Co}$  for the Peach Bottom-2 sample was, on the other hand, about the same as that for the Millstone-1 F201 sample, the values being  $1.3 \times 10^{-3}$  and  $1.4 \times 10^{-3}$ , respectively.

**Citrox Waste Forms.** Two Citrox waste-form samples were leach-tested using the ANS 16.1 procedure. The samples were collected from different liners during the solidification of decontamination resin wastes at Cooper Station. One of the samples contained mixed-bed resins consisting of 10% Purolite C-100-H cation exchange resin and 90% Purolite A-600 anion exchange resin. Sodium hydroxide was added to the resin to adjust the pH prior to solidification. The second waste-form sample contained only Purolite NRW-37 cation resin. Lime was added to the cation resin prior to solidification. The wet resin loadings of the samples, which were measured at the time of solidification, were 72 and 77 vol.% for the mixed-bed and cation resin waste forms, respectively. Other information regarding the physical characteristics of the samples may be found in Table 9.

Prior to the initiation of leaching, the surfaces of the mixed-bed resin waste form sample were

smooth and exhibited very little surface cracking. The specimen appeared sound and well cured. Photographs of the cation resin waste-form sample are shown in Figure 3. As was previously described, a relatively large crack traversed one side of the cation resin waste-form specimen just below its mid-plane and several small voids pitted the surface above the mid-plane.

During the initial 30-s prerinse of the mixed-bed resin waste form, some signs of deterioration were evident. Small particles of concrete and resin were washed from the sample, and cracks formed along the circumferences of the ends of the sample. When the sample was placed in leachant to commence the first leaching, it began to crumble within 2 min after it was initially immersed. After 5 min, large cracks appeared on all surfaces of the sample. During the following 20 min, the sample continued to crumble until only about 10% of the original sample was intact. After 1 h, essentially all of the sample had decomposed into loose granular debris and had fallen through the sample holding net to the bottom of the leaching container.

The Citrox cation resin waste-form sample suffered a similar fate, but its decomposition occurred over a longer time. After it was immersed for 6 min during the first leaching interval, the sample began shedding small flakes from its ends and the crack that traversed the side of the sample prior to leaching grew wider and deeper. Within 9 min, new cracks appeared on the surfaces of the sample and the amount of surface flaking increased. After 8 h, the sample had completely decomposed into loose debris that had fallen to the bottom of the leaching container.

When the Citrox samples disintegrated during initial phases of leach-testing, it was decided to continue leaching the debris in order to determine what effect decomposition would have on the releases of radionuclides they contained. Due to the breakup of the samples, the standard leachate changeout procedure had to be modified. The granular debris in the bottoms of the original leaching containers could not easily be transferred to new leaching containers. Under the circumstances, it was decided to leave the samples in their original containers and decant the leachates at the end of each leaching interval. Leachant was added to the same container to continue leaching.

Because the Citrox waste-form samples disintegrated during initial phases of leach-testing, several of the requirements of the ANS 16.1 procedure were violated. Consequently, the following discussion will be limited to radionuclide release rates and

cumulative release fractions. Average leachability indexes for the Citrox waste forms are presented later in this report.

The ranges of radionuclide release rates for the Citrox waste forms are presented in Table 15, where they are expressed as fraction of initial inventory released per second (F/s). The release rates were not normalized by the surface areas of the samples, since the surface areas of the samples following disintegration were not well defined. Release rates for individual leaching intervals are tabulated in Appendix E. Because the leachate samples often contained free-floating resin beads and particulates, they were filtered through 0.45- $\mu\text{m}$  filters prior to being analyzed for gamma-emitting radionuclides. However, the samples that were analyzed for concentrations of beta-emitting radionuclides were unfiltered. The release rate data presented in Table 15 are segregated according to whether or not the leachate samples were filtered prior to analysis.

Neither  $^{125}\text{Sb}$  nor  $^{137}\text{Cs}$  were detected in the Citrox cation resin waste; their concentrations were reported in Table 10 as  $<2 \times 10^{-4} \mu\text{Ci}/\text{cm}^3$  wet resin. Consequently, their release rates are reported as lower-limit values in Table 15. The concentrations of both of these radionuclides in the mixed-bed resin waste were determined. The

concentration of  $^{137}\text{Cs}$  in the mixed-bed resin was comparable to the  $^{137}\text{Cs}$  upper-limit concentration determined for the cation resin. However, the  $^{125}\text{Sb}$  concentration in the mixed-bed resin waste was about a factor of 200 higher than its upper-limit concentration in the cation resin. Common valence states of  $^{125}\text{Sb}$  are +3, +5, and -3. The fact that  $^{125}\text{Sb}$  was retained exclusively on the mixed-bed resin suggests that it existed in the decontamination solution as an anionic species rather than as a cationic species.

As was the case with the Can-Decon waste forms, maximum release rates occurred during the first or second leaching interval and minimum release rates were associated with the last three leaching intervals. For the mixed-bed waste form,  $^{137}\text{Cs}$  exhibited the highest release rate, with a maximum value of about  $1.6 \times 10^{-5}$  F/s. The maximum rate of release of  $^{137}\text{Cs}$  from the cation resin sample was  $>2.3 \times 10^{-6}$  F/s. For most leaching intervals, the average rate of release of  $^{60}\text{Co}$  from the mixed-bed resin sample was a factor of two to three higher than from the cation resin sample. For the mixed-bed sample,  $^{60}\text{Co}$  release rates ranged from about  $5.2 \times 10^{-8}$  to  $2.9 \times 10^{-12}$  F/s, while similar values for the cation resin sample ranged from about  $1.6 \times 10^{-8}$  to  $4.0 \times 10^{-12}$  F/s. In the

**Table 15. Radionuclide release rates for Citrox waste forms**

Nuclide	Release Rate <sup>a</sup> (F/s)	
	Cooper Mixed Bed	Cooper Cation
<u>Filtered:</u>		
$^{54}\text{Mn}$	6.75 (-10) <sup>b</sup>	1.43 (-8) to 3.58 (-11)
$^{60}\text{Co}$	5.19 (-8) to 2.93 (-12)	1.55 (-8) to 3.99 (-12)
$^{125}\text{Sb}$	8.94 (-8) to 4.34 (-11)	$>9.95$ (-6) to $>3.73$ (-7) <sup>c</sup>
$^{137}\text{Cs}$	1.63 (-5) to 4.65 (-9)	$>2.27$ (-6) to $>4.63$ (-8) <sup>c</sup>
<u>Unfiltered:</u>		
$^{55}\text{Fe}$	6.87 (-8) to 3.50 (-10)	8.10 (-6) to 6.51 (-11)
$^{63}\text{Ni}$	1.54 (-9) to 4.64 (-12)	1.60 (-7) to 6.82 (-13)
$^{90}\text{Sr}$	1.47 (-7) to 2.66 (-10)	1.27 (-4) to 9.42 (-10)

a. Fraction of initial inventory released per second.

b. Release rate determined for only one leach interval.

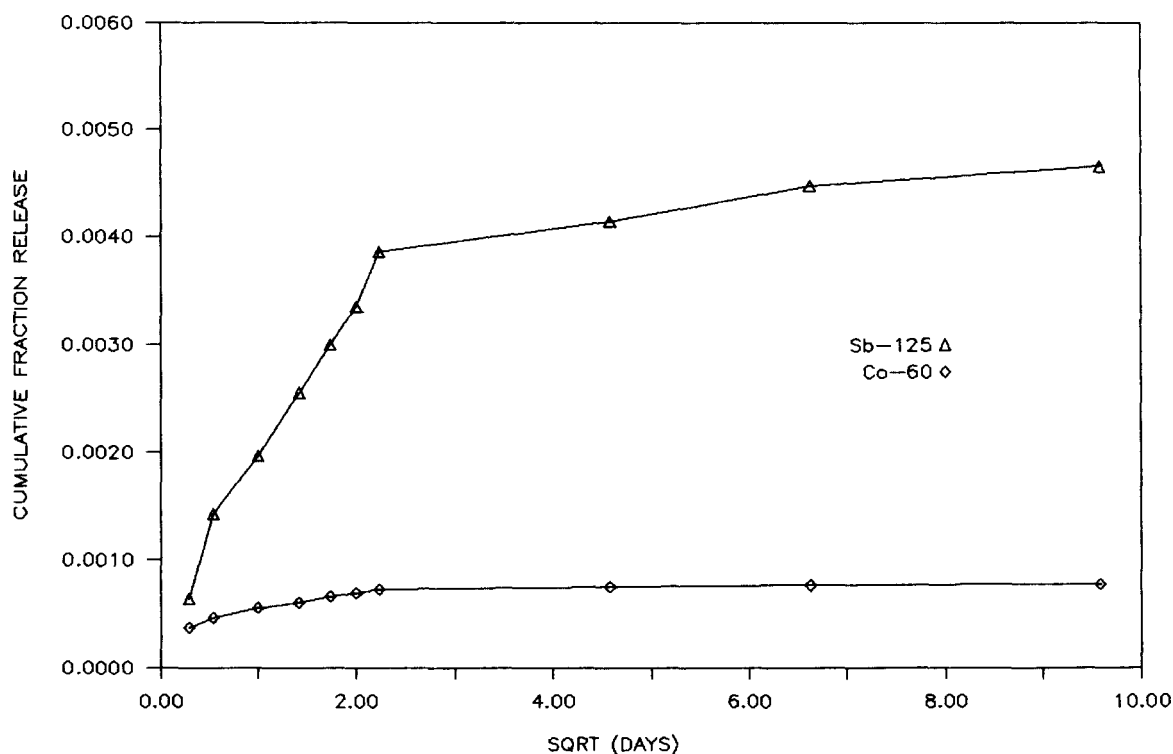
c. Concentration in resin was less than detection limit.

case of the Cooper cation resin waste form, maximum release rates of  $^{55}\text{Fe}$  and  $^{90}\text{Sr}$  were actually higher than the maximum release rate determined for  $^{137}\text{Cs}$ , indicating that the unfiltered leachate samples which were analyzed for beta-emitting radionuclides contained significant quantities of particulate matter.

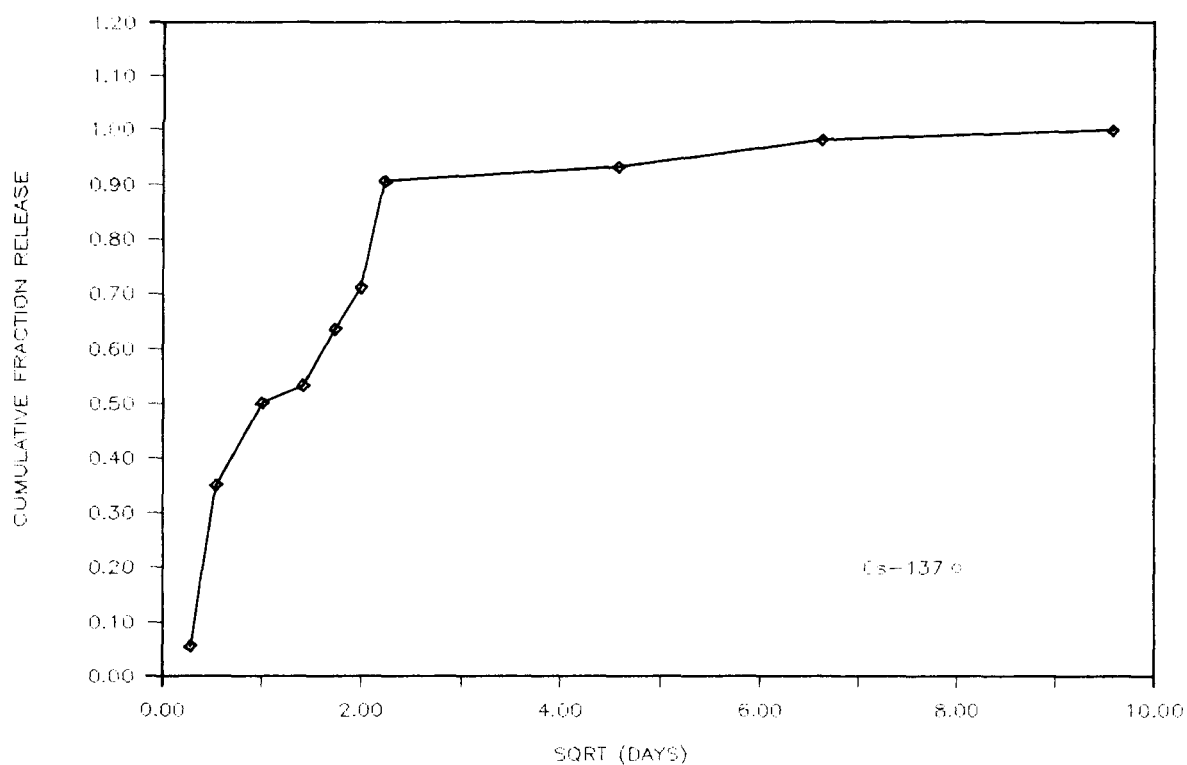
The CFR data for  $^{60}\text{Co}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  for the Cooper mixed-bed resin waste form are plotted in Figures 19 and 20, and similar results for  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  for the Cooper cation resin waste form are shown in Figures 21 and 22. In the case of the mixed-bed resin sample, the CFR profiles were very similar to the CFR profiles determined for the Millstone-1 Can-Decon samples. Through the first 5 days of leaching, CFR values increased much more rapidly than they did during the last three leaching periods. After 5 days, the CFR data were approximately linear as a function of the square root of elapsed leaching time, indicating that diffusion became the dominant release mechanism after the particulate debris had been washed (i.e., leached) seven times. About 48% of the total quantity of  $^{60}\text{Co}$  leached from the mixed-bed resin sample was lost from the sample during the first 2 h of leaching. That percentage increases to 71% by the end of the first day and to 93% by the end of the

end of the fifth day. The CFR profile of  $^{60}\text{Co}$  for the cation resin sample indicates a more abrupt change in release rate. Approximately 81% of the total amount of  $^{60}\text{Co}$  leached from the cation resin sample was lost from the sample after 7 h of leaching. In contrast to the mixed-bed resin sample, the cumulative fraction of  $^{60}\text{Co}$  released from the cation resin sample during the first leaching period was only about  $2.6 \times 10^{-6}$ . Apparently the cation resin waste form began to seriously decompose during the second leaching interval. After 7 h, the CFR data for the cation resin sample were approximately proportional to the square root of elapsed leaching time, indicating that surface effect releases were minimal after the second leaching interval with diffusion becoming the controlling release mechanism. After 92 days of leaching, the cumulative fractions of  $^{60}\text{Co}$  released from the mixed-bed and cation resin waste forms were  $7.8 \times 10^{-4}$  and  $3.5 \times 10^{-4}$ , respectively. These releases were both below the  $^{60}\text{Co}$  CFR determined for the Peach Bottom-2 waste form, a sample which maintained its physical integrity throughout 3 months of leaching.

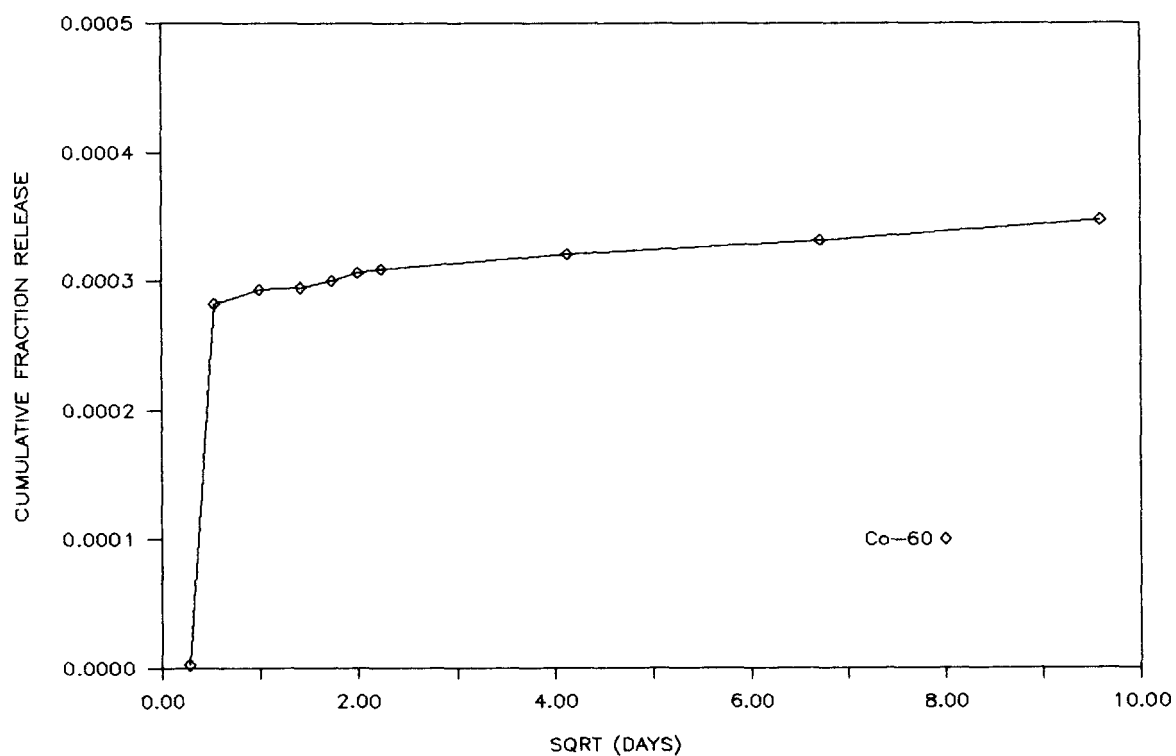
In the case of the mixed-bed resin sample, the behavior of the CFR data for  $^{125}\text{Sb}$  and  $^{137}\text{Cs}$  was similar to that of  $^{60}\text{Co}$ . About 83% of the total



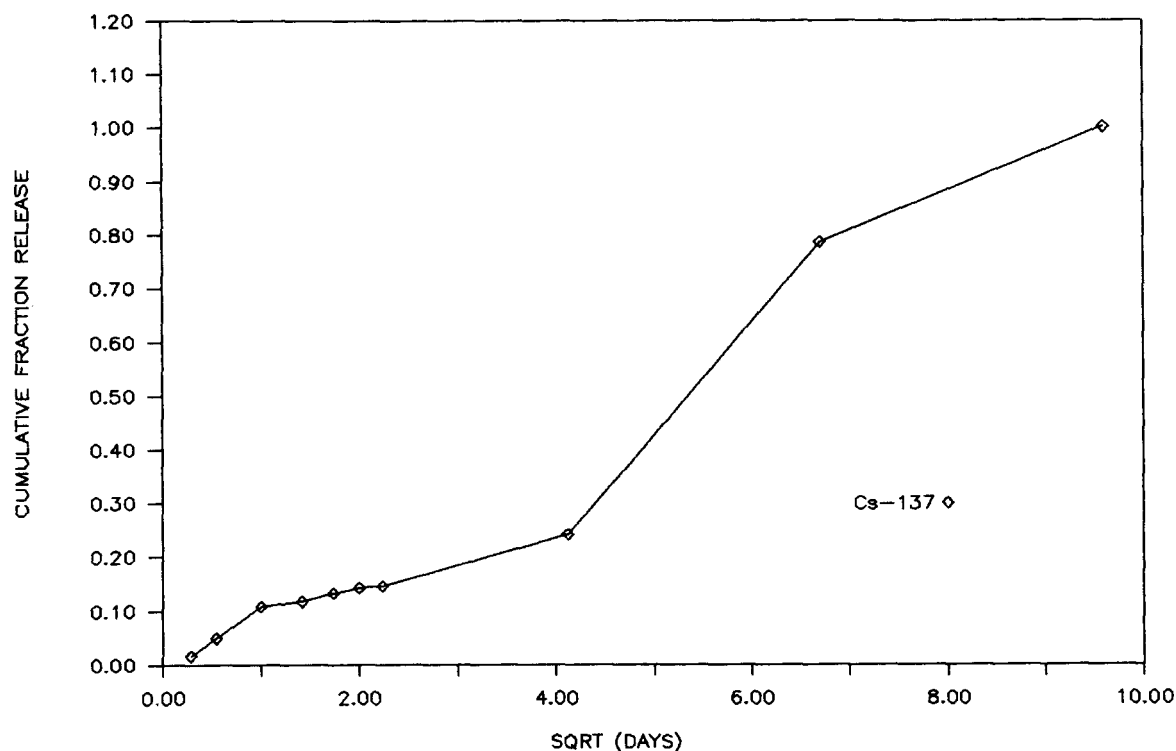
**Figure 19.** Cumulative fraction release of  $^{60}\text{Co}$  and  $^{125}\text{Sb}$  from the Cooper mixed-bed resin Citrox waste form.



**Figure 20.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Cooper mixed-bed resin Citrox waste form.



**Figure 21.** Cumulative fraction release of  $^{60}\text{Co}$  from the Cooper cation resin Citrox waste form.



**Figure 22.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Cooper cation resin CitroX waste form.

quantity of  $^{125}\text{Sb}$  and 91% of the total amount of  $^{137}\text{Cs}$  leached from the sample were leached from the sample after 5 days. The cumulative fraction of  $^{125}\text{Sb}$  released from the mixed-bed sample was  $4.7 \times 10^{-3}$ , a value comparable to the quantities of  $^{125}\text{Sb}$  released from the Millstone-1 samples. As was the case with the Millstone-1 samples, the entire inventory of  $^{137}\text{Cs}$  in the Cooper waste-form samples was leached from the samples. While 91% of the  $^{137}\text{Cs}$  inventory in the mixed-bed resin waste form was lost within 5 days, only 24% of the inventory in the cation resin sample had leached from the cation resin waste form after 17 days of leaching. That percentage increased to 79% by the end of 45 days. The rapid rise in the  $^{137}\text{Cs}$  CFR between 17 and 45 days probably coincides with the breakdown of sample debris into smaller fragments which would have resulted in new surfaces being exposed to the leachate.

**Dow NS-1 Waste Form.** One Dow NS-1 waste form sample was leach-tested. The waste-form sample was prepared by Chem-Nuclear Services, Inc., using a sample of the Dow NS-1 decontamination resin waste and the same mixing parameters used for the full-scale solidification at the Pilgrim

station. The types of resins that were solidified are not known. Following curing for 654 days, the surfaces of the waste form were smooth and exhibited very few surface imperfections. From its general appearance, it was considered to be one of the better waste forms that had been collected. The dimensions and other physical characteristics of the sample are provided in Table 9. During leach-testing, no noticeable deterioration was observed. The only significant observation was that after the sample was first immersed in leachant, it effervesced more than any other sample that was tested. The effervescence continued during the 30-s-long prerinse and during the first minute of leaching.

A summary of the radionuclide leach-test results for the Dow NS-1 waste form sample is presented in Table 16. The cumulative quantity of each radionuclide released during leaching, expressed as fraction of initial inventory, is presented in Table 16, along with the ranges of the measured effective diffusivities and leachability indexes. The cumulative fractions of  $^{54}\text{Mn}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  released from the Pilgrim waste form are given as upper-limit values. Manganese-54 and  $^{125}\text{Sb}$  were detected in only one leachate sample, and  $^{137}\text{Cs}$  was detected in only four of the nine leachate samples generated during

**Table 16. Radionuclide leach-test results for Pilgrim Dow NS-1 waste form**

Nuclide	Maximum CFR	Range Effective Diffusivity (cm <sup>2</sup> /s)	Range Leachability Index
<sup>54</sup> Mn	<2.0 (–3)	6.2 (–14) to <8.0 (–11)	>10.1 to 13.2
<sup>55</sup> Fe	— <sup>a</sup>	3.3 (–18) to 1.2 (–15)	14.9 to 17.5
<sup>60</sup> Co	5.1 (–2)	9.8 (–11) to 2.2 (–9)	8.7 to 10.0
<sup>63</sup> Ni	— <sup>a</sup>	2.2 (–8) to 3.6 (–7)	6.5 to 7.7
<sup>90</sup> Sr	— <sup>a</sup>	3.3 (–10) to 6.7 (–8)	7.2 to 9.5
<sup>125</sup> Sb	<7.1 (–2)	<1.5 (–9) to <9.0 (–8)	> 7.1 to 8.5
<sup>137</sup> Cs	<4.0 (–3)	4.3 (–14) to <1.8 (–10)	> 10.4 to 13.4
<sup>241</sup> Pu	— <sup>a</sup>	<3.2 (–14) to <2.9 (–12)	> 11.5 to > 13.5

<sup>a</sup>. Not all leachate samples were analyzed for this radionuclide.

leach testing. Cumulative release fractions for <sup>55</sup>Fe, <sup>63</sup>Ni, <sup>90</sup>Sr, and <sup>241</sup>Pu are not presented because only five of the nine leachate samples were analyzed for these radionuclides. Average radionuclide release rates during individual leaching intervals are presented in Appendix E, and radionuclide concentrations in the leachate samples are tabulated in Appendix G.

The results presented in Table 16 show that, as was the case with the Can-Decon waste forms, the effective diffusivities of individual radionuclides spanned relatively large ranges. Of the radionuclides that were consistently detected, <sup>55</sup>Fe and <sup>90</sup>Sr exhibited the largest ranges in effective diffusivities, the differences between the high and low values being greater than two orders of magnitude. The effective diffusivities of <sup>60</sup>Co and <sup>63</sup>Ni showed the smallest ranges, the high and low values varying by no more than factors of 22 and 16, respectively. The relatively large ranges in effective diffusivities exhibited by some of the radionuclides reflect the fact that their rates of release changed substantially over the duration of the leach test. As was typical of the release rates determined for the Can-Decon and Citrox waste forms, the release rates of the radionuclides contained in the Dow NS-1 sample uniformly decreased as cumulative leaching time increased. The smallest average release rates occurred during

the last two leaching periods, which were 20 and 23 days long.

Comparing the leaching results in Table 16 with those in Table 13, significant differences are obvious. The effective diffusivities of <sup>137</sup>Cs determined for the Dow NS-1 sample were five orders of magnitude or more lower than the effective diffusivities of <sup>137</sup>Cs for the Millstone-1 and Peach Bottom-2 Can-Decon samples. The leachability index of <sup>137</sup>Cs ranged from >10.4 to 13.4 for the Dow NS-1 sample, whereas similar results for the three Can-Decon samples collectively ranged from 5.8 to 7.9. In addition, the leachability indexes of <sup>60</sup>Co, <sup>63</sup>Ni, and <sup>125</sup>Sb for the Dow NS-1 waste form were considerably lower than their corresponding values for the Can-Decon samples. The leachability index of <sup>60</sup>Co ranged from only 8.7 to 10.0 for the Dow NS-1 sample, compared to a typical range of 12 to 14 for the three Can-Decon samples. Similarly, the leachability index of <sup>63</sup>Ni for the Dow NS-1 sample ranged from only 6.5 to 7.7, whereas similar values for the Millstone-1 F201 sample ranged from 10.5 to 11.8. For the Millstone-1 samples, the leachability index of <sup>125</sup>Sb ranged from 10.8 to 13.6, but similar results for the Dow NS-1 sample were only >7.1 to 8.5. As was discussed previously, resin wastes can be treated with additives that help reduce the leachability of soluble radionuclides



such as  $^{137}\text{Cs}$ . Additives such as zeolites and certain clays and silica can reduce cesium leachability by factors of a hundred or more. It is not known what additives, if any, were used during the solidification of the Dow NS-1 waste-form specimen. The results suggest that additives were used that reduced the leachability of  $^{137}\text{Cs}$  at the expense of increasing the leachability of  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ , and  $^{125}\text{Sb}$ .

The ranges of radionuclide release rates for the Pilgrim Dow NS-1 waste-form sample are presented in Table 17, where they are expressed as fraction of initial inventory released per second per square centimeter of sample external surface area ( $\text{F}/\text{cm}^2/\text{s}$ ). In almost all cases, maximum average release rates occurred during the first, 2-hour-long leaching period and minimum release rates occurred during the last two leaching intervals, each of which was about 3 weeks long. Consistent with the results previously discussed, the release rates of  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ , and  $^{125}\text{Sb}$  were higher than the release rates determined for  $^{137}\text{Cs}$ . The release rates of  $^{90}\text{Sr}$  were also higher than those for  $^{137}\text{Cs}$ . The maximum release rates of  $^{63}\text{Ni}$  and  $^{90}\text{Sr}$  were both about  $2 \times 10^{-8} \text{ F}/\text{cm}^2/\text{s}$ , while similar results for  $^{60}\text{Co}$  and  $^{125}\text{Sb}$  were about  $2 \times 10^{-9}$  and  $< 8 \times 10^{-9} \text{ F}/\text{cm}^2/\text{s}$ , respectively. Release rates determined for  $^{137}\text{Cs}$  ranged from  $< 5 \times 10^{-10}$  to  $6.7 \times 10^{-13}$

$\text{F}/\text{cm}^2/\text{s}$ . The radionuclide exhibiting the lowest release rates was  $^{55}\text{Fe}$ ; its release rates ranged from about  $2.8 \times 10^{-12}$  to  $5.9 \times 10^{-15} \text{ F}/\text{cm}^2/\text{s}$ .

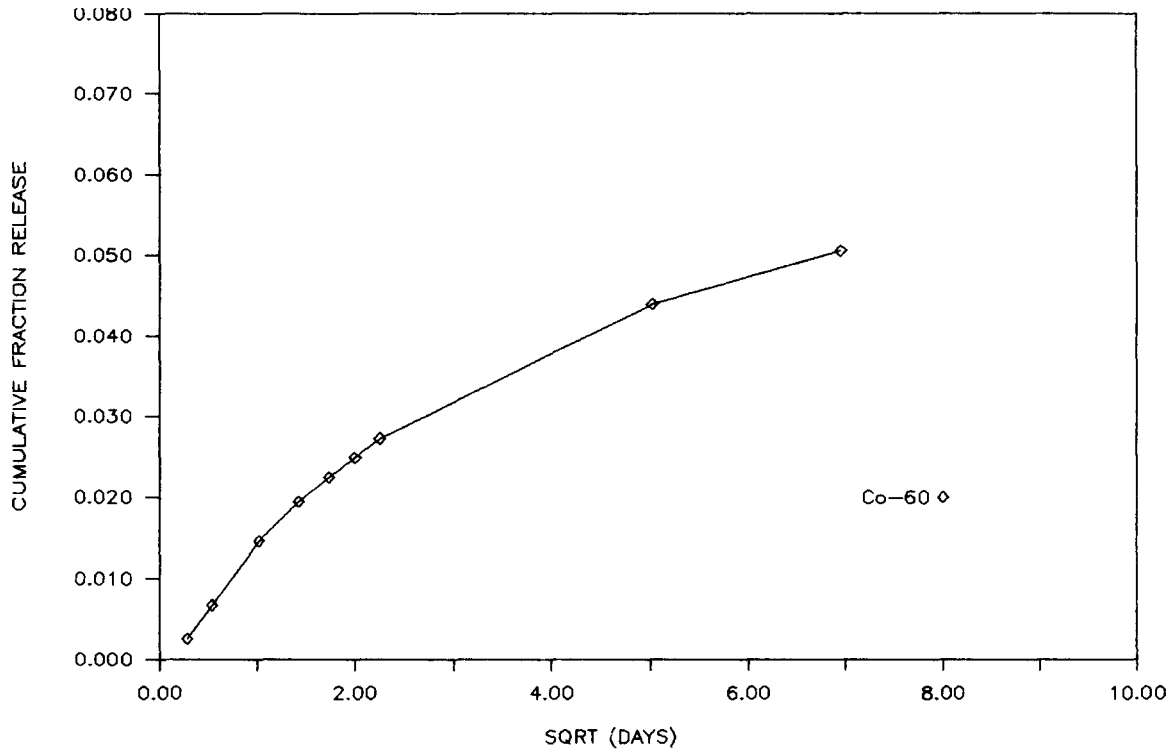
The CFR data for  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  for the Dow NS-1 waste form are plotted against the square root of elapsed leaching time in Figures 23 and 24, respectively. Results are plotted for nine leaching intervals, since only nine leachates were obtained during the leach-testing of the sample. As shown in Table 16 and Figure 23, the total quantity of  $^{60}\text{Co}$  released from the Dow NS-1 waste form after 48 days of leaching was about 5% of the initial inventory. Similar results for the Can-Decon and Citrox waste forms indicated  $^{60}\text{Co}$  releases, after 92 days of continuous leaching, that ranged from a low of 0.035% for the Cooper cation resin waste form to a high of 0.15% for the Millstone-1 F201 waste form. About 54% of the total quantity of  $^{60}\text{Co}$  released from the Dow NS-1 waste form was leached from the sample during the first 5 days of leaching. After this period of relatively rapid release, the rate of release of  $^{60}\text{Co}$  dropped substantially. During the 25- to 48-day leaching interval, the release rate was only one-seventh of its value during the 4- to 5-day leaching interval. The curvature of the  $^{60}\text{Co}$  graph through 25 days indicates that more than one release mechanism was operative through the eighth leaching interval.

The CFR results shown in Table 16 and Figure 24 indicate that less than 0.4% of the initial inventory of  $^{137}\text{Cs}$  in the Dow NS-1 waste form was leached from the sample after 48 days of leaching. Similar results for the Can-Decon and Citrox waste forms indicate total depletion of  $^{137}\text{Cs}$  inventories, except in the case of the Peach Bottom-2 waste form, which lost about 36% of its  $^{137}\text{Cs}$  inventory during 95 days of leaching. When initially analyzed, the concentrations of  $^{137}\text{Cs}$  in the first five Pilgrim waste-form leachates were less than detection limits, in part due to the Compton distribution of gamma rays emitted by  $^{60}\text{Co}$ . To improve detection sensitivity, samples of the second, third, and seventh leachates were subjected to radiochemical treatment to separate the  $^{137}\text{Cs}$  from the  $^{60}\text{Co}$ , and the  $^{137}\text{Cs}$  fractions were analyzed by gamma ray spectrometry. The method improved detection sensitivity by about a factor of ten. The fourth and fifth leachate samples were not analyzed using the separation technique, and the analyses yielded relatively high upper-limit concentrations. Therefore, it is possible that the actual  $^{137}\text{Cs}$  release profile more closely resembles a delta function, with the first significant release occurring during the sixth leaching interval.

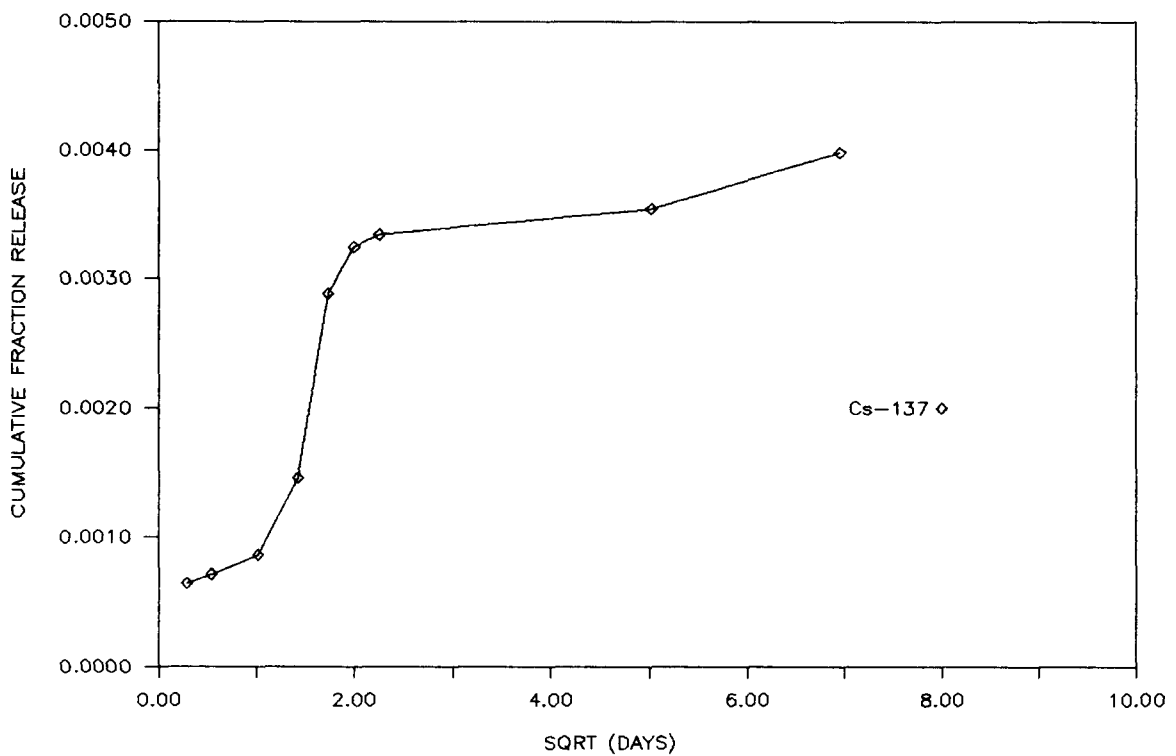
**Table 17. Radionuclide release rates for Pilgrim Dow NS-1 waste form**

Nuclide	Release Rate ( $\text{F}/\text{cm}^2/\text{s}$ )
$^{54}\text{Mn}$	$< 1.02 (-10)$ to $1.36 (-12)$
$^{55}\text{Fe}$	$2.84 (-12)$ to $5.88 (-15)$
$^{60}\text{Co}$	$2.05 (-9)$ to $1.93 (-11)$
$^{63}\text{Ni}$	$2.11 (-8)$ to $4.75 (-10)$
$^{90}\text{Sr}$	$2.10 (-8)$ to $5.83 (-11)$
$^{125}\text{Sb}$	$< 8.47 (-9)$ to $< 5.78 (-10)$
$^{137}\text{Cs}$	$< 5.16 (-10)$ to $6.68 (-13)$
$^{241}\text{Pu}$	$< 1.38 (-10)$ to $< 5.72 (-13)$

a. Fraction of initial inventory released per second per  $\text{cm}^2$  of sample surface area.



**Figure 23.** Cumulative fraction release of  $^{60}\text{Co}$  from the Pilgrim Dow NS-1 waste form.



**Figure 24.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Pilgrim Dow NS-1 waste form.

**LOMI Waste Form.** One LOMI decontamination resin waste form was leach-tested. The sample was collected from a solidification liner during the solidification of decontamination resin wastes at Indian Point-3 station. The sample contained Ionac A-365 anion and Rohm and Haas IRN-77 cation exchange resins, which were used to process the spent LOMI decontamination solution, and cation exchange resins from the plant radwaste system. Resins from the radwaste system constituted about 34 vol.% of the resins that were solidified. The wet resin loading of the sample was estimated to be 70 vol.% based on information provided by Chem-Nuclear Services, Inc. The physical parameters of the waste form following 539 days of curing are given in Table 9.

Photographs of the Indian Point-3 LOMI waste form prior to the start of leach-testing are shown in Figure 4. The surface of one end of the specimen was not perpendicular to the specimen's longitudinal axis, and a region of the surface on one side of the sample was rough and pitted with small voids. However, as shown in Figure 4, the majority of the surface was smooth and unpitted. After the sample was immersed in leachant, it effervesced to a degree typical of many of the waste-form samples. No significant deterioration of the sample was observed during 3 months of leach-testing. A few very small pieces of concrete flaked off the circumferences of the ends of the sample, but surface cracking or crumbling were not observed.

A summary of the radionuclide leach-test results for the LOMI waste-form sample is given in Table 18. The cumulative quantity of each radionuclide released during leaching, expressed as fraction of initial inventory, and the ranges of the measured effective diffusivities and leachability indexes are given in Table 18. Not all of the leachates generated during testing of the LOMI waste form were analyzed for  $^{55}\text{Fe}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ , and  $^{241}\text{Pu}$ ; therefore, CFR values for these radionuclides are not reported. Neither  $^{125}\text{Sb}$  nor  $^{241}\text{Pu}$  were detected in any of the leachates, so their CFR results are also absent from Table 18. The effective diffusivities and leachability indexes of  $^{125}\text{Sb}$  and  $^{241}\text{Pu}$  were calculated using upper-limit concentrations determined for each leachate analysis. Of the radionuclides that were consistently detected,  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ , and  $^{63}\text{Ni}$  exhibited the largest ranges in effective diffusivities. Effective diffusivities of  $^{55}\text{Fe}$  varied by as much as three orders of magnitude. The effective diffusivities of  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$  were, on the other hand, each relatively constant, the high and low values varying by a maximum of a factor of 18. As

was the case with all other waste forms that were leach-tested, radionuclide release rates decreased as cumulative leaching time increased. The smallest average release rates occurred during the last two leaching intervals, which were 28 and 41 days long.

Comparing the leaching results in Table 18 with those in Table 13, the radionuclide leachability indexes for the LOMI waste form were similar to those determined for the Can-Decon waste forms. The leachability index of  $^{137}\text{Cs}$  ranged from 6.4 to 7.4 for the LOMI waste form, while similar values for the Can-Decon samples collectively ranged from 5.8 to 7.9. The leachability index of  $^{60}\text{Co}$  ranged from 12.5 to 13.6 for the LOMI waste form, while corresponding results for the Can-Decon samples ranged from 12.2 to 14.6. In general, the leaching results for the LOMI and Can-Decon waste forms were comparable and were different from the leaching results for the Dow NS-1 waste form in the respects discussed previously.

The ranges of radionuclide release rates determined for the Indian Point-3 LOMI waste form are presented in Table 19, where they are expressed as fraction of initial inventory released per second per square centimeter of sample external surface area ( $\text{F}/\text{cm}^2/\text{s}$ ). In all cases, maximum average radionuclide release rates occurred during the first 2-h-long leaching interval, while minimum average release rates occurred during the last two leaching periods which were about 4 and 6 weeks long. The maximum rates of release of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  were about  $1.7 \times 10^{-8}$  and  $1.9 \times 10^{-9} \text{ F}/\text{cm}^2/\text{s}$ , respectively, while similar results for  $^{60}\text{Co}$  and  $^{54}\text{Mn}$  were about  $1.4 \times 10^{-11}$  and  $1.1 \times 10^{-11} \text{ F}/\text{cm}^2/\text{s}$ , respectively. Minimum release rates ranged from a low value of  $1.2 \times 10^{-14} \text{ F}/\text{cm}^2/\text{s}$  for  $^{54}\text{Mn}$  to a high value of  $1.7 \times 10^{-10} \text{ F}/\text{cm}^2/\text{s}$  for  $^{137}\text{Cs}$ .

The CFR data for  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$  for the LOMI waste-form specimen are plotted as a function of the square root of elapsed leaching time in Figures 25 and 26. Results are plotted for 10 leaching intervals extending through 87 days. As shown in Figure 25 and Table 18, the total quantities of  $^{58}\text{Co}$  and  $^{60}\text{Co}$  released from the waste form agreed very well, their CFR values being  $1.2 \times 10^{-3}$  and  $1.3 \times 10^{-3}$ , respectively. These CFR values are comparable to the fractions of  $^{60}\text{Co}$  released from the Millstone-1 F201 and Peach Bottom-2 waste forms and are factors of between two and four higher than the  $^{60}\text{Co}$  release fractions determined for the Millstone-1 F33, Cooper mixed-bed resin, and Cooper cation resin waste forms. The total quantity of  $^{137}\text{Cs}$  released from the LOMI waste form at

**Table 18. Radionuclide leach-test results for Indian Point-3 LOMI waste form**

Nuclide	Range Maximum CFR	Range Effective Diffusivity (cm <sup>2</sup> /s)	Leachability Index
<sup>54</sup> Mn	— <sup>a</sup>	3.0 (– 17) to 1.6 (– 14)	13.8 to 16.5
<sup>55</sup> Fe	— <sup>b</sup>	5.6 (– 17) to 3.1 (– 13)	12.5 to 16.3
<sup>58</sup> Co	1.2 (– 3)	2.1 (– 14) to 3.1 (– 13)	12.5 to 13.7
<sup>60</sup> Co	1.3 (– 3)	2.8 (– 14) to 3.2 (– 13)	12.5 to 13.6
<sup>63</sup> Ni	— <sup>b</sup>	3.3 (– 15) to 1.5 (– 13)	12.8 to 14.5
<sup>90</sup> Sr	— <sup>b</sup>	5.1 (– 10) to 9.0 (– 9)	8.0 to 9.3
<sup>125</sup> Sb	— <sup>a</sup>	< 7.0 (– 13) to < 3.3 (– 10)	> 9.5 to > 12.2
<sup>137</sup> Cs	1.0 (+ 0)	4.3 (– 8) to 4.1 (– 7)	6.4 to 7.4
<sup>241</sup> Pu	— <sup>b</sup>	< 1.2 (– 14) to < 8.1 (– 12)	> 11.1 to > 13.9

a. Radionuclide was not detected in some of the leachates.

b. Not all leachate samples were analyzed for this radionuclide.

**Table 19. Radionuclide release rates for Indian Point-3 LOMI waste form**

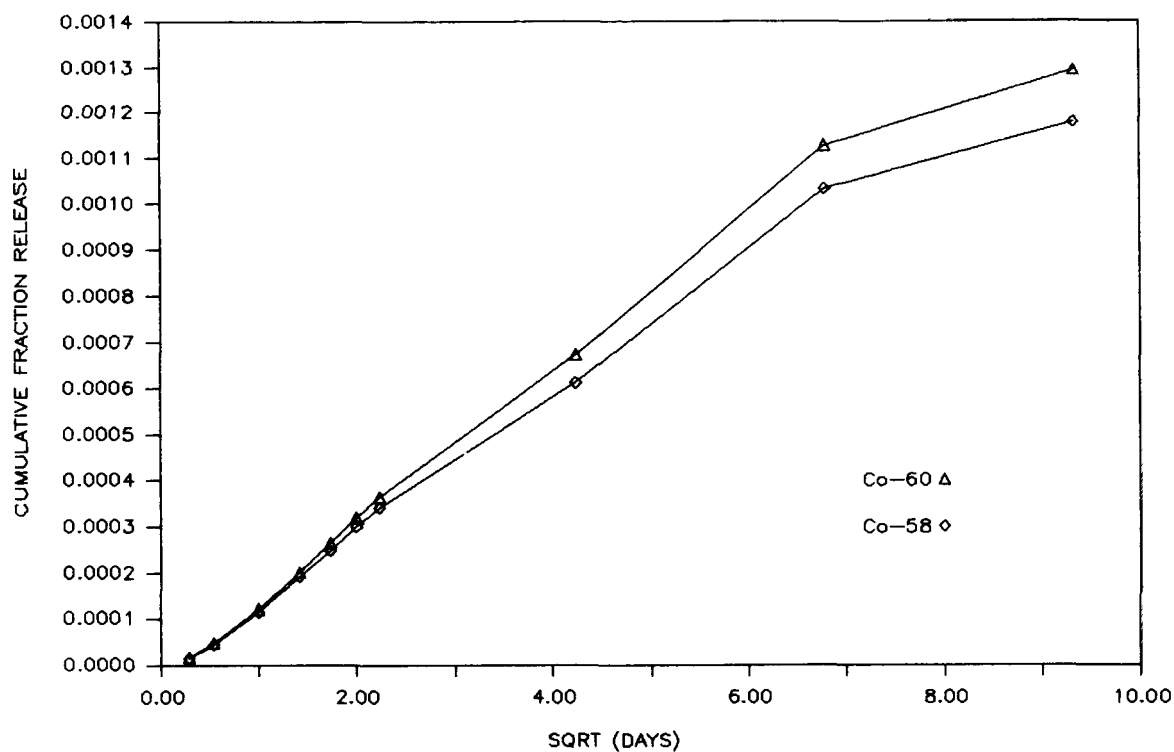
Nuclide	Release Rate <sup>a</sup> (F/cm <sup>2</sup> /s)
<sup>54</sup> Mn	1.07 (– 11) to 1.20 (– 14)
<sup>55</sup> Fe	4.68 (– 11) to 1.64 (– 14)
<sup>58</sup> Co	1.20 (– 11) to 2.41 (– 13)
<sup>60</sup> Co	1.40 (– 11) to 2.79 (– 13)
<sup>63</sup> Ni	3.24 (– 11) to 8.61 (– 14)
<sup>90</sup> Sr	1.90 (– 9) to 4.83 (– 11)
<sup>125</sup> Sb	< 4.13 (– 10) to < 1.26 (– 12)
<sup>137</sup> Cs	1.73 (– 8) to 1.70 (– 10)
<sup>241</sup> Pu	< 2.39 (– 10) to < 1.62 (– 13)

a. Fraction of initial inventory released per second per cm<sup>2</sup> of sample surface area.

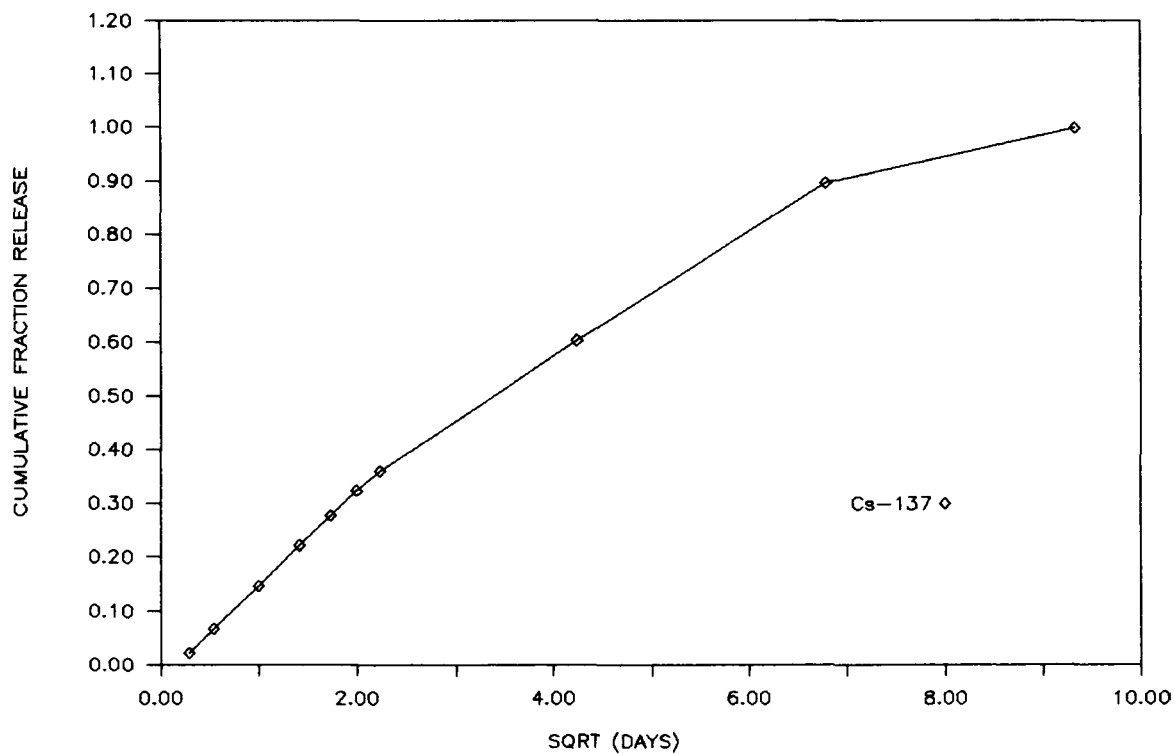
the end of 87 days of leaching was equal to the estimated initial inventory of <sup>137</sup>Cs in the sample.

As shown in Figures 25 and 26, the CFR data for <sup>58</sup>Co, <sup>60</sup>Co, and <sup>137</sup>Cs were linear over two or more time regions. The CFR data were approximately linear from 0 to 5 days and from 5 to 46 days. During the 46- to 87-day leaching interval, release rates of these radionuclides all dropped by about a factor of four compared to their average release rates during the previous leaching interval. The changes in slopes that are evident in Figures 25 and 26 indicate changes in release mechanisms (i.e., surface effect releases versus diffusion controlled releases) and/or changes in the forms of the species being leached. The similarity of the trends in the CFR plots of <sup>58</sup>Co, <sup>60</sup>Co, and <sup>137</sup>Cs indicates that both insoluble and soluble species were affected in the same way by changes in the operative release mechanisms.

**Summary.** The leaching data presented in the preceding subsections show that, with the exception of <sup>137</sup>Cs, only very small fractions of the inventories of the radionuclides contained in the waste forms were leached from the forms during leach-testing. The total quantity of <sup>54</sup>Mn released ranged from <0.005% for the Millstone-1 F201 waste form to <0.2% for the Pilgrim waste form, where the results are expressed as percent of initial



**Figure 25.** Cumulative fraction release of  $^{58}\text{Co}$  and  $^{60}\text{Co}$  from the Indian Point-3 LOMI waste form.



**Figure 26.** Cumulative fraction release of  $^{137}\text{Cs}$  from the Indian Point-3 LOMI waste form.

inventory. Excluding the results for the Pilgrim waste form, release fractions for  $^{60}\text{Co}$  ranged from 0.03% for the Cooper cation resin waste form to 0.2% for the Millstone-1 F201 waste form. The fraction of the initial inventory of  $^{60}\text{Co}$  released from the Pilgrim waste form was much higher than usual, being about 5%. Similar results for  $^{125}\text{Sb}$  ranged from 0.2% for the Millstone-1 F33 sample to <7% for the Pilgrim waste form. As was the case with  $^{60}\text{Co}$ , the fraction of  $^{125}\text{Sb}$  released from the Pilgrim sample was higher than normal. The  $^{125}\text{Sb}$  release fractions determined for the Millstone-1 F201 and Cooper mixed-bed resin samples were comparable to the result for the Millstone-1 F33 sample, the release fractions of the former samples being 0.3% and 0.5%, respectively.

The fact that the fractions of the initial inventories of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ , and  $^{125}\text{Sb}$  leached from the waste forms were usually very low suggests that releases of some species may have been confined to regions near the surfaces of the waste forms. In order to assess this possibility, calculations were performed to determine to what depths the surfaces of the samples would have to have been depleted of activity in order to produce the observed release fractions. For a cylindrical solid whose surface is depleted to a depth  $\alpha$ , the volume of the depleted region,  $V_D$ , is given by

$$V_D = S_A\alpha - \pi(h + 4r)\alpha^2 + 2\pi\alpha^3 \quad (7)$$

where

$V_D$  = volume of depleted surface layer ( $\text{cm}^3$ ),

$S_A$  = external surface area of sample ( $\text{cm}^2$ ),

$r$  = radius of sample (cm),

$h$  = height of sample (cm), and

$\alpha$  = depth of depleted region (cm).

Volumes of depleted regions were estimated by assuming that activity was homogeneously distributed within the waste forms, in which case the depleted volume of each waste form for a given radionuclide would be equal to the product of the CFR of the radionuclide and the sample volume. Depleted volumes were estimated in this manner, and Equation (7) was used to calculate the depths to which the surfaces of the samples would have to

have been depleted of activity to yield the observed release fractions.

For all samples except the Dow NS-1 waste form, the total quantities of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ , and  $^{125}\text{Sb}$  released from the waste forms can be accounted for if only the first 30  $\mu\text{m}$  of the surface layer of each sample were totally depleted of activity. The results for the Indian Point-3 and Peach Bottom-2 waste forms indicate that a depleted layer only 12- $\mu\text{m}$  thick would account for the total quantities of  $^{60}\text{Co}$  released from the samples. Surfaces depleted to depths of 18 and 27  $\mu\text{m}$ , respectively, would account for the  $^{125}\text{Sb}$  released from the Millstone-1 F33 and F201 samples. Even in the case of the Pilgrim sample, which released about 5% of its inventory of  $^{60}\text{Co}$ , a surface totally depleted of  $^{60}\text{Co}$  to a depth of 0.5 mm would account for the total quantity of  $^{60}\text{Co}$  released from the waste form.

These results indicate that it is possible that the observed releases of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ , and  $^{125}\text{Sb}$  were mainly the result of release mechanisms operating exclusively on or very near the surfaces of the waste forms. The operative release mechanisms might have included corrosion and dissolution of the waste-form surfaces and diffusion from just beneath the surfaces. The sharp declines in release rates that were observed over the course of leaching are consistent with significant depletion of activities from the surfaces of the waste forms. It is known that the concentration gradient between the waste-form surface and the leachate affects the release rate of a diffusing species. If the rates of diffusion of species within the waste forms were negligible compared to their rates of release from the surfaces of the waste forms, as the waste-form surfaces became depleted of a species, the equilibrium concentration in the leachate would have decreased and, consequently, the average release rate for leaching intervals of the same length would have decreased. For longer leaching intervals, the release rate would have decreased as the equilibrium concentration was approached, resulting in lower average release rates for longer leaching intervals. The declining release rates that were observed for each sample were likely a consequence of the waste-form surfaces becoming depleted of activity because of the imbalance between internal diffusion rates and surface release rates.

While the releases of transition metal species were generally very small, in most cases the entire inventories of  $^{137}\text{Cs}$  in the waste forms were released during leach-testing. The cumulative quantities of  $^{137}\text{Cs}$  released from the Millstone-1 F33 and F201 waste forms were 98% and 96% of initial

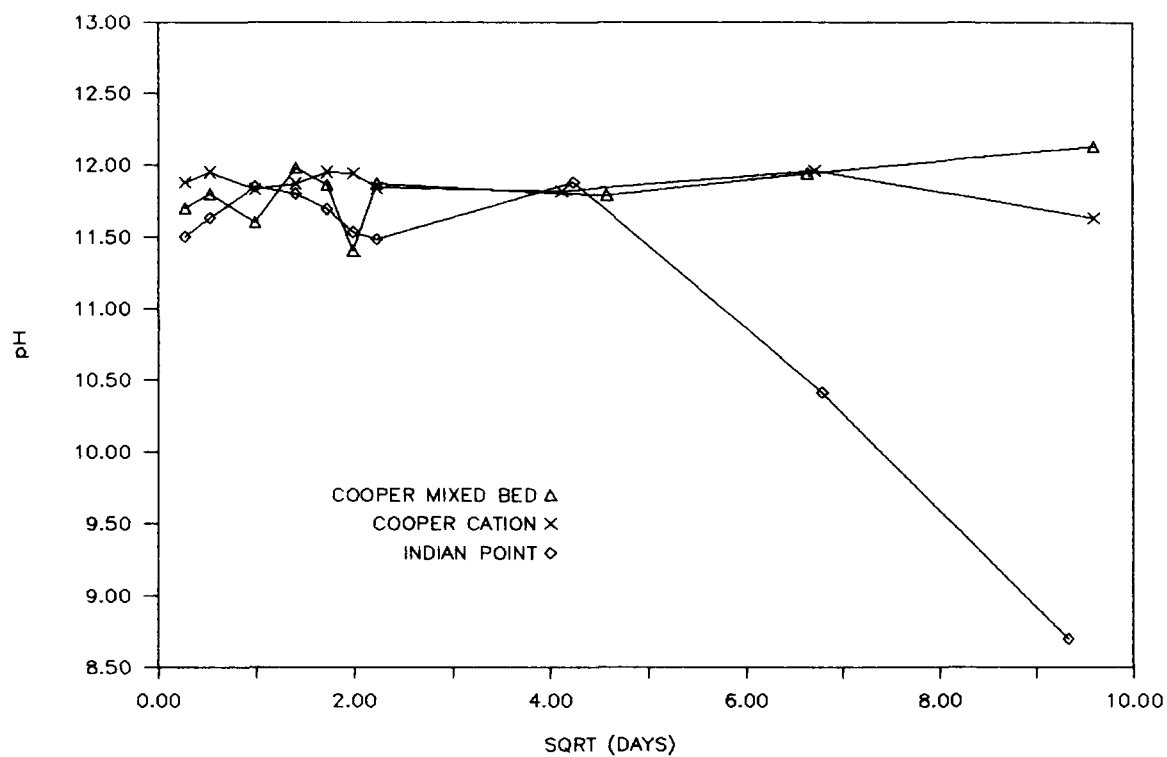
inventories, respectively; while similar values for the Cooper mixed-bed resin, Cooper cation resin, and Indian Point-3 waste forms were each 100%. The total quantity of  $^{137}\text{Cs}$  released from the Peach Bottom-2 sample was about 36% of the original  $^{137}\text{Cs}$  inventory. The Pilgrim waste form released an unusually small quantity of  $^{137}\text{Cs}$ , less than 0.4% of the amount of  $^{137}\text{Cs}$  estimated to be in the waste form prior to the start of leach-testing. It is not known if special additives were used during the solidification of the Pilgrim waste form, but the results suggest that additives such as zeolites, clay, or silica were employed to reduce the leachability of  $^{137}\text{Cs}$ . The fact that large fractions of the original inventory of  $^{137}\text{Cs}$  were released from most of the waste forms indicates that  $^{137}\text{Cs}$  normally remained soluble following cement hydration and that the diffusion of  $^{137}\text{Cs}$  was not encumbered by activity gradients extending over several centimeters.

The ANS 16.1 leach-test procedure is intended to minimize the effects of changes in leachant chemistry on the leach rate of a waste form during a leaching interval. However, if a waste-form binder material has soluble components [e.g.,  $\text{Ca}(\text{OH})_2$  in the case of cement-solidified waste forms], the leachate may quickly become saturated with one or more soluble binder compounds. Evidence of the introduction of soluble binder compounds into the leachates is shown in Figures 27 and 28, which are graphs of leachate pH as a function of the square root of elapsed leaching time. As shown in the figures, the pH of the leachates of the Cooper mixed-bed resin, the Cooper cation resin, and the Millstone-1 F33 and F201 samples were comparable and remained reasonably constant throughout the durations of the leach tests. The pH of the leachates of these four samples ranged from a low of 11.4 to a high of 12.4. In contrast to these results, the pH of the leachates of the Indian Point-3, Peach Bottom-2, and Pilgrim waste forms did not remain constant throughout the leach tests. In the case of the Indian Point-3 waste form, leachate pH dropped significantly during each of the last two leaching intervals. It decreased from 11.9 during the 5- to 18-day leaching interval to 10.4 during the next leaching interval to 8.7 during the last leaching interval. The pH of leachates of both the Peach Bottom-2 and Pilgrim waste forms dropped rather abruptly from values close to 11.0 to about 8.2 during the first week of leaching and then came back up to previous values. The leachates of the samples that showed signs of physical deterioration during testing (i.e., Cooper and, to a lesser extent, Millstone-1 samples) exhibited relatively constant

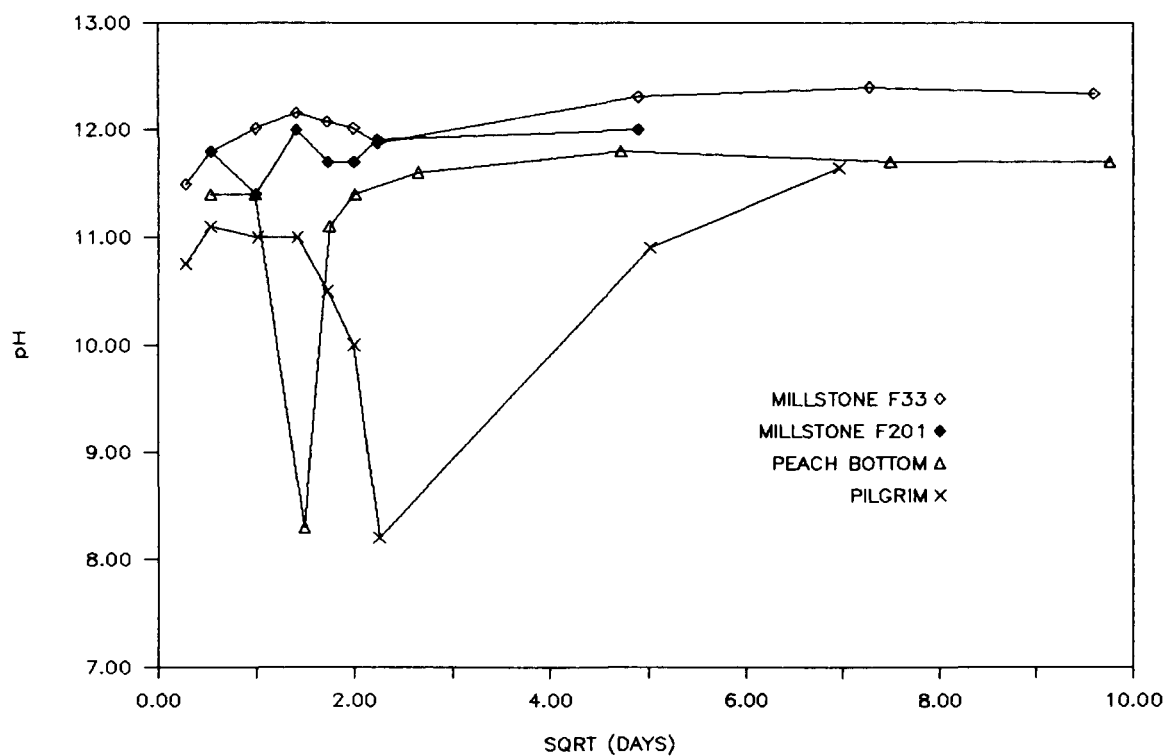
pH values; and the leachates of the remaining three waste forms, which showed no obvious signs of degradation, each exhibited significant variations in pH. The reason why the pH of leachates of decomposing samples would remain relatively constant is clear, since the buffering capacity of such samples is renewed when new surfaces are exposed. The behavior of the pH of the Peach Bottom-2 and Pilgrim leachates is not clearly understood.

As was previously discussed, a sample of each leachate was passed through a cation exchange membrane. Based on the assumption that cationic species were retained by the membrane, estimates were made of the fractions of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  that remained in solution as neutral or anionic compounds or ligands. Measurement results are presented in Appendix G, Tables G-1 through G-7. Excluding the results for the Cooper Citrox waste forms, the data indicate that  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  were present in leachate solutions primarily as neutral or anionic compounds or ligands. In general, the cation exchange membranes retained less than 10% of the total quantities of these cations in the leachate samples. In the case of the two Cooper Citrox waste forms, the fractions of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  retained by the cation exchange membranes varied from 0 to 0.85. These results indicate that the Cooper Citrox waste-form leachates were not adequately filtered prior to being passed through the cation exchange membranes. High retention fractions very likely reflect the fact that the membranes efficiently retained any particulates in solution. Because the leachates were all basic (i.e., pH normally ranged from 10 to 12), one would expect the population of free cations in solution to be negligible. More sophisticated analysis techniques will be required to determine the chemical forms of released radionuclides.

The leachability indexes of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  are plotted separately as a function of elapsed leaching time in Figures 29 through 35, respectively. The differences in the leaching behavior of the Pilgrim sample compared to that of other samples is quite striking in Figures 31 and 35, which, respectively, present the leachability indexes of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ . These two figures also show a general difference that exists between the releases of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ . As shown in Figure 31, the leachability indexes of  $^{60}\text{Co}$  occupy a relatively narrow range during the initial leaching intervals and then diverge as leaching intervals become longer. The leachability indexes of  $^{137}\text{Cs}$  occupy a relatively wide range during the first two weeks of leaching and then converge during



**Figure 27.** Leachate pH for Cooper and Indian Point-3 waste forms.



**Figure 28.** Leachate pH for Millstone-1, Peach Bottom-2, and Pilgrim waste forms.



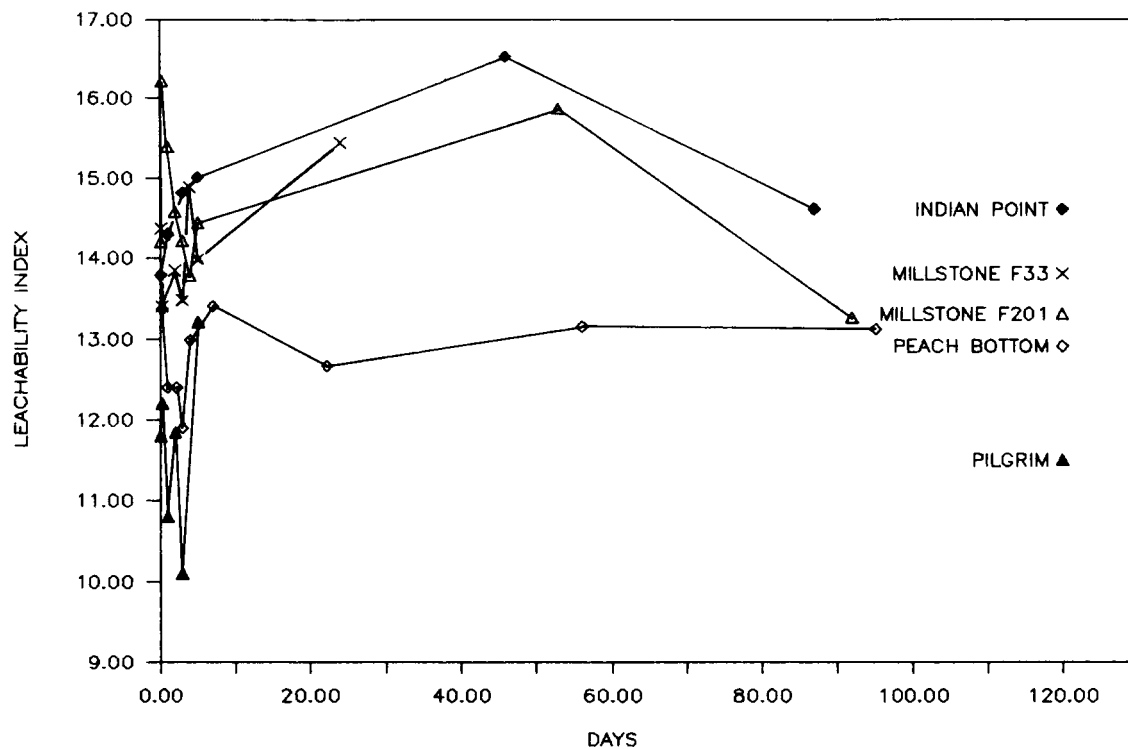


Figure 29. Leachability indexes for the release of  $^{54}\text{Mn}$ .

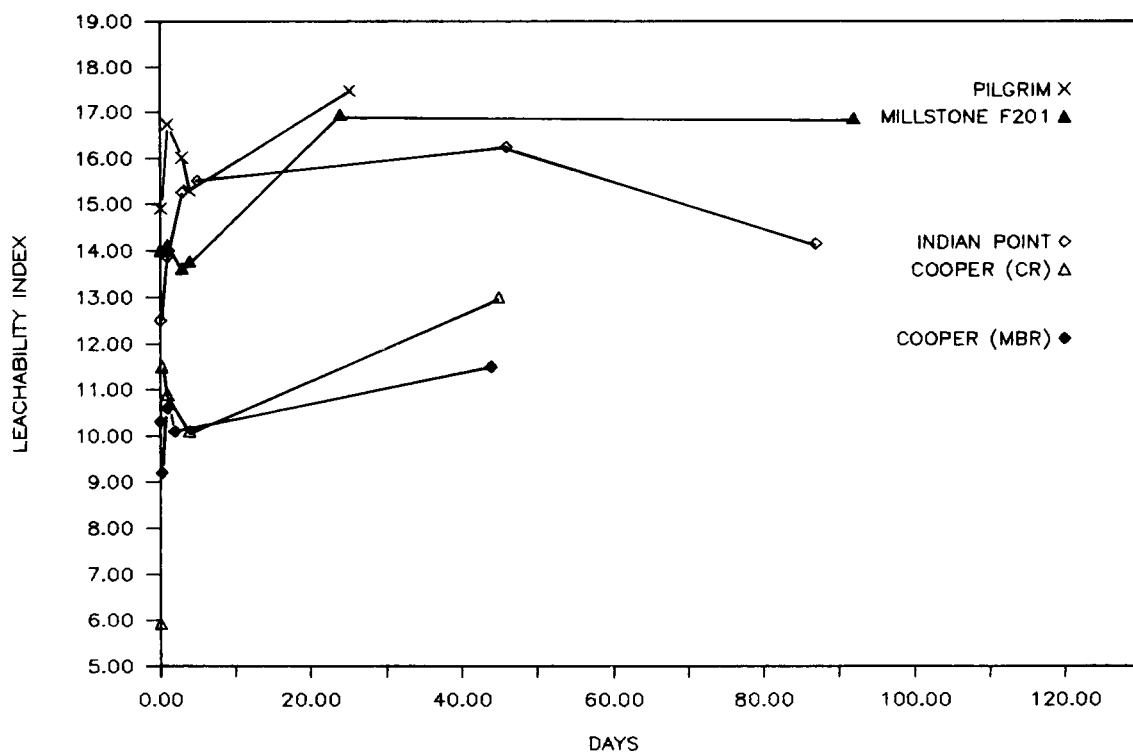


Figure 30. Leachability indexes for the release of  $^{55}\text{Fe}$ .

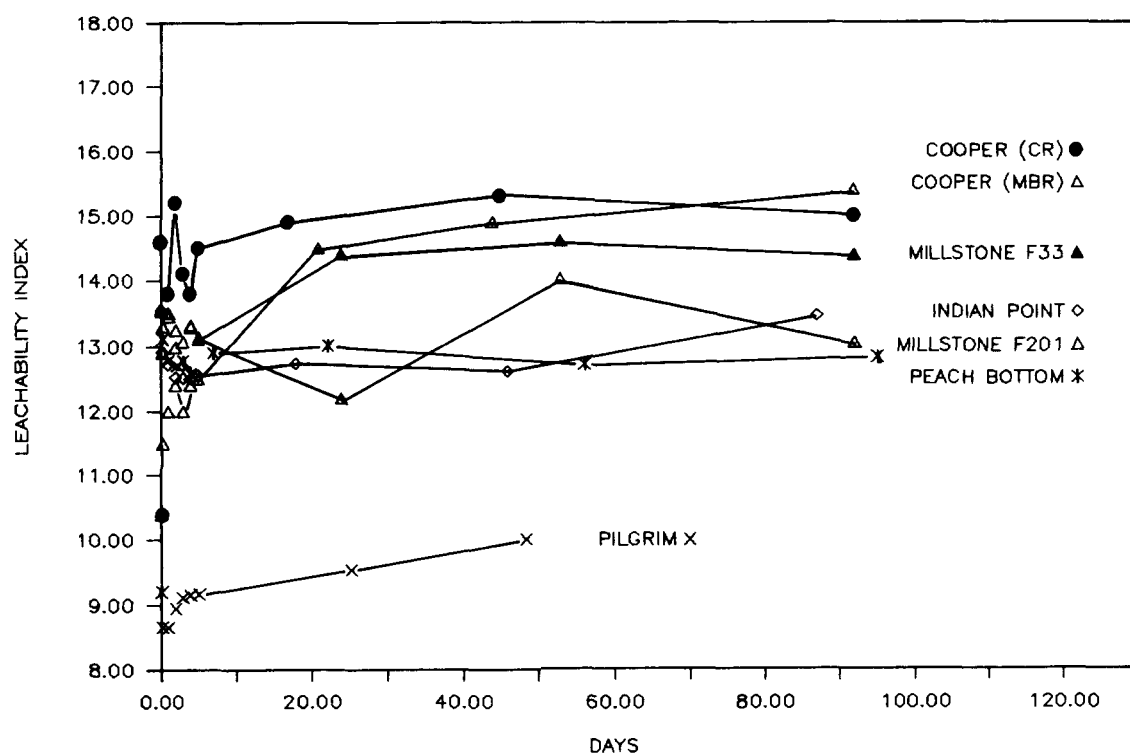


Figure 31. Leachability indexes for the release of  $^{60}\text{Co}$ .

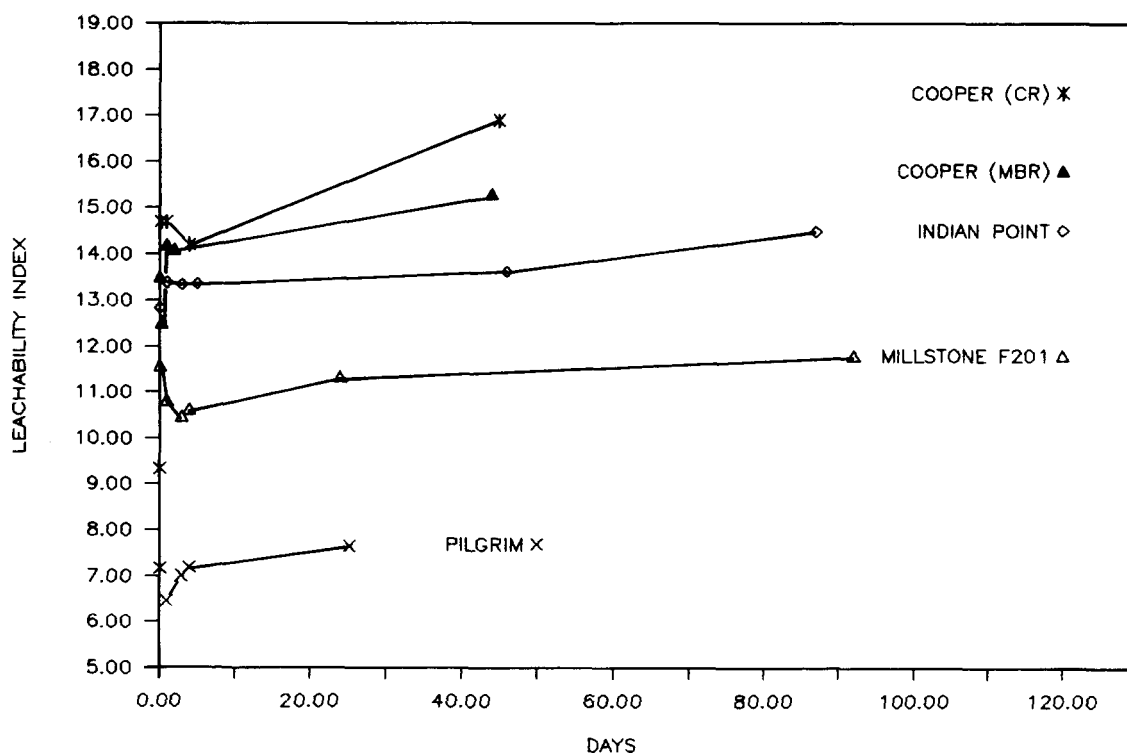
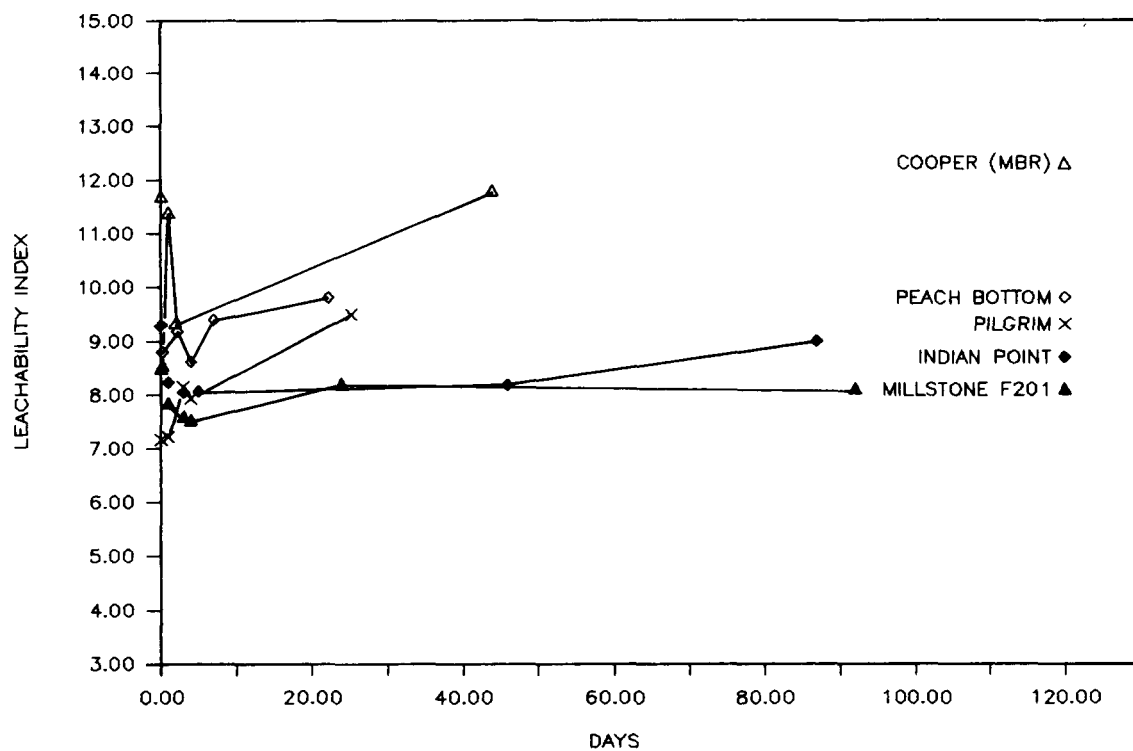
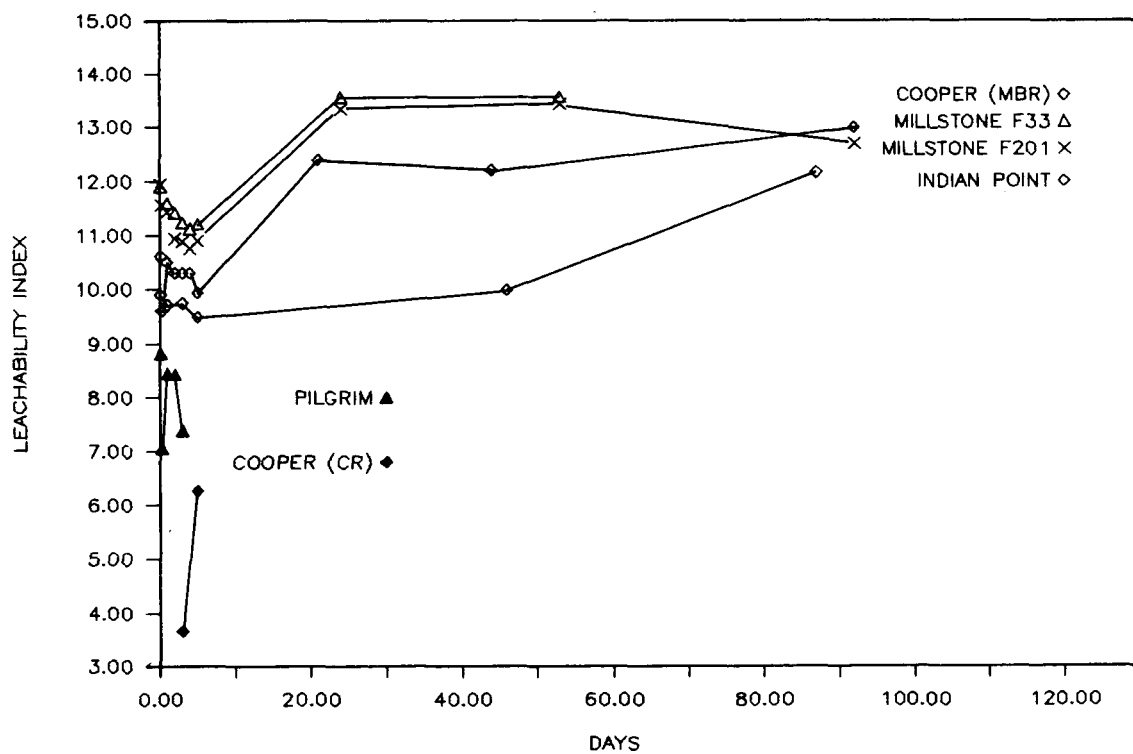


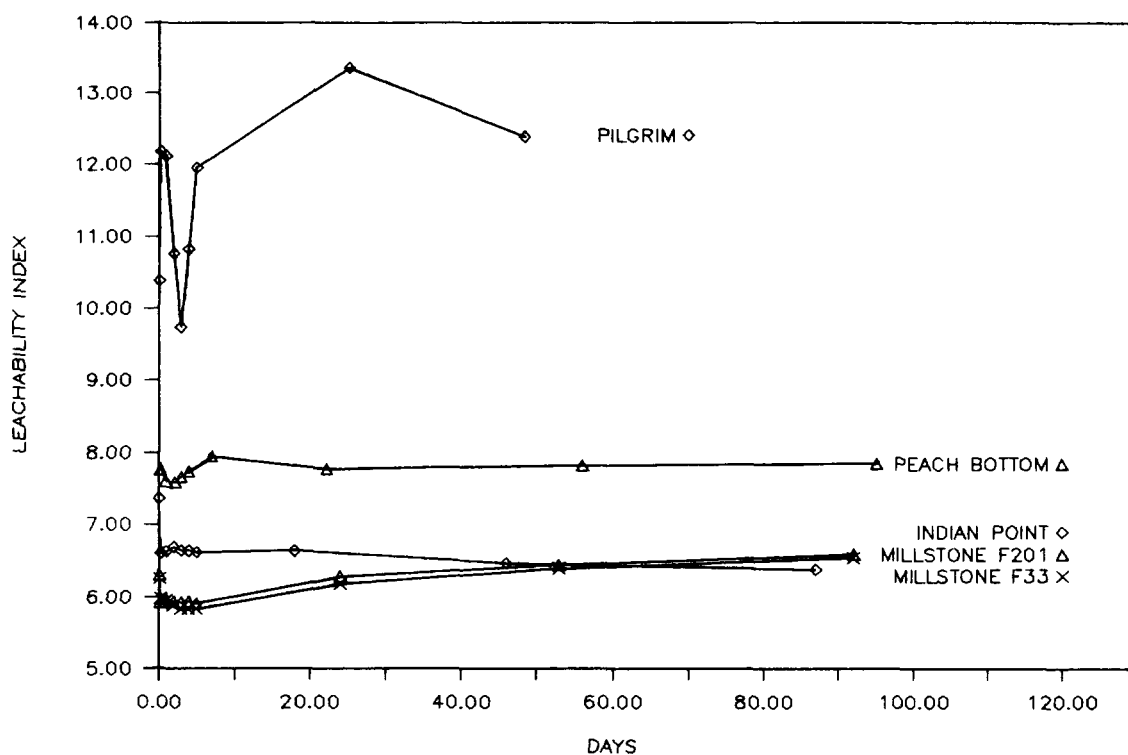
Figure 32. Leachability indexes for the release of  $^{63}\text{Ni}$ .



**Figure 33.** Leachability indexes for the release of  $^{90}\text{Sr}$ .



**Figure 34.** Leachability indexes for the release of  $^{125}\text{Sb}$ .



**Figure 35.** Leachability indexes for the release of  $^{137}\text{Cs}$ .

later leaching intervals when the samples become depleted of their  $^{137}\text{Cs}$  inventories.

Average leachability indexes of nine radionuclides are presented in Table 20 for each of the seven waste forms that were leach-tested. Uncertainties given in Table 20 are one sigma standard deviations of the mean. In those cases where only lower-limit values or combinations of lower-limit values and measured values of the leachability index were determined, the results are given as ranges rather than as average values. Because both of the Cooper waste-form samples decomposed into loose granular debris during the initial phases of leach-testing, the original models for these two samples had to be modified. The leachability indexes for the Cooper waste-form samples were calculated, in each case, assuming that the sample boundary was the top surface of the loose debris residing in the bottom of the leaching container. The debris of each sample was uniformly distributed over the bottom of the sample container, so the surface area of the top surface of the debris was assumed to be the same as the surface area of the bottom of the container, namely,  $177\text{ cm}^2$ . The volumes of the debris beds were estimated assuming 50% void fractions. Based on these assumptions,

the volume-to-surface-area ratios of the Cooper samples following decomposition were 1.7 for the mixed-bed resin sample and 1.8 for the cation resin sample. Leachability indexes were then calculated in the usual manner using the new volume-to-surface-area ratios. Average leachability indexes for the Cooper samples were calculated for two different time regions, one corresponding to the period during which decomposition occurred and the second encompassing the time region characterized by lower release rates. The time periods used are indicated in Table 20.

Average leachability indexes of  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  are plotted in Figure 36, and similar values for  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ , and  $^{63}\text{Ni}$  are plotted in Figure 37. The results presented in Table 20 and Figures 36 and 37 show that the average leachability indexes of the duplicate Millstone-1 Can-Decon samples agreed very well. Leachability indexes of  $^{54}\text{Mn}$  and  $^{60}\text{Co}$  for the Peach Bottom-2 Can-Decon waste form were somewhat lower than their corresponding values for the Millstone-1 samples. As expected, the  $^{60}\text{Co}$  leachability indexes of the Cooper mixed-bed and cation waste forms increased after surface effect releases diminished. During the initial leaching intervals when the

**Table 20. Average leachability indexes of radionuclides**

Sample	<sup>54</sup> Mn	<sup>55</sup> Fe	<sup>58</sup> Co
Millstone-1 F33	14.2 ± 0.2	— <sup>a</sup>	8.3 to >9.4
Millstone-1 F201	14.7 ± 0.3	14.9 ± 0.6	>8.1 to >9.7
Peach Bottom-2	11.9 to >13.4	— <sup>a</sup>	— <sup>b</sup>
Cooper (MBR)			
0 to 5 day	12.7 <sup>c</sup>	10.0 ± 0.3	— <sup>b</sup>
5 to 92 day	— <sup>b</sup>	11.5 <sup>d</sup>	— <sup>b</sup>
Cooper (CR)			
0 to 7 h	10.5 <sup>c</sup>	8.7 ± 2.0	— <sup>b</sup>
7 h to 92 day	14.3 <sup>c</sup>	11.5 ± 0.8	— <sup>b</sup>
Pilgrim	>10.1 to 13.2	16.1 ± 0.4	>4.5 to >5.9
Indian Point-3	14.8 ± 0.3	14.6 ± 0.5	12.9 ± 0.1
Sample	<sup>60</sup> Co	<sup>63</sup> Ni	<sup>90</sup> Sr
Millstone-1 F33	13.6 ± 0.2	— <sup>a</sup>	— <sup>a</sup>
Millstone-1 F201	13.2 ± 0.1	11.1 ± 0.2	8.0 ± 0.1
Peach Bottom-2	12.8 ± 0.1	— <sup>a</sup>	9.2 ± 0.2
Cooper (MBR)			
0 to 5 day	11.9 ± 0.3	13.6 ± 0.3	10.2 ± 0.7
5 to 92 day	14.9 ± 0.2	15.3 <sup>c</sup>	11.8 <sup>c</sup>
Cooper (CR)			
0 to 7 h	12.5 ± 1.5	12.0 ± 1.9	— <sup>d</sup>
7 h to 92 day	14.6 ± 0.2	15.3 ± 0.7	— <sup>d</sup>
Pilgrim	9.2 ± 0.1	7.1 ± 0.2	8.0 ± 0.4
Indian Point-3	12.8 ± 0.1	13.5 ± 0.2	8.5 ± 0.2
Sample	<sup>125</sup> Sb	<sup>137</sup> Cs	<sup>241</sup> Pu
Millstone-1 F33	11.9 ± 0.3	6.1 ± 0.1	— <sup>a</sup>
Millstone-1 F201	11.8 ± 0.3	6.1 ± 0.1	>12.9 to >16.4
Peach Bottom-2	— <sup>b</sup>	7.8 ± 0.1	>9.6 to >11.1
Cooper (MBR)			
0 to 5 day	10.1 ± 0.1	— <sup>d</sup>	>10.3 to >11.4
5 to 92 day	12.5 ± 0.2	— <sup>d</sup>	>12.8 <sup>c</sup>
Cooper (CR)			
0 to 7 h	— <sup>b</sup>	— <sup>d</sup>	6.9 to >10.2
7 h to 92 day	< 3.7 to <6.3	— <sup>d</sup>	>10.8 to >12.8
Pilgrim	> 7.1 to >8.8	>10.4 to 13.4	>11.5 to >13.5
Indian Point-3	> 9.5 to >12.2	6.7 ± 0.1	>11.1 to >13.9

a. Analyses of resin waste and/or leachate samples were not performed.

b. Not detected in resin waste and/or leachate samples.

c. Leachability index was determined for only one leachate sample.

d. Release fraction was greater than 20% which ruled out using approximate diffusion model.

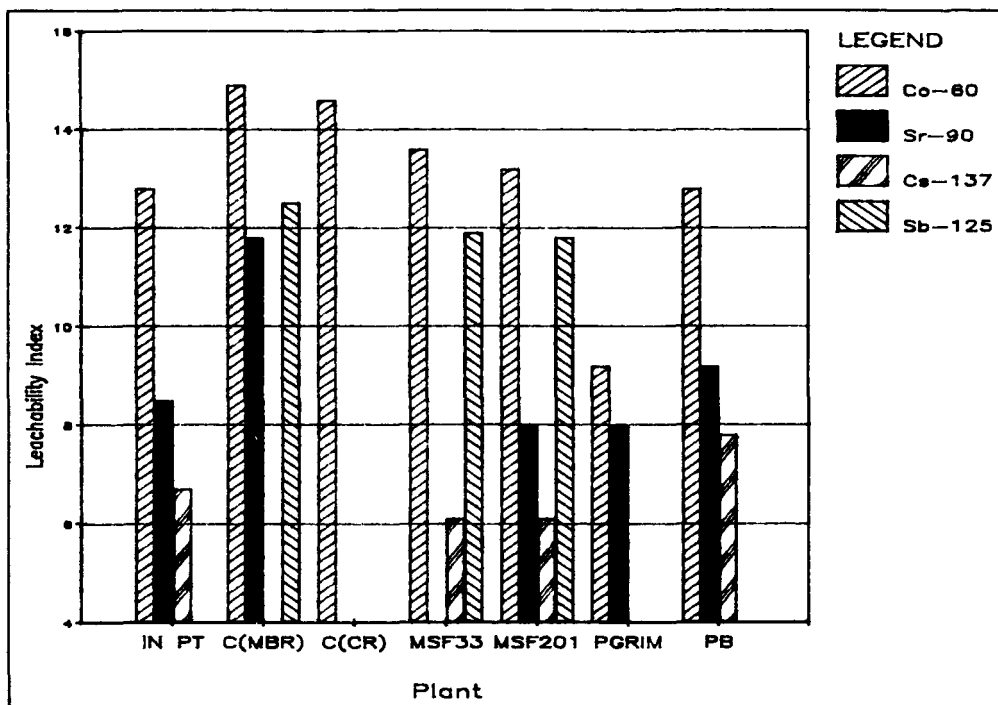


Figure 36. Average leachability indexes of  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$ .

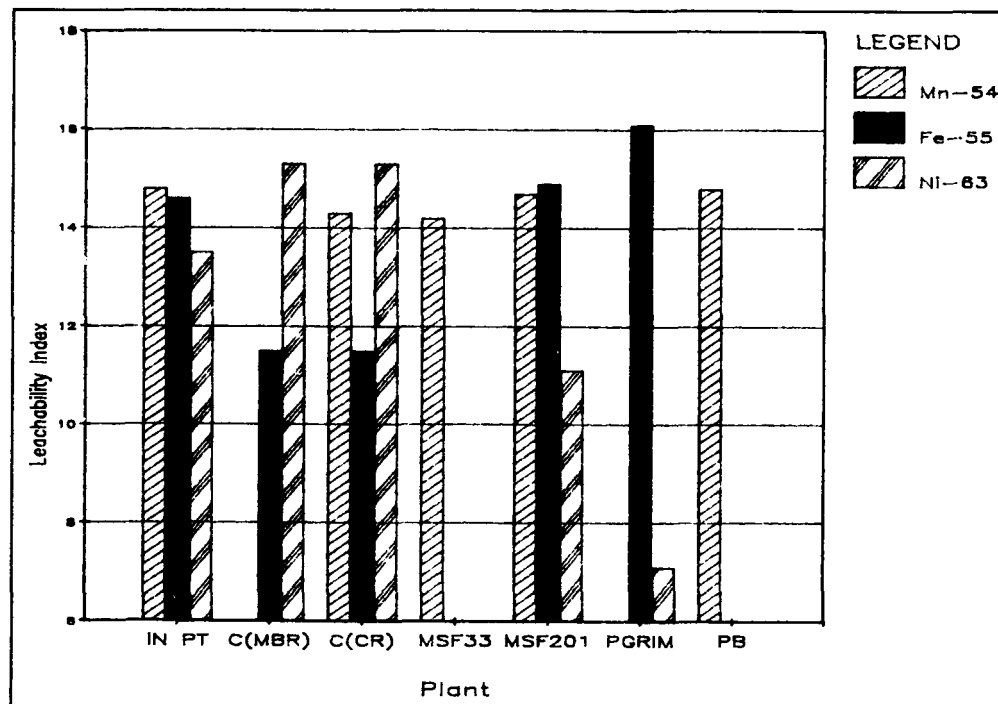


Figure 37. Average leachability indexes of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ , and  $^{63}\text{Ni}$ .

samples were decomposing, the average  $^{60}\text{Co}$  leachability indexes of the mixed-bed and cation resin waste forms were 11.9 and 12.5, respectively. Throughout the subsequent leaching intervals, the average  $^{60}\text{Co}$  leachability indexes were 14.9 and 14.6, respectively. Because both of the Cooper samples decomposed so completely, a sizeable fraction of the resin beads in the loose debris were directly exposed to the leachates. In spite of this condition, the measured releases of radionuclides from these two samples were comparable to the releases determined for samples that remained intact.

As shown in Table 20, with the exception of the results for  $^{58}\text{Co}$  for the waste-form specimen obtained from Pilgrim Station and the results for  $^{125}\text{Sb}$  for the cation resin waste-form specimen obtained from Cooper Station, the measured radionuclide leachability indexes were all greater than six, which is the lower limit considered acceptable according to the NRC Branch Technical Position on Waste Form.<sup>8</sup> Cobalt-58 was not detected in any of the Pilgrim waste-form leachates; therefore, the corresponding leachability indexes presented in Table 20 are lower-limit values. Antimony-125 was detected in only two of the Cooper cation resin waste-form leachates, and  $^{125}\text{Sb}$  was not detected in the Cooper cation resin sample. Consequently, the corresponding leachability indexes shown in Table 20 are upper-limit values. Leachability indexes of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ ,  $^{125}\text{Sb}$ , and  $^{241}\text{Pu}$  were typically greater than 10. Similar results for  $^{90}\text{Sr}$  ranged from 8.0 to 11.8, while leachability indexes of  $^{137}\text{Cs}$  varied from 6.1 to 13.4. Radionuclide leachability indexes determined for the cation and mixed-bed resin waste-form specimens obtained from Cooper Station were similar to corresponding leachability indexes measured for specimens which maintained their physical integrity during leaching.

In a separate study<sup>28</sup> previously conducted at INEL, the leachability indexes of  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$  were determined for boric acid and sulfate evaporator wastes which had been solidified in cement at two operating commercial LWRs. Different sized waste-form samples were leach-tested in deionized water using a modified IAEA leach-test procedure. The results of this study are summarized in Table 21.

The results presented in Tables 20 and 21 show that, with the exception of the result for  $^{60}\text{Co}$  for the Pilgrim sample, the average leachability indexes of  $^{60}\text{Co}$  and  $^{90}\text{Sr}$  for the decontamination waste-form samples were consistently higher than corresponding leachability indexes determined for boric

**Table 21. Leaching results for boric acid and sulfate waste forms**

Sample	Range Leachability Index		
	$^{60}\text{Co}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$
Boric acid	10.3 to 11.2	6.7 to 7.7	6.2 to 6.7
Sulfate	10.7 to 11.5	7.7 to 8.3	6.1 to 6.7

acid and sulfate waste forms. Ignoring the anomalously high value determined for the Pilgrim waste form, the leachability indexes of  $^{137}\text{Cs}$  were comparable for the evaporator and decontamination resin waste-form samples.

## Release of Stable Metals from Waste Forms

This section presents the results of measurements of the release of chromium, iron, cobalt, and nickel during leaching of the seven decontamination resin waste specimens. These transition metals were chosen for analysis because they are the primary constituents of stainless steel and Inconel, which are the materials used to line the internal surfaces of LWR primary coolant systems. Leachate samples were analyzed by ICP-AES, as described previously. Approximately one-half of the total number of leachate samples obtained during leach-testing were analyzed for these four transition metals. Consequently, it was not possible to determine the cumulative quantities of metals released during leaching. As was shown in Table 11, the Peach Bottom-2 resin waste sample was not analyzed for stable metals, nor were the Cooper mixed bed and Pilgrim resin waste samples analyzed for chromium. In these cases, effective diffusivities and leachability indexes could not be determined. In those cases where the leachate concentration was below the detection limit, leachability indexes were calculated using upper-limit concentrations.

A summary of the stable metal leach-test results for five of the waste-form samples is given in Table 22. Leachates obtained from testing the Millstone-1 F33 sample were not analyzed for stable metals; consequently, only the results for the F201 Millstone-1 waste-form sample are presented in the table. The effective diffusivities and leachability indexes of the Cooper waste forms were calculated using the same assumptions described in

**Table 22. Stable metal leach-test results for waste forms**

Element	Range Effective Diffusivity (cm <sup>2</sup> /s)	Range Leachability Index
<u>Millstone-1 F201:</u>		
Chromium	<1.5 (–13) to <1.4 (–11)	>10.8 to >12.8
Iron	<5.3 (–16) to <5.3 (–14)	>13.3 to >15.3
Cobalt	<1.8 (–9) to <1.7 (–7)	> 6.8 to > 8.8
Nickel	2.7 (–12) to 7.6 (–11)	10.1 to 11.6
<u>Cooper Mixed Bed Resin:</u>		
Chromium	— <sup>a</sup>	— <sup>a</sup>
Iron	<3.4 (–13) to <2.3 (–11)	>10.6 to >12.5
Cobalt	— <sup>b</sup>	— <sup>b</sup>
Nickel	<4.3 (–10) to <92.8 (–8)	7.6 to > 9.4
<u>Cooper Cation Resin:</u>		
Chromium	1.1 (–11) to 1.0 (–6)	6.0 to 11.0
Iron	<96.8 (–14) to 6.7 (–6)	5.2 to >13.2
Cobalt	<96.2 (–10) to 7.9 (–7)	6.1 to > 9.2
Nickel	2.1 (–12) to 2.0 (–6)	5.7 to 11.7
<u>Pilgrim:</u>		
Chromium	— <sup>b</sup>	— <sup>b</sup>
Iron	<99.7 (–15) to <91.0 (–12)	>12.0 to >14.0
Cobalt	<93.2 (–9) to <93.4 (–7)	> 6.5 to > 8.5
Nickel	2.7 (–8) to 4.6 (–7)	6.3 to 7.6
<u>Indian Point-3:</u>		
Chromium	<92.0 (–12) to <92.3 (–10)	> 9.6 to >11.7
Iron	<93.7 (–14) to 1.8 (–11)	10.7 to >13.4
Cobalt	— <sup>b</sup>	— <sup>b</sup>
Nickel	7.3 (–12) to 2.9 (–10)	9.5 to 11.1

a. Concentration in resin waste was not determined.

b. Concentrations in both resin waste and leachates were less than detection limits.



the preceding subsection. The ranges of the effective diffusivities and leachability indexes of chromium, iron, cobalt, and nickel were generally comparable in magnitude to the ranges determined for radionuclides. However, the effective diffusivities of the Cooper cation waste form exhibited unusually large ranges. For example, the effective diffusivities determined for iron varied by as much as eight orders of magnitude, resulting in corresponding leachability indexes that ranged from 5.2 to greater than 13.2. For all metals, the highest effective diffusivities for the Cooper cation sample were those determined for the first leaching interval. The sample began to decompose when first immersed in leachant and released significant amounts of particulate matter into the leachate. The presence of this particulate matter likely accounts for the unusually low stable metal leachability indexes determined for the first leaching period.

The ranges of the release rates of chromium, iron, cobalt, and nickel are presented in Table 23. The release rates for the Millstone-1 F201, Pilgrim, and Indian Point-3 samples are expressed as fraction of initial inventory released per second per square centimeter of sample external surface area, while those for the Cooper samples were not normalized by sample surface area. The results shown in Table 23 indicate that nickel was released from the Pilgrim sample at a rate about ten times higher than the rate it was released from the Millstone-1 F201 and Indian Point-3 samples. This behavior correlates well with the results for  $^{63}\text{Ni}$ . Comparing the  $^{63}\text{Ni}$  release rates presented in Tables 14 and 17 with similar results for nickel indicates that for the Millstone-1 and Pilgrim samples,  $^{63}\text{Ni}$  was released at about the same rate as nickel. As was the case with radionuclides, the release rates of the stable metals decreased as leaching time increased.

The leachability indexes of chromium, iron, cobalt, and nickel for five of the waste-form samples are plotted as a function of elapsed leaching time in Figures 38 through 41, respectively. Because chromium, iron, and cobalt were frequently not detected in leachate samples, the majority of the leachability indexes for these metals had to be estimated using their corresponding detection limit concentrations. Therefore, the graphs of the chromium, iron, and cobalt data indicate the lower limits of the leachability indexes as a function of time but are not particularly useful for other purposes. (Leachability indexes of stable metals for individual leaching intervals are tabulated in Appendix E.) However, nickel was detected

and quantified in almost all of the leachate samples. Comparing Figure 41 with Figure 32, it is clear that for the Pilgrim and Millstone-1 F201 samples the leachability indexes of  $^{63}\text{Ni}$  and nickel were about equal and behaved similarly as a function of time. But this correlation did not apply to the Indian Point-3 results, which indicated a much higher release of nickel compared to  $^{63}\text{Ni}$ .

Average leachability indexes of chromium, iron, cobalt, and nickel are presented in Table 24. Uncertainties given in Table 24 are one-sigma standard deviations of the mean. In most cases, only lower-limit values or combinations of lower-limit values and measured values of the leachability index were determined. In these cases, the results are reported as ranges rather than as average values. Using the same approach used for radionuclides, average leachability indexes of the Cooper samples were calculated for two different time regions, one corresponding to the period during which sample decomposition occurred and the second encompassing the time region characterized by lower release rates. The time periods used are the same as those used to calculate average radionuclide leachability indexes. Average leachability indexes of nickel for the Millstone-1 F201 and Pilgrim samples were  $10.7 \pm 0.2$  and  $6.9 \pm 0.3$ , respectively. Similar results for  $^{63}\text{Ni}$  for these two samples were approximately the same as the results for nickel, the average leachability indexes of  $^{63}\text{Ni}$  being  $11.1 \pm 0.2$  and  $7.1 \pm 0.2$ , respectively. In the case of the Indian Point-3 sample, the average leachability index of nickel was  $10.1 \pm 0.2$ , a value well below the average leachability index determined for  $^{63}\text{Ni}$ ,  $13.5 \pm 0.2$ . The ranges of the leachability indexes of iron were generally comparable to the corresponding ranges determined for  $^{55}\text{Fe}$ , while the ranges of the leachability indexes of cobalt were typically well below the corresponding ranges determined for  $^{60}\text{Co}$ .

## Release of Organic Acids from Waste Forms

This section presents the results of measurements of the release of organic acids from the seven solidified decontamination resin waste forms which were leach-tested according to the ANS 16.1 procedure. Releases of oxalic, citric, formic, and picolinic acids, EDTA, and DTPA were measured. Measurements were performed using the gas-liquid and ion chromatography methods described previously.

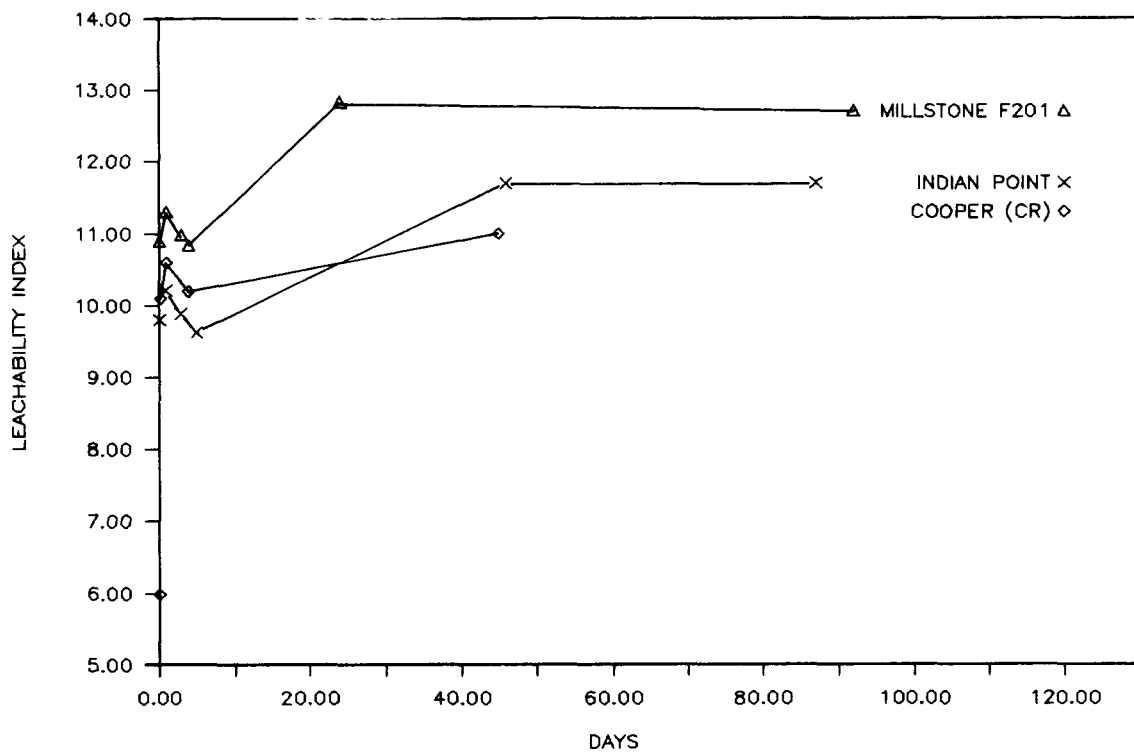
**Table 23. Release rates of stable metals**

Sample	Release Rate (F/cm <sup>2</sup> /s) <sup>a</sup>	
	Chromium	Iron
Millstone-1 F201	<5.0 (–10) to <1.1 (–12)	<3.0 (–11) to <6.5 (–14)
Pilgrim	— <sup>b</sup>	<8.5 (–12) to <1.8 (–10)
Indian Point-3	<1.1 (–9) to <2.1 (–12)	3.6 (–10) to <2.9 (–13)
Sample	Release Rate (F/cm <sup>2</sup> /s) <sup>c</sup>	
	Chromium	Iron
Millstone-1 F201	<5.5 (–8) to <1.2 (–10)	6.3 (–10) to 4.0 (–12)
Pilgrim	<4.9 (–9) to <1.8 (–10)	1.0 (–8) to 5.3 (–10)
Indian Point-3	— <sup>b</sup>	1.2 (–9) to 4.1 (–12)
Sample	Release Rate (F/cm <sup>2</sup> /s) <sup>c</sup>	
	Chromium	Iron
Cooper (MBR)	— <sup>b</sup>	<3.3 (–8) to <1.2 (–10)
Cooper (CR)	7.6 (–6) to 6.5 (–10)	1.9 (–5) to 1.8 (–10)
Sample	Release Rate (F/cm <sup>2</sup> /s) <sup>c</sup>	
	Cobalt	Nickel
Cooper (MBR)	— <sup>b</sup>	1.3 (–6) to <4.2 (–9)
Cooper (CR)	6.6 (–6) to <4.9 (–9)	1.0 (–5) to 2.9 (–10)

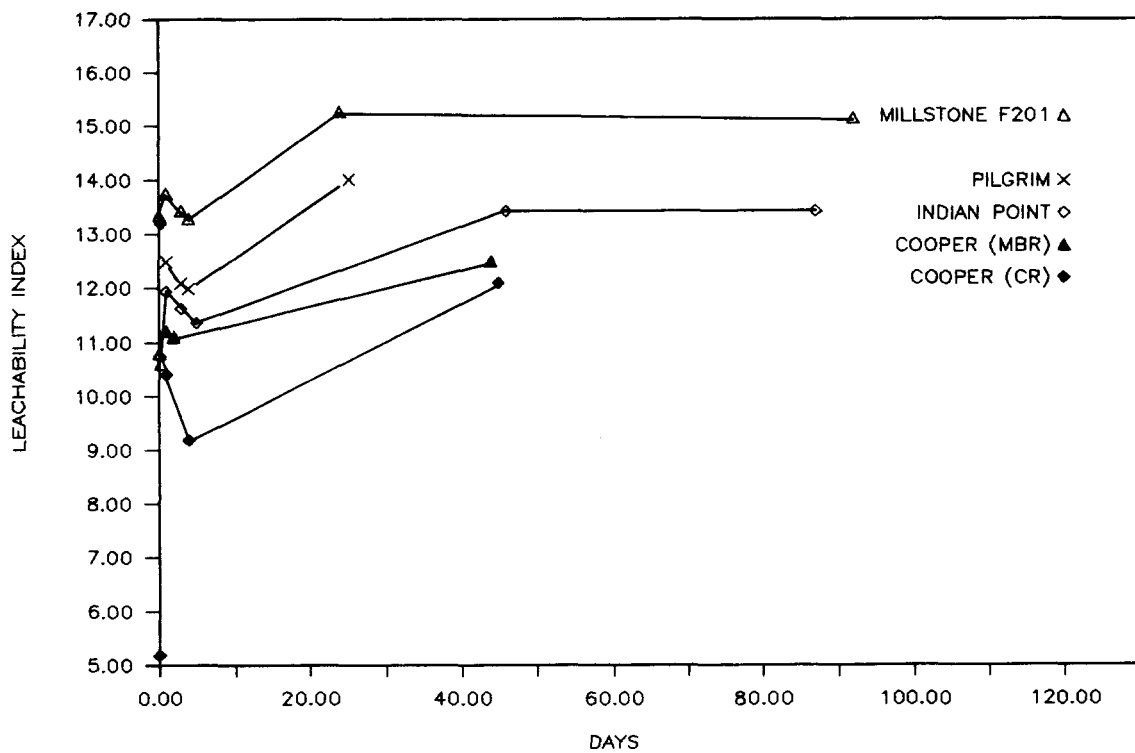
a. Fraction of initial inventory released per second per cm<sup>2</sup> of sample external surface area.

b. Analyses of resin waste and/or leachate samples were not performed.

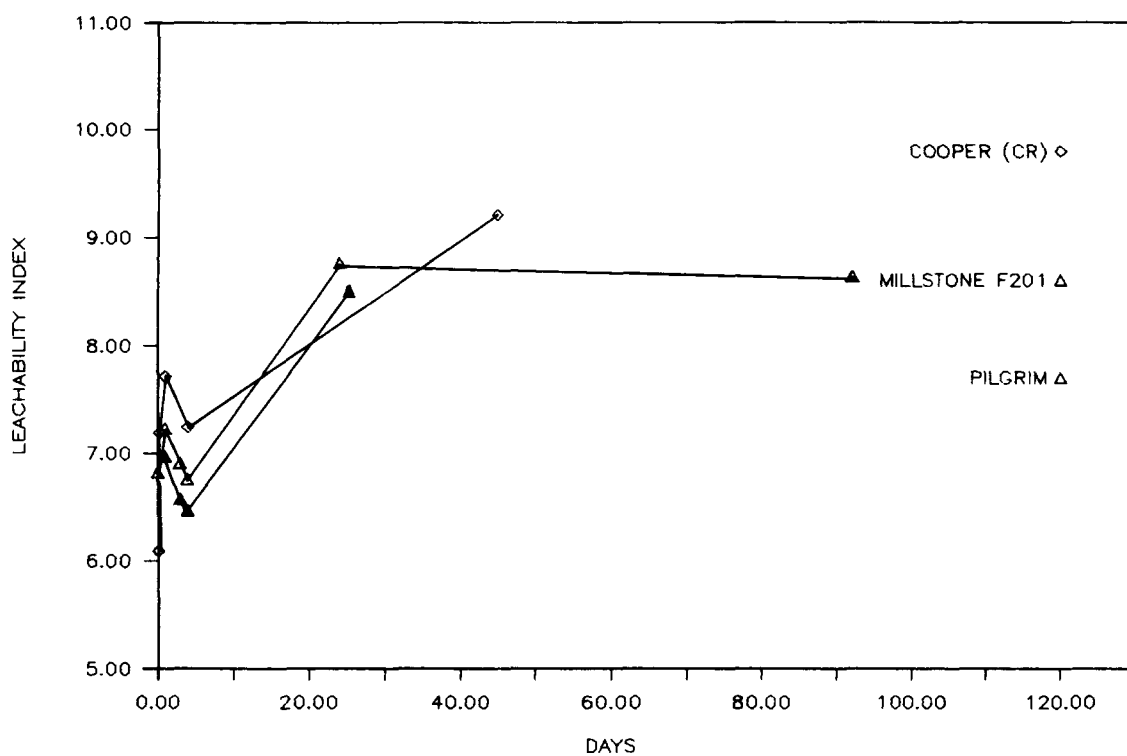
c. Fraction of initial inventory released per second.



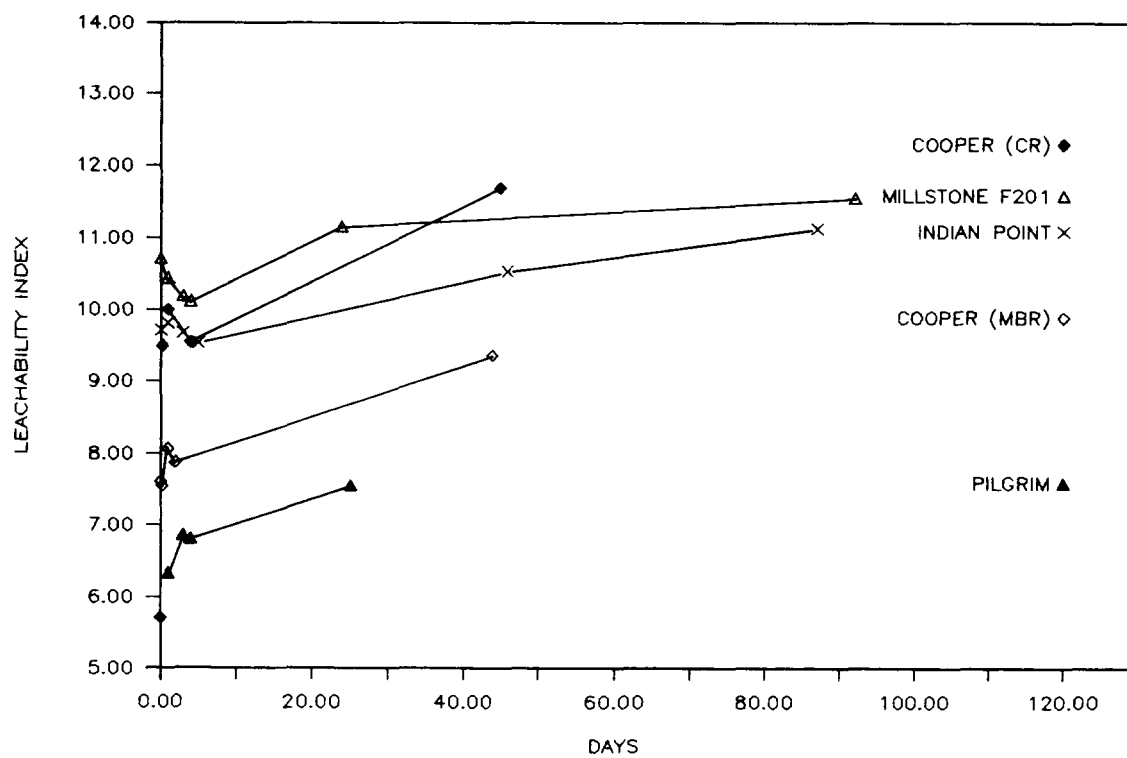
**Figure 38.** Leachability indexes for the release of chromium.



**Figure 39.** Leachability indexes for the release of iron.



**Figure 40.** Leachability indexes for the release of cobalt.



**Figure 41.** Leachability indexes for the release of nickel.

**Table 24. Average leachability indexes of stable metals**

Sample	Chromium	Iron	Cobalt	Nickel
Millstone-1 F33	— <sup>a</sup>	— <sup>a</sup>	— <sup>a</sup>	— <sup>a</sup>
Millstone-1 F201	>10.8 to >12.8	>13.3 to >15.3	>6.8 to >8.8	10.7 ± 0.2
Peach Bottom-2	— <sup>a</sup>	— <sup>a</sup>	— <sup>a</sup>	— <sup>a</sup>
Cooper (MBR)				
0 to 5 day	— <sup>a</sup>	>10.6 to >11.1	6.4 ± 0.1	7.6 to >8.1
5 to 92 day	— <sup>a</sup>	>12.5 <sup>b</sup>	8.0 <sup>b</sup>	>9.4 <sup>b</sup>
Cooper (CR)				
0 to 7 h	6.0 to >10.1	5.2 to >13.2	6.1 to >7.2	7.6 ± 2.0
7 h to 92 day	>10.2 to 11.0	10.6 ± 0.8	>7.2 to >9.2	10.4 ± 0.7
Pilgrim	— <sup>a</sup>	>12.0 to >14.0	>6.5 to >8.5	6.9 ± 0.3
Indian Point-3	>9.6 to >11.7	10.7 to >13.4	— <sup>c</sup>	10.1 ± 0.2

a. Analyses of resin waste and/or leachate samples were not performed.

b. Leachability index was determined for only one leachate sample.

c. Not detected in resin waste and/or leachate samples.

The concentrations of organic acids in leachates and their average release rates during individual leaching intervals are tabulated in Appendix F.

A summary of the leach-test results for the three Can-Decon waste-form samples is given in Table 25. For each sample, the cumulative quantity of each organic acid released during leaching, expressed as fraction of initial inventory, is presented in Table 25 along with the ranges of the measured effective diffusivities and leachability indexes. While all of the Millstone-1 F33 leachates were analyzed for organic acids, only three of the ten leachates generated during leach-testing of the Millstone-1 F201 waste form were analyzed for organic acids. The ranges of the effective diffusivities and leachability indexes for the Millstone-1 F201 sample were smaller than corresponding ranges for the other two Can-Decon samples because of the limited number of F201 leachates which were analyzed. The results for the Millstone-1 F33 and Peach Bottom-2 samples show that the effective diffusivities of a given organic acid typically varied by factors of 200 or more. The relatively large ranges in effective diffusivities reflect the fact that the rates at which organic acids were released decreased as cumulative leaching time

increased. This behavior is similar to that observed for radionuclides and stable metals.

A summary of the leach-test results for the Citrox, Dow NS-1, and LOMI waste forms is presented in Table 26. Because the concentrations of citric and oxalic acids in the Cooper cation resin waste were well below the concentrations of organic acids in other resin wastes, only a limited number of the Cooper cation resin waste-form leachates were analyzed for organic acids. (See Table 12 for resin waste organic acid concentrations.) The results presented in Tables 25 and 26 show that the effective diffusivities of organic acids in the Pilgrim and Indian Point-3 samples were much higher than the effective diffusivities determined for the other waste forms. While CFR values for oxalic and citric acids for the Millstone-1 F33, Peach Bottom-2, and Cooper mixed-bed samples were no more than about 2%, CFR results for the Pilgrim sample indicate that 34% of the oxalic acid and 40% of the citric acid originally present in the sample were released during leaching. The results also indicate that 100% of the EDTA and 33% of the DTPA inventories in the Pilgrim sample were released during leaching. The results for the Indian Point-3 sample indicate that 83% of the formic

**Table 25. Organic acid leach-test results for Can-Decon waste forms**

<u>Acid</u>	<u>Maximum CFR</u>	<u>Range Effective Diffusivity (cm<sup>2</sup>/s)</u>	<u>Range Leachability Index</u>
<u>Millstone-1 F33:</u>			
Oxalic	2.4 (– 2)	2.2 (– 12) to 4.4 (– 10)	9.4 to 11.7
Citric	1.4 (– 2)	5.3 (– 12) to 6.6 (– 10)	9.2 to 11.3
EDTA	2.3 (– 1)	1.3 (– 9) to 2.4 (– 8)	7.6 to 8.9
<u>Millstone-1 F201:</u>			
Oxalic	— <sup>a</sup>	2.6 (– 10) to 4.1 (– 10)	9.4 to 9.6
Citric	— <sup>a</sup>	1.0 (– 11) to 5.2 (– 10)	9.3 to 11.0
EDTA	— <sup>a</sup>	2.5 (– 9) to 1.3 (– 8)	7.9 to 8.6
<u>Peach Bottom-2:</u>			
Oxalic	1.8 (– 2)	1.8 (– 12) to 3.6 (– 10)	9.4 to 11.7
Citric	2.2 (– 2)	2.1 (– 12) to 1.9 (– 9)	8.7 to 11.7
EDTA	7.1 (– 2)	2.4 (– 12) to 7.4 (– 9)	8.1 to 11.6

a. Not all leachate samples were analyzed for this organic acid.

**Table 26. Organic acid leach-test results for Citrox, Dow NS-1, and LOMI waste forms**

<u>Acid</u>	<u>Maximum CFR</u>	<u>Range Effective Diffusivity (cm<sup>2</sup>/s)</u>	<u>Range Leachability Index</u>
<u>Cooper Mixed Bed Resin:</u>			
Oxalic	1.0 (– 2)	1.6 (– 12) to 1.5 (– 9)	8.8 to 11.8
Citric	8.4 (– 3)	4.8 (– 13) to 1.7 (– 9)	8.8 to 12.3
<u>Cooper Cation Resin:</u>			
Oxalic	— <sup>a</sup>	— <sup>a</sup>	— <sup>a</sup>
Citric	— <sup>b</sup>	5.0 (– 8) to 2.9 (– 7)	6.5 to 7.3
<u>Pilgrim:</u>			
Oxalic	3.4 (– 1)	2.3 (– 8) to 2.9 (– 7)	6.5 to 7.6
Citric	4.0 (– 1)	3.2 (– 8) to 2.4 (– 6)	5.6 to 7.5
EDTA	1.0 (+ 0)	1.9 (– 7) to 4.7 (– 6)	5.3 to 6.7
DTPA	3.3 (– 1)	7.6 (– 9) to 1.6 (– 7)	6.8 to 8.1
<u>Indian Point-3:</u>			
Formic	8.3 (– 1)	6.4 (– 8) to 2.9 (– 7)	6.5 to 7.2
Picolinic	6.0 (– 1)	2.5 (– 8) to 1.9 (– 7)	6.7 to 7.6

a. Acid was not detected in the leachate samples.

b. Not all leachates samples were analyzed for this organic acid.

acid and 60% of the picolinic acid originally in the waste form were released from the sample. The relatively narrow ranges of the effective diffusivities of the Pilgrim and Indian Point-3 samples reflect the fact that the rates of release of organic acids from these two samples remained comparatively high even during the longer leaching intervals.

The ranges of the rates of release of organic acids from the waste-form samples are presented in Table 27. Results for the Cooper waste forms are expressed as fraction of initial inventory released per second (F/s), while the release rates for other waste forms are expressed as fraction of initial inventory, F, released per second per square centimeter of sample external surface area (F/cm<sup>2</sup>/s). In

all cases, maximum release rates occurred during the first or second leaching intervals; and, in most cases, minimum release rates occurred during the last leaching interval. As shown in Table 27, the rates of release of acids from the Pilgrim sample during the first 2-h-long leaching interval were factors of 10 or more higher than their corresponding rates of release from the Millstone-1 and Peach Bottom-2 samples. Initial rates of release of formic and picolinic acids from the Indian Point-3 sample were comparable to the initial organic acid release rates measured for the Pilgrim sample.

The organic acid CFR data for the Millstone-1 F33, Peach Bottom-2, Cooper mixed-bed resin, Pilgrim, and Indian Point-3 waste-form samples

**Table 27. Release rates of organic acids**

Sample	Release Rate <sup>a</sup> (F/cm <sup>2</sup> /s)		
	Oxalic	Citric	EDTA
Millstone-1 F33	2.9 (– 9) to 3.9 (– 12)	4.7 (– 10) to 7.7 (– 12)	5.6 (– 9) to 1.0 (– 10)
Millstone-1 F201	1.0 (– 9) to 1.6 (– 10)	1.6 (– 10) to 5.7 (– 11)	2.5 (– 9) to 6.0 (– 10)
Peach Bottom-2	8.6 (– 10) to 1.9 (– 12)	2.0 (– 9) to 2.0 (– 12)	1.5 (– 9) to 2.2 (– 12)
Pilgrim <sup>b</sup>	2.5 (– 8) to 1.2 (– 10)	4.4 (– 8) to 3.3 (– 11)	6.9 (– 8) to 4.7 (– 11)
Indian Point-3	Formic	Picolinic	
	2.1 (– 8) to 5.2 (– 10)	1.3 (– 8) to 3.4 (– 10)	
	Release Rate <sup>c</sup> (F/s)		
Cooper (MBR)	Oxalic	Citric	
	3.1 (– 7) to 2.0 (– 10)	3.3 (– 7) to 9.8 (– 11)	

a. Fraction of initial inventory released per second per cm<sup>2</sup> of sample external surface area.

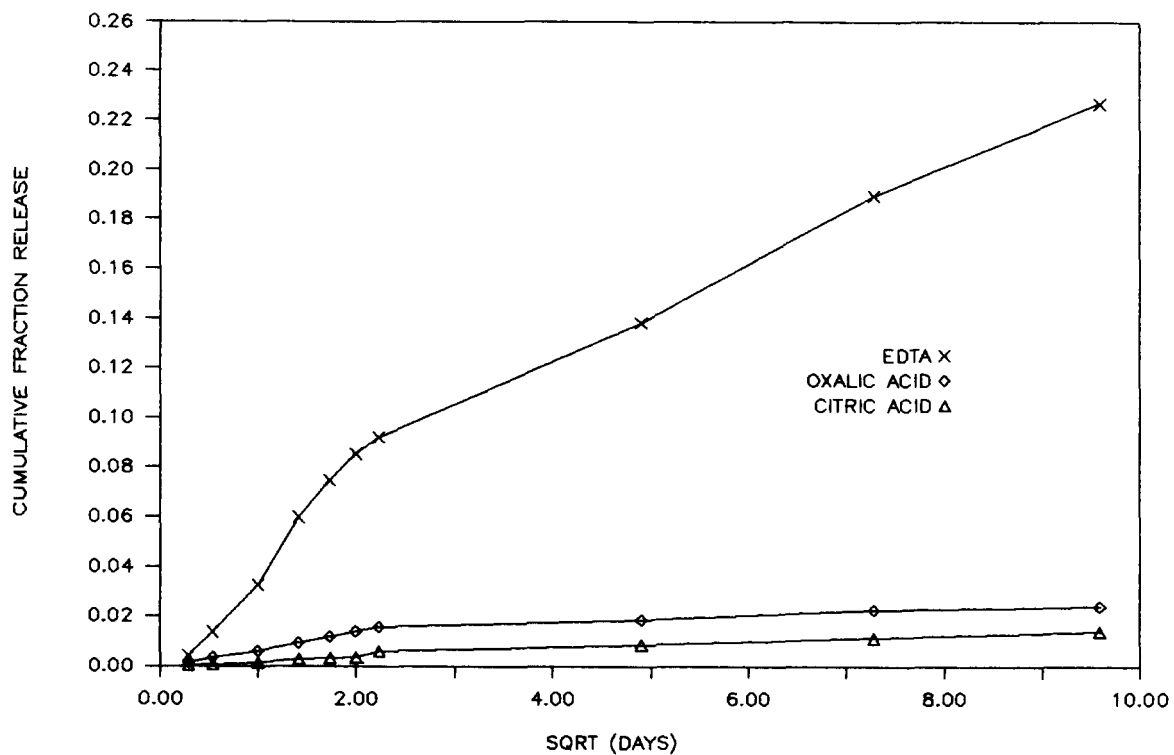
b. Pilgrim DTPA release rates were: 9.9 (– 9) to 1.3 (– 10) F/cm<sup>2</sup>/s.

c. Fraction of initial inventory released per second.

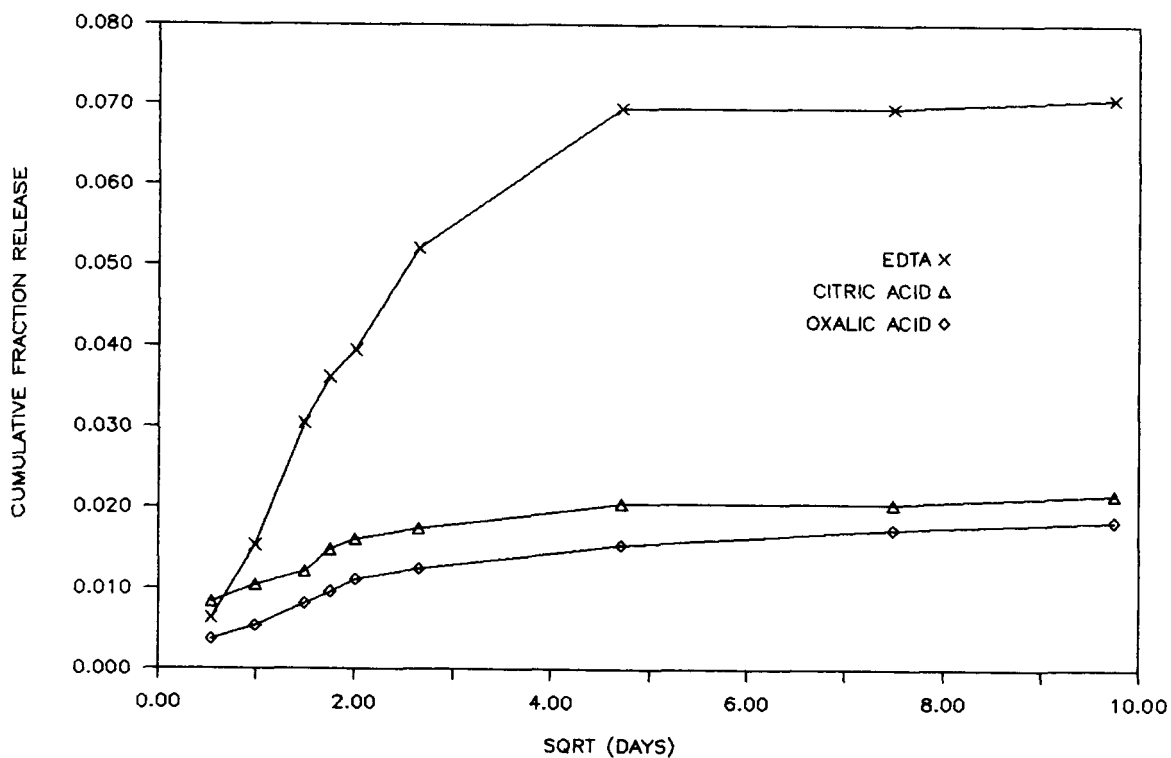
are plotted as a function of the square root of elapsed leaching time in Figures 42 through 46, respectively. According to Equation (2), the CFR data will be linear with  $t^{1/2}$  if diffusion is the controlling release mechanism. As shown in Table 25 and Figures 42 and 43, considerably more EDTA was released from the Can-Decon waste forms than oxalic or citric acid. The cumulative quantities of EDTA released from the Millstone-1 F33 and Peach Bottom-2 samples corresponded to 23% and 7%, respectively, of their original inventories of EDTA. Similar CFR values for oxalic and citric acids were, respectively, only 2.4% and 1.4% for the Millstone-1 F33 sample and 1.8% and 2.2%, respectively, for the Peach Bottom-2 waste form. Thus the fractions of the initial inventories of oxalic and citric acids released from the two Can-Decon samples were similar, but substantially less EDTA was released from the Peach Bottom-2 sample compared to the quantity released from the Millstone-1 F33 sample.

As was the case with releases of radionuclides from the Millstone-1 F33 sample, the release rates of organic acids were highest during the first 5 days of leaching. Leaching intervals were no longer than 1 day during this period, while subsequent leaching intervals were 19, 29, and 39 days long. Of the total quantity of EDTA released from the sample after 92 days of leaching, about 41% of that amount was released after only 5 days of leaching. Similar values for oxalic and citric acids after 5 days of leaching were 66% and 43%, respectively. The trends in the CFR data shown in Figure 42 were similar to the trends in the radionuclide CFR data for this sample, which were shown in Figures 13 and 14. The results indicate that about 26% of the total quantity of organic acids in the waste form at the start of leaching was ultimately released from the sample. Figure 16 shows that about 96% of the <sup>137</sup>Cs inventory was leached from the sample. As a monovalent alkali metal, cesium forms only weak complexes with organic acids; and, in the presence

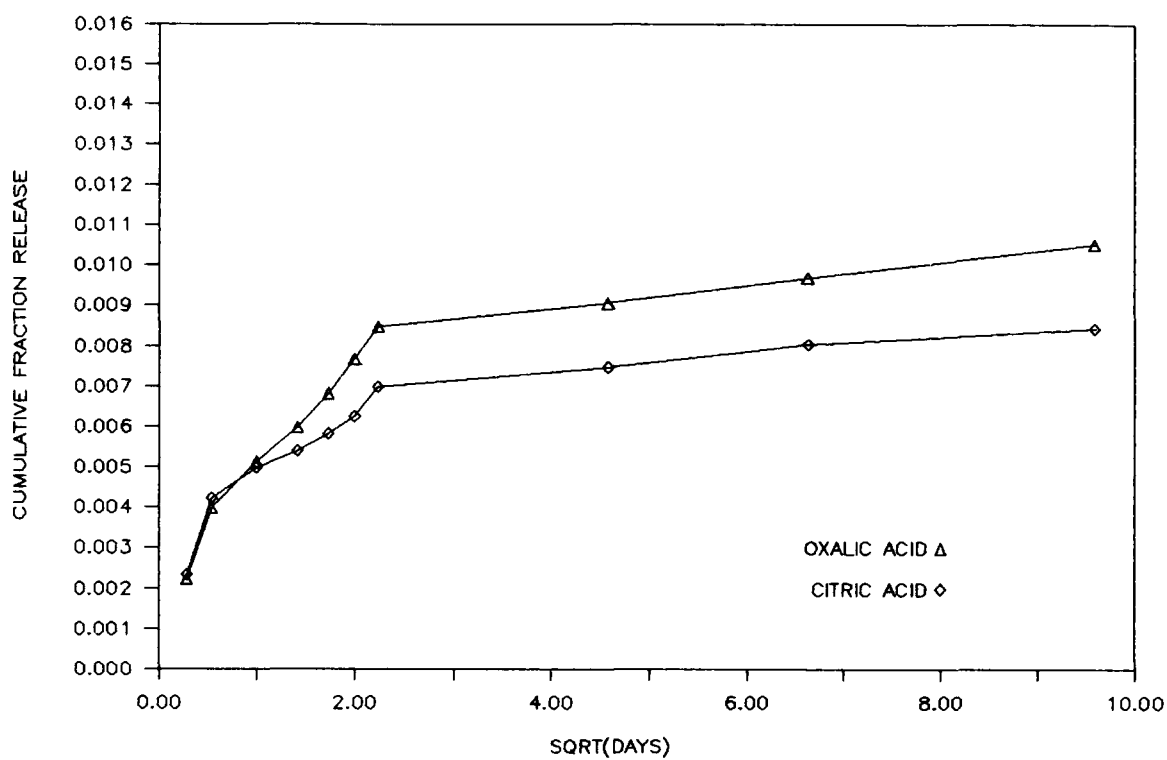




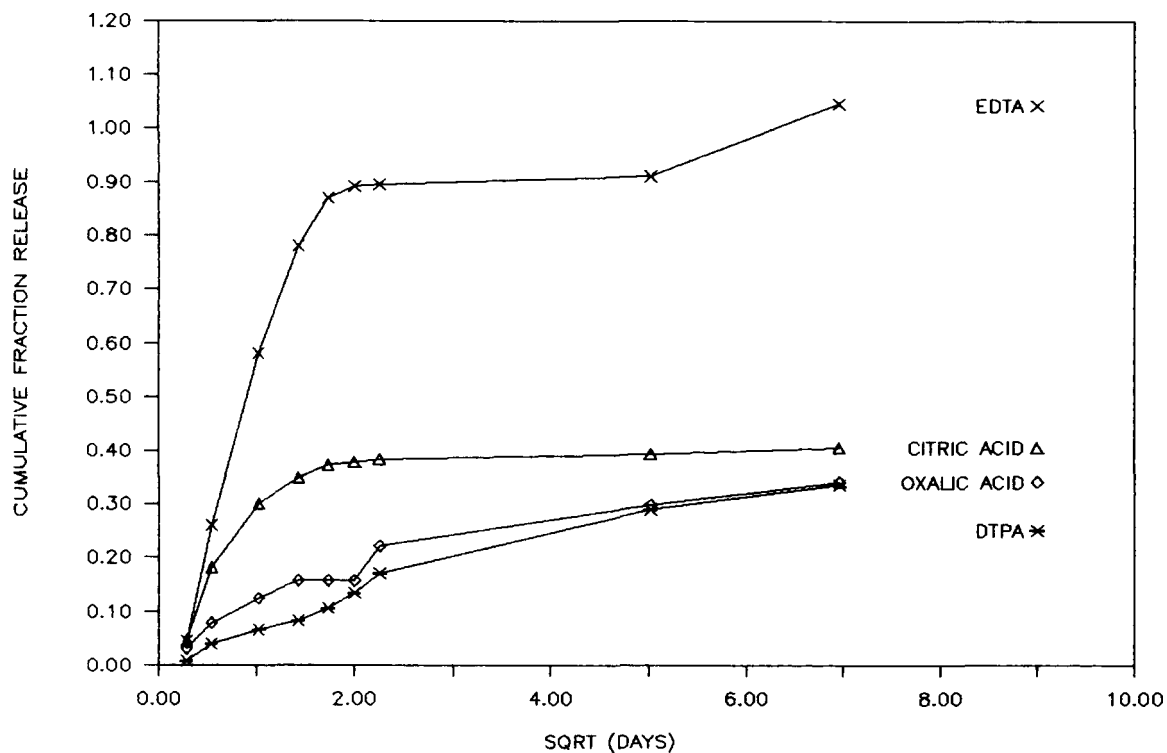
**Figure 42.** Cumulative fraction release of oxalic acid, citric acid, and EDTA from the Millstone-1 F33 Can-Decon waste form.



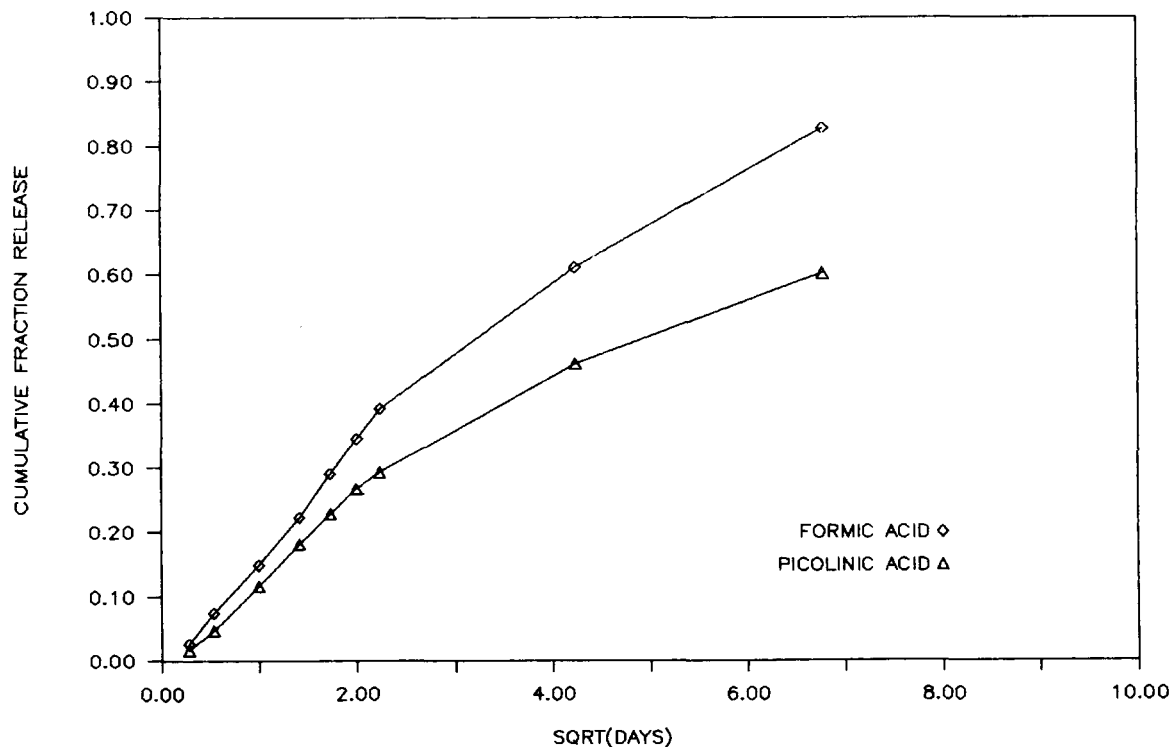
**Figure 43.** Cumulative fraction release of oxalic acid, citric acid, and EDTA from the Peach Bottom-2 Can-Decon waste form.



**Figure 44.** Cumulative fraction release of oxalic and citric acids from the Cooper mixed-bed resin Citrox waste form.



**Figure 45.** Cumulative fraction release of oxalic acid, citric acid, EDTA, and DTPA from the Pilgrim Dow NS-1 waste form.



**Figure 46.** Cumulative fraction release of formic and picolinic acids from the Indian Point-3 LOMI waste form.

of water, cesium is readily dissociated from such complexing agents. The differences in the releases of  $^{137}\text{Cs}$  and organic acids confirm that  $^{137}\text{Cs}$  was released from the waste form as an uncomplexed soluble species.

In the case of the Peach Bottom-2 sample, the region of relatively rapid releases of oxalic and citric acids extended to 4 days, while relatively rapid releases of EDTA continued through 22 days. After 22 days, the releases of EDTA and citric acid essentially stopped. For this sample, the sixth leaching interval was 3 days long rather than the customary 1-day length. Figure 43 shows that while the release rates of citric and oxalic acids decreased during the sixth leaching interval, the rate of release of EDTA actually increased. The sharp drop in leachate pH which was measured during the third leaching interval is not reflected in the CFR data of the organic acids. The trends in the CFR data for  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ , which were shown in Figures 17 and 18, respectively, appear to be uncorrelated to the trends in the EDTA releases.

As shown in Table 26 and Figure 44, about 1.0% and 0.8% of the original inventories of oxalic and citric acids were released from the Cooper mixed-bed resin waste form after 92 days of continuous

leaching. Approximately 80% of the total amount of oxalic acid released from the sample was released during the first 5 days of leaching, and about 83% of the total quantity of citric acid released from the sample was released during the same period. Comparing Figure 44 with Figures 19 and 20, which show the radionuclide CFR data for the Cooper mixed-bed resin waste form, it is apparent that the trends in both sets of data are similar. Both the radionuclide and organic acid CFR data are characterized by relatively rapid releases during the first 5 days of leaching, followed by slower but continuous releases throughout the remaining 87 days of leaching. The CFRs of oxalic and citric acids were each 1% or less at the end of leaching, but the CFR of  $^{137}\text{Cs}$  was 100%, indicating that  $^{137}\text{Cs}$  was released from the waste form as an uncomplexed soluble species.

The results presented in Table 26 indicate that between 30% and 40% of the inventories of oxalic acid, citric acid, and DTPA were released from the Pilgrim waste form during 48 days of continuous leaching, and 100% of the inventory of EDTA was lost from the sample during the same period. The CFR data shown in Figure 45 indicate that the rate of release of EDTA from the Pilgrim waste form

was relatively rapid during the first 3 days of leaching. Approximately 84% of the total amount of EDTA released from the sample was lost during this time period. Release of EDTA essentially stopped between 3 days and 25 days, but resumed after 25 days. The period of very low EDTA releases corresponds to a period when the leachate pH remained below 11 (see Figure 28), presumably because less  $\text{Ca}(\text{OH})_2$  was being released to the leachant. The behavior of the release of citric acid from the Pilgrim waste form was similar to that of EDTA except that the citric acid release rate dropped off sooner. About 86% of the total amount of citric acid lost from the waste form was released by the end of the second day of leaching. Contrasting the behavior of EDTA, the release of citric acid did not increase significantly during the 25-to-48-day leaching interval. The CFR profiles of oxalic acid and DTPA were similar in the respect that after 3 days of leaching their release rates did not decrease as much as those of EDTA and citric acid.

The trends in the  $^{137}\text{Cs}$  CFR data for the Pilgrim waste form, which were shown in Figure 24, closely duplicated the trends in the EDTA CFR data for the Pilgrim sample. However, the results indicate that while the entire inventory of EDTA was released from the waste form, only about 0.4% of the initial inventory of  $^{137}\text{Cs}$  was leached from the sample. The results suggest that  $^{137}\text{Cs}$  was released from the waste form as an uncomplexed soluble species. While the releases of  $^{137}\text{Cs}$ , EDTA, and citric acid were minimal between 3 and 25 days, the relative releases of  $^{60}\text{Co}$  were significant during this period. Approximately 43% of the total quantity of  $^{60}\text{Co}$  released from the waste form was released between 3 and 25 days after leaching commenced.

The cumulative quantities of formic and picolinic acids leached from the Indian Point-3 waste form are plotted in Figure 46. As explained previously, the concentration of formic acid in the resin waste was not measured because the gas-liquid chromatography method used to quantify the concentrations of organic acids in the resin wastes could not be used to detect formic acid. The concentration of formic acid was assumed to be the same as that of picolinic acid based on typical conditions for experimental decontaminations described in Reference 31. The results in Table 26 and Figure 46 indicate that about 83% and 60%, respectively, of the initial inventories of formic and picolinic acids were released from the waste form during 46 days of continuous leaching. As usual, the CFR plots of these acids exhibited relatively

steep slopes during the first 5 days of leaching. However, unlike the organic acid CFR data for many of the waste forms, significant releases occurred after 5 days of leaching. The results indicate that about one-half of the total quantity of each acid released was released between 5 and 46 days after the start of leaching.

The trends in the CFR profiles of formic and picolinic acids closely resemble those in the CFR profiles of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  for the Indian Point-3 waste form (see Figures 25 and 26). At the end of 46 days of leaching, the CFR of  $^{137}\text{Cs}$  was comparable to the CFR of formic acid, their respective values being 90% and 83%. However, while the majority of the inventories of  $^{137}\text{Cs}$  and formic and picolinic acids were released from the waste form by the end of 46 days, only about 0.1% of the original quantity of  $^{60}\text{Co}$  was released from the sample during this same time. At the end of the eighth leaching interval, leachate pH was 11.9; but it decreased to 10.4 at the end of the ninth leaching interval and to 8.7 at the end of the tenth leaching interval. The regions of lower pH correspond to regions of lower release rates for both radionuclides and organic acids.

The leachability indexes of oxalic acid, citric acid, EDTA, DTPA, and formic and picolinic acids are plotted as a function of elapsed leaching time in Figures 47 through 51, respectively. (Data for the Millstone-1 F201 and Cooper cation resin waste forms are not shown because of the limited number of leachability indexes determined for these two samples.) Figures 47 through 49 show that oxalic acid, citric acid, and EDTA were released much more readily from the Pilgrim sample than from the other waste-form samples. This difference in leaching behavior is similar to that observed for  $^{60}\text{Co}$  (see Figure 31). The Pilgrim waste form leachability indexes of oxalic and citric acids ranged from 6.5 to 7.6 and from 5.6 to 7.5, respectively; while the leachability indexes of these same acids for the Citrox and Can-Decon waste forms ranged from 8.8 to 11.8 and from 8.7 to 12.3, respectively. Similarly, for EDTA the Pilgrim leachability indexes ranged from 5.3 to 6.7, while the EDTA leachability indexes of the three Can-Decon samples ranged from 7.6 to 11.6. The leachability indexes of oxalic and citric acids for the Citrox and Can-Decon samples were comparable throughout 3 months of leaching. The EDTA leachability indexes of the Millstone-1 F33 and Peach Bottom-2 waste forms agreed quite well through 24 days of leaching but diverged during the last leaching interval.

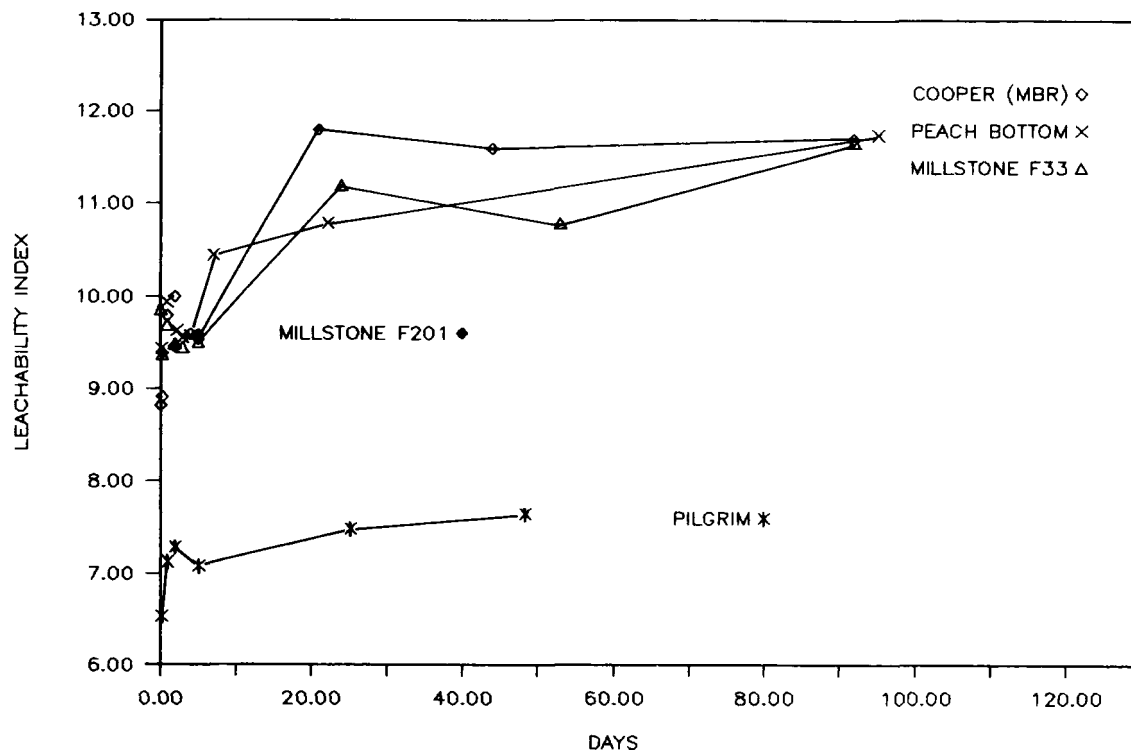


Figure 47. Leachability indexes for the release of oxalic acid.

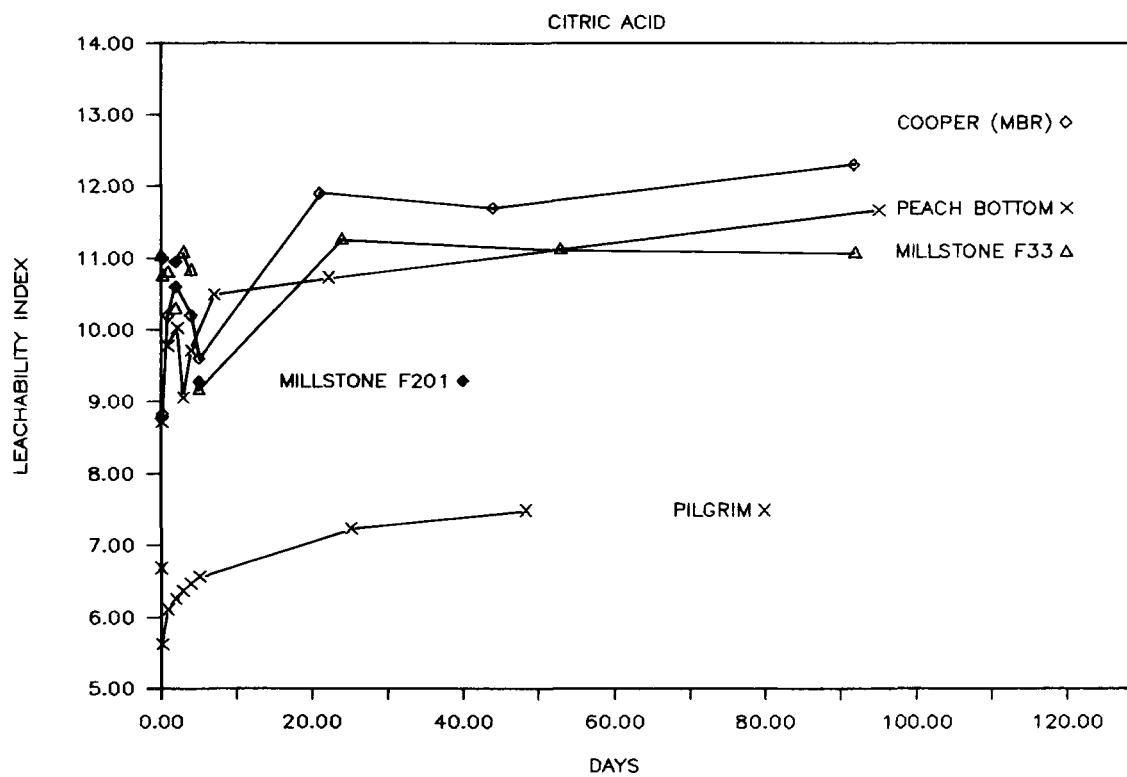
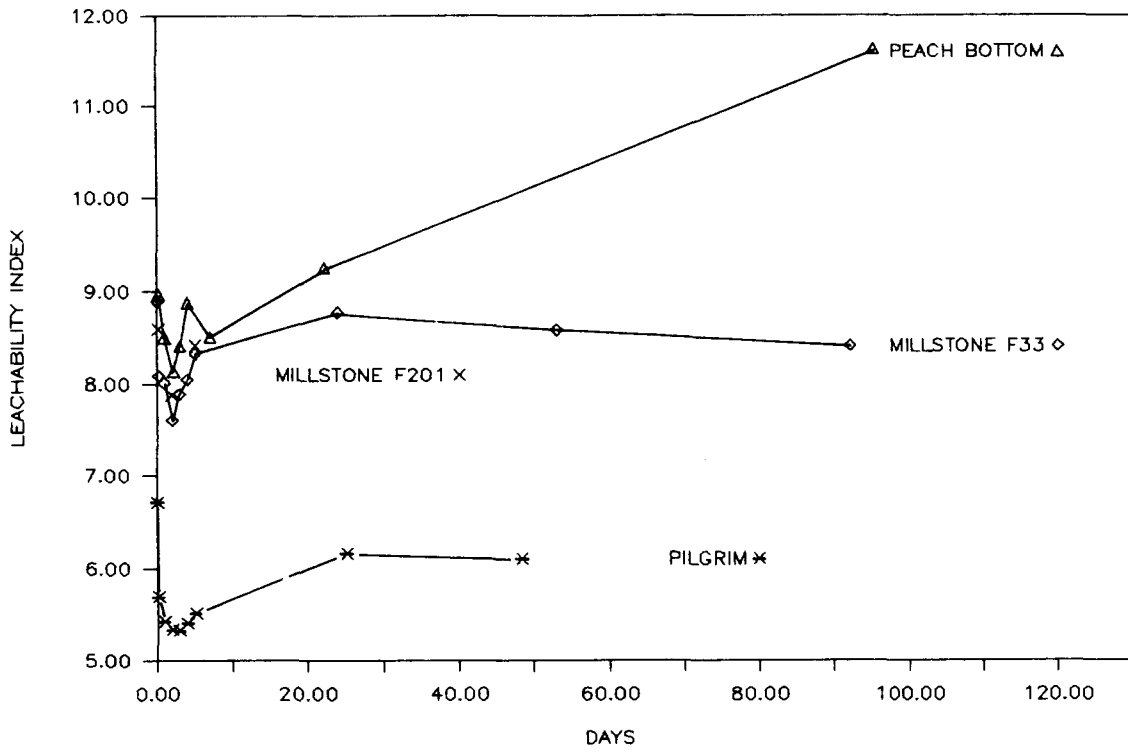
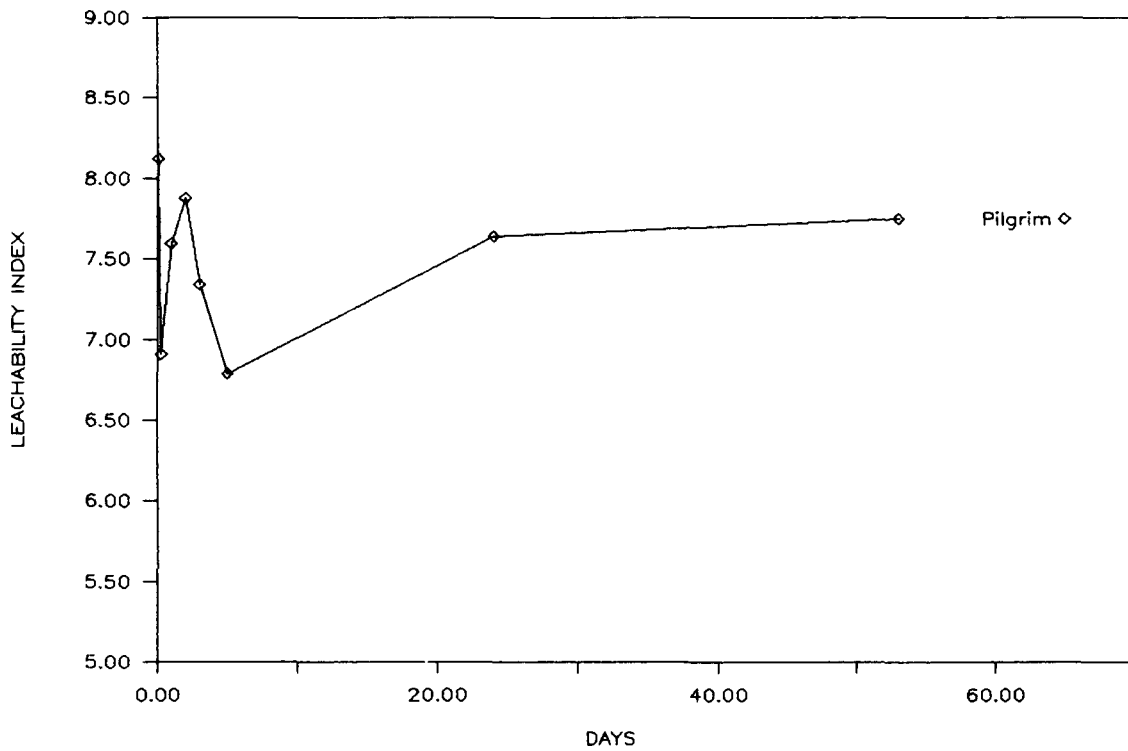


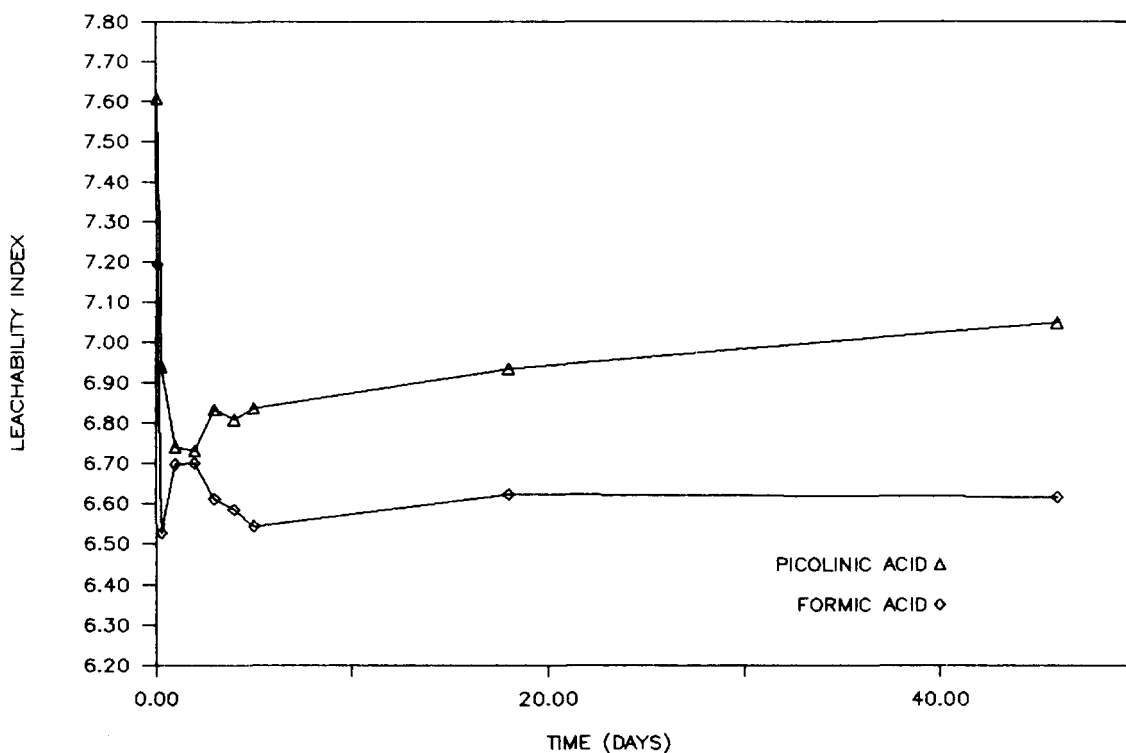
Figure 48. Leachability indexes for the release of citric acid.



**Figure 49.** Leachability indexes for the release of EDTA.



**Figure 50.** Leachability indexes for the release of DTPA.



**Figure 51.** Leachability indexes for the release of formic and picolinic acids.

The valley and peak evident in the Indian Point-3 picolinic and formic acid leachability indexes, which occurred during the second and fourth leaching intervals, coincide with a peak in the leachate pH (see Figure 27). Leachate pH was about 11.8 during this period. The opposite trends in the leachability indexes of picolinic and formic acids during the first week of leaching indicate that, at least during initial leachings, the releases of these two acids are affected in opposite ways by changes in leachate pH. The decrease in leachate pH that was observed between the eighth and ninth leaching intervals was accompanied by a slight increase in the leachability index of picolinic acid and no change in the leachability index of formic acid.

Average leachability indexes of EDTA, DTPA, and oxalic, citric, formic, and picolinic acids are presented in Table 28 for each of the seven waste-form samples that were leach-tested. Uncertainties given in Table 28 are one-sigma standard deviations of the mean. Because both Cooper samples decomposed during initial phases of leaching, organic acid leachability indexes of these waste forms were calculated using the same assumptions described previously. Average leachability indexes for the two

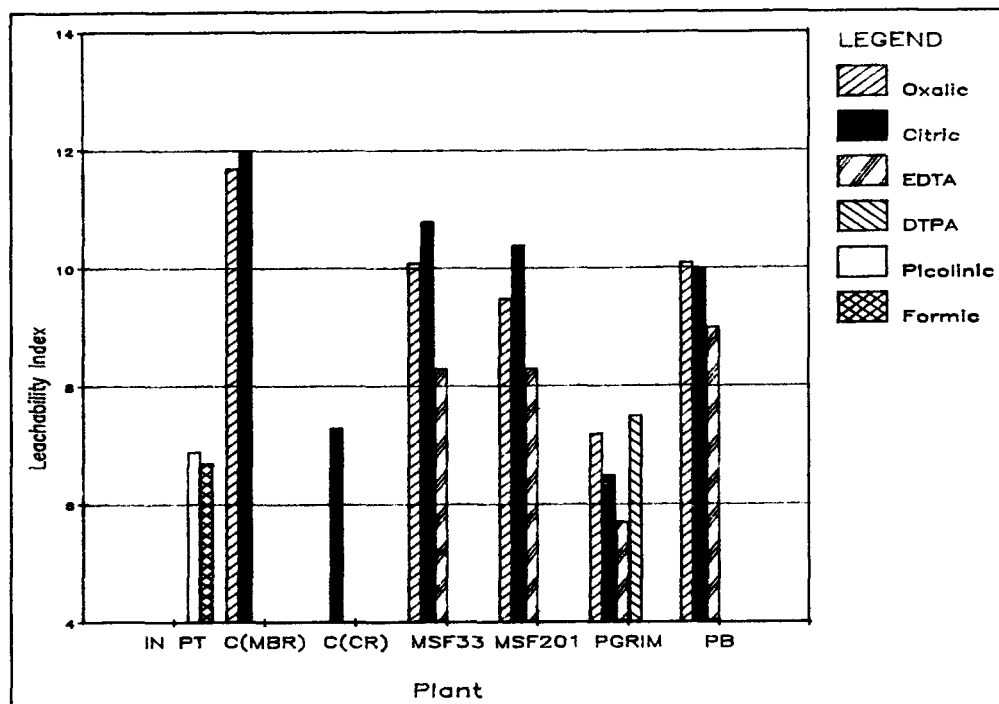
Cooper samples were determined for two different time regions, one corresponding to the period during which decomposition occurred and the second encompassing the time region characterized by lower release rates. Results are presented graphically in Figure 52. The data shown in Table 28 and Figure 52 show that the average leachability indexes of organic acids contained in the Can-Decon waste forms agreed quite well. Average leachability indexes of oxalic acid, citric acid, and EDTA were  $10.1 \pm 0.3$ ,  $10.8 \pm 0.2$ , and  $8.3 \pm 0.1$ , respectively, for the Millstone-1 F33 sample, while similar results for the Peach Bottom-2 sample were  $10.1 \pm 0.3$ ,  $10.0 \pm 0.3$ , and  $9.0 \pm 0.4$ , respectively. Results for the Cooper mixed-bed resin Citrox sample were comparable to corresponding results for the Can-Decon samples. Leachability indexes of oxalic and citric acids during initial leaching intervals were slightly lower, and leachability indexes of these acids during later leaching intervals were somewhat higher than corresponding average leachability indexes for the three Can-Decon samples. Average leachability indexes of oxalic and citric acids contained in the Pilgrim sample were only  $7.2 \pm 0.1$  and  $6.5 \pm 0.1$ , respectively. The results for formic and

**Table 28. Average leachability indexes of organic acids**

Sample	Oxalic	Citric	EDTA	DTPA
Millstone-1 F33	10.1 ± 0.3	10.8 ± 0.2	8.3 ± 0.1	— <sup>a</sup>
Millstone-1 F201	9.5 ± 0.1	10.4 ± 0.5	8.3 ± 0.2	— <sup>a</sup>
Peach Bottom-2	10.1 ± 0.3	10.0 ± 0.3	9.0 ± 0.4	— <sup>a</sup>
Cooper (MBR)				
0 to 5 day	9.4 ± 0.2	9.7 ± 0.3	— <sup>a</sup>	— <sup>a</sup>
5 to 92 day	11.7 ± 0.1	12.0 ± 0.1	— <sup>a</sup>	— <sup>a</sup>
Cooper (CR)				
0 to 7 h	— <sup>a</sup>	6.5 <sup>b</sup>	— <sup>a</sup>	— <sup>a</sup>
7 h to 92 day	— <sup>a</sup>	7.3 <sup>b</sup>	— <sup>a</sup>	— <sup>a</sup>
Pilgrim	7.2 ± 0.1	6.5 ± 0.2	5.7 ± 0.2	7.5 ± 0.2
	Formic	Picolinic		
Indian Point-3	6.7 ± 0.1	6.9 ± 0.1		

a. Not detected in resin waste and/or leachate samples.

b. Leachability index was determined for only one leachate sample.



**Figure 52. Average leachability indexes of organic acids.**



picolinic acids contained in the Indian Point-3 waste form were  $6.7 \pm 0.1$  and  $6.9 \pm 0.1$ , respectively.

A recent study<sup>10</sup> conducted at BNL determined the releases of organic complexing agents from solidified simulated decontamination resin wastes. Simulated resin wastes were made by loading various commercially available resins with different combinations of organic acids. The loaded resins were solidified in Portland I cement and leach-tested according to the ANS 16.1 procedure. The results of this BNL study are summarized in Table 29.

The results presented in Tables 28 and 29 show that the average leachability indexes of oxalic and citric acids contained in the Can-Decon and Citrox waste forms collected from operating LWRs agree reasonably well with the corresponding results for the simulated resin waste forms. However, the average leachability indexes of EDTA determined using actual resin waste forms were generally lower than the corresponding values determined using simulated samples. The average leachability indexes determined for formic and picolinic acids were also lower than the corresponding leachability indexes measured using simulated waste form samples.

**Table 29. Leaching results for simulated resin waste forms<sup>10</sup>**

Reagents <sup>a</sup> in Form	Average Leachability Index		
	Oxalic	Citric	EDTA
EOC	11.2 to 11.9	9.8 to 10.5	10.5 to 10.6
EOC	10.8 to 11.0	9.7 to 9.9	10.5 to 10.6
	Formic	Picolinic	
LOMI	8.3	8.8	
LOMI	8.1 to 8.4	8.1 to 8.6	

a. EOC = combination of EDTA, oxalic acid, and citric acid. LOMI = combination of formic and picolinic acids.

## CONCLUSIONS AND RECOMMENDATIONS

### Concentrations of Radionuclides, Stable Metals, and Organic Acids in Resin Wastes

Of the radionuclides that were detected in the resin wastes,  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{63}\text{Ni}$  were the major activities. Concentrations of these radionuclides ranged from about 0.1 to  $20\ \mu\text{Ci}/\text{cm}^3$  of tap-settled, wet resin. Cobalt-60 made up from 24% to 59%,  $^{55}\text{Fe}$  from 15% to 62%,  $^{63}\text{Ni}$  from 3% to 43%, and  $^{54}\text{Mn}$  from 4% to 8% of the total radionuclide inventories of the resins. The concentrations of  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  were, on the other hand, relatively low, their maximum concentrations being  $5.9 \times 10^{-4}$ ,  $1.6 \times 10^{-1}$ , and  $1.3 \times 10^{-1}\ \mu\text{Ci}/\text{cm}^3$  of wet resin, respectively. Among the transuranics,  $^{241}\text{Pu}$  exhibited the highest activity;  $^{241}\text{Pu}$  concentrations ranged from  $5 \times 10^{-3}$  to  $3.5 \times 10^{-1}\ \mu\text{Ci}/\text{cm}^3$ . The concentrations of transuranics in the resin waste obtained from Millstone-1 were typically an order of magnitude higher than corresponding concentrations in other resin wastes which, excluding  $^{241}\text{Pu}$ , normally ranged from  $1 \times 10^{-6}$  to  $1 \times 10^{-4}\ \mu\text{Ci}/\text{cm}^3$ . Omitting  $^{58}\text{Co}$ , which has a half-life of only 70.9 days, total radionuclide concentrations in the resins ranged from about 1.7 to  $31.7\ \mu\text{Ci}/\text{cm}^3$ . Excluding the result for the mixed-bed resin obtained from Cooper Station (its radionuclide concentrations were much lower than those of the other resin waste samples), the average loading on the remaining five resin waste samples was  $17 \pm 4\ \mu\text{Ci}/\text{cm}^3$ .

Iron and nickel were the major transition metals in the resin wastes, while chromium and cobalt were present at trace levels. Concentrations of iron ranged from about  $5 \times 10^2$  to  $1 \times 10^4\ \mu\text{g}/\text{cm}^3$  wet resin, while concentrations of nickel ranged from  $7.4 \times 10^1$  to about  $2 \times 10^3\ \mu\text{g}/\text{cm}^3$  wet resin. Maximum concentrations of chromium and cobalt were  $3.5 \times 10^2$  and  $6 \times 10^1\ \mu\text{g}/\text{cm}^3$ , respectively.

Concentrations of organic reagents in resin wastes were measured using GLC. The ranges of concentrations of organic acids in the resins, expressed as  $\mu\text{g}/\text{cm}^3$  of tap-settled, wet resin, were measured to be: oxalic acid,  $2.7 \times 10^2$  to  $1.6 \times 10^4$ ; citric acid,  $9.6 \times 10^1$  to  $2.5 \times 10^4$ ; and EDTA,  $3.7 \times 10^3$  to  $6.1 \times 10^3$ . The concentration of DTPA in the resin collected from Pilgrim Station was  $3.6 \times 10^2\ \mu\text{g}/\text{cm}^3$ , and the concentration of picolinic acid in the resin obtained from Indian Point-3

Station was  $4.1 \times 10^3\ \mu\text{g}/\text{cm}^3$ . The total reagent loadings of the resin wastes ranged from 0.04 wt. % for the cation resin sample obtained from Cooper Station to 4.1 wt. % for the mixed-bed resin sample obtained from Cooper Station. The reagent loadings of other resins were typically between 1 and 2 wt. %.

### Waste-Form Stability During Leaching

Of the seven cement-solidified resin waste-form samples which were leach-tested in deionized water, two completely decomposed into loose granular rubble within 8 h following immersion in leachant and two others deteriorated to a lesser extent over the course of 90 days of leaching. The samples which completely decomposed were the cation and mixed-bed resin waste forms collected from Cooper Station. The primary coolant recirculation system at Cooper had been decontaminated using the AP/Citrox process, which employs alkaline permanganate, citric acid, and oxalic acid. The decontamination solution was cycled through a cation resin bed during the decontamination operation; and following the completion of decontamination, it was processed through mixed-bed resin. The two types of resin were solidified separately.

The two samples which suffered modest deterioration were the duplicate samples obtained from the Millstone-1 Station. They lost no more than 5% of their masses during leaching; but the leaching results indicate that in addition to spallation, cracking also occurred. Millstone-1 Station had been decontaminated using the Can-Decon process, which employs citric acid, oxalic acid, and EDTA. The observed deterioration of the Millstone-1 waste forms might be the result of the condition of the cement/resin paste at the time of their collection. The cement/resin mixture in the solidification liner had begun to set up when the samples were being collected; and as result, following curing, the surfaces of the two waste forms were coarser than usual and their upper ends were concave and irregular.

Studies<sup>10,11</sup> conducted at BNL using simulated waste forms have indicated that the stability of cement-solidified decontamination resin waste forms is dependent on a number of factors, including types of resins and reagents solidified,

waste-to-binder ratios used during solidification, and aging time prior to leach-testing. One of these studies<sup>10</sup> demonstrated that simulated cement-solidified waste forms containing oxalic acid, citric acid, and EDTA loaded on anion exchange resin did not withstand immersion testing, yet mixed-bed resins containing the same reagents did pass immersion tests. During the same study, three of five replicate waste forms containing citric acid deteriorated so badly during leaching in deionized water that no recognizable form remained. Failure mechanisms have been suggested<sup>11</sup> that include the swelling of cation resin beads, as monovalent ions (e.g.,  $H^+$ ,  $Na^+$ ) attached to the beads are replaced by higher-valent cations present in the cement (e.g.,  $Ca^{+2}$  and  $Al^{+3}$ ). Also, the release of EDTA has been shown to cause anion resin beads to swell. For waste forms containing picolinic acid, the release of this acid produced the reverse effect, causing anion resin beads to shrink.

The complete decompositions of the Cooper Citrox waste forms following immersion in deionized water might have been caused by different failure mechanisms. The Cooper mixed-bed resin contained relatively high concentrations of citric acid (i.e.,  $2.5 \times 10^4 \mu g/cm^3$ ). The citric acid might have combined with calcium in the cement to form insoluble calcium citrate precipitates. The formation of calcium citrate would have interfered with the hydration of the cement, producing a waste form lacking structural stability. The failure of the Cooper cation resin waste form, which contained relatively small quantities of reagents, may have been due to the swelling of cation resin beads due to the ion-exchange processes previously described.

## Release of Radionuclides from Waste Forms

The leaching results show that, with the exception of  $^{137}Cs$ , only very small fractions of the inventories of the radionuclides contained in the decontamination resin waste forms were released from the forms during leach-testing. The total quantity of  $^{54}Mn$  released from any waste form ranged from less than 0.005% to less than 0.2%, where the results are expressed as percent of initial inventory. Excluding the results for the Dow NS-1 waste form obtained from Pilgrim Station, release fractions for  $^{60}Co$  ranged from 0.03% to 0.2%. The fraction of  $^{60}Co$  released from the Pilgrim sample was much higher than usual, being about 5%. Sim-

ilar results for  $^{125}Sb$  ranged from 0.2% to less than 7%.

For all waste-form samples except the Pilgrim Dow NS-1 sample, the total quantities of  $^{54}Mn$ ,  $^{60}Co$ , and  $^{125}Sb$  released from the waste forms can be accounted for if only the first 30  $\mu m$  of the surface layer of each sample were totally depleted of activity. Even in the case of the Pilgrim sample, which released about 5% of its inventory of  $^{60}Co$ , a surface totally depleted of  $^{60}Co$  to a depth of 0.5 mm would account for the total quantity of  $^{60}Co$  released from the waste form. These results indicate that it is possible that the observed releases of  $^{54}Mn$ ,  $^{60}Co$ , and  $^{125}Sb$  were the result of release mechanisms operating exclusively on or very near the surfaces of the waste forms. The operative release mechanisms might have included corrosion and dissolution of the waste-form surfaces and diffusion from just beneath the surfaces. The sharp declines in release rates that were observed over the course of leaching are consistent with significant depletion of activities from the surfaces of the waste forms.

Five of the seven resin waste forms released their entire inventories of  $^{137}Cs$  during leaching. The waste forms which were the exceptions released about 36% and 0.4% of their initial inventories of  $^{137}Cs$ . The waste form which released an unusually small fraction of its  $^{137}Cs$  inventory was the Dow NS-1 sample obtained from Pilgrim. It is not known if special additives were used during the solidification of the Pilgrim sample, but the anomalously low  $^{137}Cs$  release suggests that additives such as zeolites, clay, or silica were employed to reduce the leachability of  $^{137}Cs$ . The results show that while significantly less  $^{137}Cs$  was released from the Pilgrim sample compared to the quantities released from other waste-form samples, the releases of transition metal radionuclides were much higher than usual. The high  $^{137}Cs$  release fractions measured for most of the waste forms indicates that  $^{137}Cs$  remained soluble following cement hydration.

The leaching results obtained during the course of this study show that, with the exception of  $^{137}Cs$ , releases of radionuclides contained in the decontamination resin waste forms were well below the NRC Branch Technical Position limits<sup>8</sup> for solidified waste forms. But, in the case of  $^{137}Cs$ , one of the seven waste-form specimens released about 36% of its inventory of  $^{137}Cs$  and five specimens released their entire inventories of  $^{137}Cs$ . For several of these waste-form specimens, the leachability index of  $^{137}Cs$  was only slightly above the acceptable lower limit of 6.0 as defined by

the NRC Branch Technical Position on Waste Form.<sup>8</sup> However, in the case of the Dow NS-1 waste form obtained from the Pilgrim Station, only 0.4% of the original quantity of <sup>137</sup>Cs in the specimen was released during leaching; and the leachability index of <sup>137</sup>Cs for this sample ranged from greater than 10.4 to 13.4. Leachability indexes of this magnitude are typical of <sup>60</sup>Co. Although it is not known whether or not zeolites or other additives were used during the preparation of the Pilgrim sample, it is apparent that <sup>137</sup>Cs was immobilized by some means.

With the exception of the results for <sup>58</sup>Co for the waste-form specimen obtained from Pilgrim Station and the results for <sup>125</sup>Sb for the cation resin waste-form specimen obtained from Cooper Station, the measured radionuclide leachability indexes were all greater than six, which is the lower limit considered acceptable according to the NRC Branch Technical Position on Waste Form.<sup>8</sup> Leachability indexes of <sup>54</sup>Mn, <sup>55</sup>Fe, <sup>60</sup>Co, <sup>63</sup>Ni, <sup>125</sup>Sb, and <sup>241</sup>Pu were typically greater than 10. Similar results for <sup>90</sup>Sr ranged from 8.0 to 11.8, while leachability indexes of <sup>137</sup>Cs varied from 6.1 to 13.4. Radionuclide leachability indexes determined for the cation and mixed-bed resin waste-form specimens obtained from Cooper Station were similar to corresponding leachability indexes measured for specimens which maintained their physical integrity during leaching.

A previous study<sup>28</sup> conducted at the INEL determined the releases of <sup>60</sup>Co, <sup>90</sup>Sr, and <sup>137</sup>Cs from boric acid and sulfate evaporator wastes which had been solidified in cement at two operating LWRs. The average leachability indexes of <sup>60</sup>Co and <sup>90</sup>Sr for the majority of the decontamination resin waste forms were consistently higher than the corresponding leachability indexes determined for the boric acid and sulfate waste forms. The leachability indexes of <sup>137</sup>Cs for the majority of the decontamination resin waste forms were comparable to the <sup>137</sup>Cs leachability indexes determined for the evaporator waste forms. Assuming that the evaporator wastes were free of organic reagents, the results of this study indicate that the organic reagents contained in the solidified decontamination resin waste forms did not adversely affect their leaching characteristics.

## Release of Stable Metals from Waste Forms

The ranges of the effective diffusivities and leachability indexes of chromium, iron, cobalt, and nickel were generally comparable in magnitude to the corresponding ranges determined for radionu-

clides. The results show that nickel was released from the Pilgrim Dow NS-1 sample at a rate about ten times higher than the rate it was released from the Can-Decon and LOMI waste forms. This behavior correlates well with similar results for <sup>63</sup>Ni. In several instances, <sup>63</sup>Ni release rates were about the same as the release rates of nickel. The ranges of the leachability indexes of iron were generally comparable to the corresponding ranges determined for <sup>55</sup>Fe, while the ranges of the leachability index of cobalt were typically well below the corresponding ranges determined for <sup>60</sup>Co. The results for iron and cobalt are qualitative because, in most cases, the concentrations of these two metals in the leachates were below detection limits.

With the exception of the results for chromium and iron for the Cooper cation resin waste form, the measured stable metal leachability indexes were all greater than 6. The leachability indexes of these two metals were 6.0 and 5.2, respectively, during the initial 2-h-long leach of the Cooper cation resin waste form. Because this sample began to deteriorate during the first leaching interval and the leachate was not filtered prior to elemental analysis, the initial low leachability indexes for this waste form were very likely due to the presence of particulates in the leachate. The ranges of the leachability indexes of stable metals for all seven waste-form specimens were: chromium, 6.0 to >12.8; iron, 5.2 to >15.3; cobalt, 6.1 to >9.2; and nickel, 7.6 to 10.7.

## Release of Organic Acids from Waste Forms

The leaching results show that the total quantities of oxalic acid, citric acid, and EDTA which were released from the Can-Decon and CitroX waste forms were all quite small. Cumulative release fractions of organic acids for these waste forms ranged from 0.8% to 2.4% of initial inventories. However, organic acids were readily released from the Dow NS-1 waste form obtained from Pilgrim Station and from the LOMI waste form collected from Indian Point-3 Station. Results for the Pilgrim waste form indicated that between 30% and 40% of the original inventories of oxalic acid, citric acid, and DTPA in the sample were released during leaching. The results also indicated that the entire inventory of EDTA was leached from the Pilgrim waste form during 48 days of continuous leaching. In the case of the LOMI waste form obtained from Indian Point-3, 83% of the formic acid and 60% of the picolinic acid originally in the waste form were released

from the sample during 46 days of leaching. Measurements indicated that the release rates of organic acids from the Pilgrim and Indian Point-3 waste forms remained comparatively high even during the later, longer leaching intervals.

The trends in the releases of organic acids were, in many instances, similar to observed trends in releases of radionuclides. Release rates of both species generally decreased significantly as elapsed leaching time increased, with decreases in release rates being more pronounced during the longer leaching intervals. In the case of the Cooper mixed-bed resin Citrox waste form, which disintegrated completely within 1 h following immersion in leachant, releases of both radionuclides and organic acids decreased significantly after five days of leaching, indicating that surface effect release mechanisms dominated the release of both species during the initial leaching intervals. Thereafter, the releases of both species appeared to have been controlled by diffusion.

In almost all cases, considerably larger fractions of the inventories of organic acids were released compared to the fractions of the inventories of  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{63}\text{Ni}$ , and  $^{125}\text{Sb}$  which were released. Conversely, in several instances, the cumulative release fraction of  $^{137}\text{Cs}$  was much higher than the cumulative release fractions of the organic acids. The specific forms of the radionuclides when released from the waste forms are not known. Because the waste forms are chemically complex media, radionuclides may exist combined with binder materials as cationic or anionic ligands, or they may be complexed with organic acids.

With the exception of the result for EDTA for the Pilgrim waste-form specimen, all of the measured organic acid leachability indexes were greater than 6. The ranges of the leachability indexes of organic acids for all seven waste-form specimens were: oxalic acid, 7.2 to 11.7; citric acid, 6.5 to 12.0; EDTA, 5.7 to 9.0; DTPA, 7.5; formic acid, 6.7, and picolinic acid, 6.9.

## Leachability Indexes

Leachability indexes of radionuclides and organic reagents for each of the seven waste forms are plotted against elapsed leaching time in Appendix D, Figures D-1 through D-7. The average leachability indexes of radionuclides and organic acids contained in each of the waste forms are ranked from highest to lowest in Table 30. (Results for the Cooper Citrox waste forms are not included in Table 30, since they decomposed during initial

leaching.) For four of the five waste forms shown in Table 30,  $^{137}\text{Cs}$  exhibited the lowest leachability index among the radionuclides and organic acids. The results for the Pilgrim Dow NS-1 sample are remarkably different. In the case of the Pilgrim waste form, only  $^{55}\text{Fe}$  exhibited a higher leachability index than  $^{137}\text{Cs}$ . While very little  $^{137}\text{Cs}$  was released from the Pilgrim waste form, an anomalously high fraction of the  $^{63}\text{Ni}$  inventory was released. As shown in Table 30,  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ , and  $^{60}\text{Co}$  were normally the most intractable species and  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and the organic acids were the species most readily released from the waste forms.

## Recommendations for Future Studies

A primary objective of the current study is to establish a data base on the immersion stability and leachability of decontamination resin waste forms obtained from commercial LWRs. Because of the limited number of LWR decontamination waste forms which have been tested to date, there is a compelling need to test additional waste forms which are representative of commonly used decontamination processes. Special emphasis should be placed on testing Citrox and LOMI waste forms. Additional Citrox waste forms should be tested to determine if the observed lack of physical stability of the Cooper samples during immersion is a generic problem for waste forms containing relatively high concentrations of citric and oxalic acids. Additional LOMI waste forms should be tested because, at the current time, the LOMI process is the most commonly used decontamination process.

The waste forms tested during the current study were cured at ambient temperature, whereas resins solidified in large liners experience elevated temperatures during the hydration of cement. Previous studies performed using different sized waste forms have shown that the leachability of small (5- x 10-cm) and large [208-L (55-gal)] waste forms are similar, indicating that differences in hydration exotherm temperatures in this size range do not significantly affect leachability. Leaching studies should be performed using duplicate waste forms cured at ambient temperature and at temperatures representative of actual solidification liner exotherm temperatures. A study of this type is needed to verify that the leachability and physical stability characteristics of small samples cured at ambient temperature are representative of the behavior of the large monoliths in the solidification liners, which experience

**Table 30. Rankings of leachability indexes for waste forms**

Sample	Order of Ranking <sup>a</sup>
Millstone-1 F33	$^{54}\text{Mn} > ^{60}\text{Co} > ^{125}\text{Sb} > \text{CA} > \text{OA} > \text{EDTA} > ^{137}\text{Cs}$
Millstone-1 F201	$^{55}\text{Fe} > ^{54}\text{Mn} > ^{60}\text{Co} > ^{125}\text{Sb} > ^{63}\text{Ni} > \text{CA} > \text{OA} > \text{EDTA} > ^{90}\text{Sr} > ^{137}\text{Cs}$
Peach Bottom-2	$^{60}\text{Co} > ^{54}\text{Mn} > \text{OA} > \text{CA} > ^{90}\text{Sr} > \text{EDTA} > ^{137}\text{Cs}$
Pilgrim	$^{55}\text{Fe} > ^{137}\text{Cs} > ^{54}\text{Mn} > ^{60}\text{Co} > ^{90}\text{Sr} > \text{DTPA} > \text{OA} > ^{63}\text{Ni} > \text{CA} > \text{EDTA}$
Indian Point-3	$^{54}\text{Mn} > ^{55}\text{Fe} > ^{63}\text{Ni} > ^{60}\text{Co} > ^{90}\text{Sr} > \text{PA} > ^{137}\text{Cs} > \text{FA}$

a. OA = oxalic acid; CA = citric acid; FA = formic acid; PA = picolinic acid.

temperatures as high as 343 K (70°C) during initial phases of cement hydration.

The waste forms tested during the current study were aged a minimum of about 18 months prior to initiation of leaching. Many of the waste forms tested in other studies have been aged for 28 days prior to leaching. Leaching studies should be performed to determine the degree to which leachability and physical stability are affected by the length of time a sample is aged prior to the start of leaching.

Because the characteristics of the groundwaters at the various burial sites are different, leaching studies should be conducted using leachants representative of burial site groundwaters. These leaching tests should be performed using both groundwaters having a range of pH values and deionized water so as to allow comparison of the leaching data with historical data obtained using only deionized water. Leaching studies should also be performed using seawater leachant.

Very little information is currently available regarding the specific chemical forms of radionuclides contained in decontamination ion-exchange resin waste forms. Resin waste forms are extremely complex media in so far as the number and types of reactions which can occur between the resins, reagents, additives, and binder materials. Studies should be performed to identify the specific chemical forms of the radionuclides of interest so as to allow a more precise interpretation of leaching

data. Specific information of this type is needed to evaluate postulated release mechanisms.

Given the fact that the leachability index of  $^{137}\text{Cs}$  was only slightly above the acceptable lower limit of 6.0 for several of the waste-form specimens that were leach-tested, it is recommended that the benefits of the addition of zeolites and other materials during solidification be determined experimentally. The effect of these additives on the leachability of transition metal radionuclides and transuranics should also be determined.

Compressive-strength testing of waste-form specimens which have been aged for different lengths of time should be performed before and after leaching to determine if the length of aging prior to initiation of leaching affects the compressive strength of waste forms.

Long-term leaching studies should be performed to determine how accurately the results of 90-day leach tests predict the leaching behavior of decontamination ion-exchange resin waste forms leached for periods much longer than 90 days.

Decontamination ion-exchange waste forms should be tested, using the methods recommended by the NRC Branch Technical Position on Waste Form.<sup>8</sup> In addition to compressive strength and leach-testing, decontamination resin waste forms should be tested for resistance to degradation as a result of exposure to radiation, thermal cycling, and microorganisms.

## REFERENCES

1. H. Nishita and E. Essington, "Effect of Chelating Agents on the Movement of Fission Products in Soils," *Soil Science*, 103, p. 168, 1967
2. R. Jackson, W. Merritt, D. Champ. J. Gulens, and K. Inch, *The Distribution Coefficient as a Geochemical Measure of the Mobility of Contaminants in a Ground Water Flow System*, Vienna: International Atomic Energy Agency, 1977.
3. J. Means, D. Crerar, and J. Duguid, "Application of Gel Filtration Chromatography to Evaluation of Organo-Metallic Interactions in Natural Waters," *Limn. and Ocean.*, 22, p. 557, 1977.
4. J. Means, D. Crerar, and J. Duguid, "Migration of Radioactive Wastes: Radionuclide Mobilization by Complexing Agents," *Science*, 200, p. 1477, 1978.
5. J. Cleveland and T. Rees, "Characterization of Plutonium in Maxey Flats Radioactive Trench Leachates," *Science*, 212, p. 1506, 1981.
6. J. L. Means and C. A. Alexander, "The Environmental Biogeochemistry of Chelating Agents and Recommendations for the Disposal of Chelated Radioactive Waste," *Nucl. and Chem. Waste Management*, 2, p. 183, 1981.
7. Code of Federal Regulations, 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Wastes," Office of the Federal Register, December 1982.
8. "Technical Position on Waste Form," Rev. 0, U.S. Nuclear Regulatory Commission, Low-Level Waste Management Branch, Washington, D. C., May 1983.
9. "Compliance with 10 CFR Part 61 and Implementation of the Radiological Effluent Technical Specifications (RETS) and Attendant Process Control Program (PCP)," Generic Letter 84-12, U. S. Nuclear Regulatory Commission, Washington, D. C., April 30, 1984.
10. P. L. Piciulo, J. W. Adams, M. S. Davis, L. W. Milian, and C. I. Anderson, *Release of Organic Chelating Agents from Solidified Decontamination Wastes*, NUREG/CR-4709, July 1986.
11. P. Soo, L. W. Milian, and P. L. Piciulo, *The Leachability and Mechanical Integrity of Simulated Decontamination Resin Wastes Solidified in Cement and Vinyl Ester-Styrene*, NUREG/CR-5153, May 1988.
12. N. Moriyama, S. Dojiri, and H. Matsuzuru, "Leaching of Cs-137 from the Ion-Exchange Resin Incorporated in Polyethylene or Cement Composite," *Health Physics*, 32, p. 549, 1977.
13. P. Colombo and R. M. Neilson, Jr., *Properties of Radioactive Wastes and Waste Containers*, NUREG/CR-0619, August 1979.
14. R. Dayal, H. Arora, and N. Morcos, *Estimation of Cesium-137 Release from Waste/Cement Composites Using Data from Small-Scale Specimens*, NUREG/CR-3382, July 1983.
15. H. Arora and R. Dayal, *Leaching Studies of Cement-Based Low-Level Radioactive Waste Forms*, NUREG/CR-4756, October 1986.
16. N. Morcos, R. Dayal, and A. J. Weiss, *Properties of Radioactive Wastes and Waste Containers, Status Report October 1980 - September 1981*, NUREG/CR-2617, April 1982.

17. R. M. Neilson, Jr., P. D. Kalb, and P. Colombo, *Lysimeter Study of Commercial Reactor Waste Forms: Waste Form Acquisition, Characterization and Full-Scale Leaching*, BNL 51613, September 1982.
18. R. M. Neilson, Jr. and P. Colombo, *Waste Form Development Program Annual Progress Report, October 1981 - September 1982*, BNL 51614, September 1982.
19. R. E. Barletta, K. J. Swyler, S. F. Chan, and R. E. Davis, *Leach Behavior and Mechanical-Integrity Studies of Irradiated EPICOR-II Waste Products*, BNL-NUREG-31929, 1982.
20. K. J. Swyler, C. J. Dodge, and R. Dayal, *Irradiation Effects on the Storage and Disposal of Radwaste Containing Organic Ion-Exchange Media*, NUREG/CR-3383, October 1983.
21. R. E. Barletta, J. W. Adams, and R. E. Davis, *Physical Tests on Solidified Decontamination Wastes from Dresden Unit 1*, NUREG/CR-3165, December 1983.
22. H. Arora and R. Dayal, *Solidification and Leaching of Boric Acid and Resin LWR Wastes*, NUREG/CR-3909, June 1984.
23. J. H. Westsik, Jr. (Comp.), *Characterization of Cement and Bitumen Waste Forms Containing Simulated Low-Level Waste Incinerator Ash*, NUREG/CR-3798, August 1984.
24. R. M. Neilson, Jr., J. W. McConnell, Jr., and R. D. Rogers, "Performance Testing of High Specific Activity Waste Forms per 10 CFR Part 61," *Waste Management '87, Proceedings of the Symposium on Waste Management, Tucson, Arizona, March 1-5, 1987*.
25. E. D. Hespe, "Leach Testing of Immobilized Radioactive Waste Solids - A Proposal for a Standard Method," *Atomic Energy Review*, 9, p. 195, 1971.
26. "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure," ANSI/ANS-16.1-1986, American Nuclear Society Standards Committee, April 1986.
27. S. W. Duce and J. T. Case, *Effectiveness and Safety Aspects of Selected Decontamination Processes*, NUREG/CR-4445, August 1986.
28. S. T. Croney, *Leachability of Radionuclides from Cement Solidified Waste Forms Produced at Operating Nuclear Power Plants*, NUREG/CR-4181, March 1985.
29. M. S. Davis, *The Impact of LWR Decontaminations on Solidification, Waste Disposal and Associated Occupational Exposure*, NUREG/CR-3444, August 1983.
30. R. Soto, *Decontamination Waste Management*, EPRI NP-4240, September 1985.
31. *Development of LOMI Chemical Decontamination Technology*, EPRI NP-3177, July 1983.
32. D. L. Kantro and L. E. Copeland, "The Stoichiometry of the Hydration of Portland Cement," *National Bureau of Standards Monograph*, 1, 43, p. 442, 1962.
33. R. M. Wheaton and A. H. Seamster, "A Basic Reference on Ion Exchange," in *Encyclopedia of Chemical Technology*, 2nd Edition, II, pp. 871-899, 1966.
34. D. R. Dougherty and P. Colombo, *Leaching Mechanisms of Solidified Low-Level Waste, The Literature Survey*, BNL-51899, June 1985.



35. J. E. Mendel et al., *A State-of-the-Art Review of Materials Properties of Nuclear Waste Forms*, PNL-3802, 1981.
36. H. W. Godbee and D. S. Joy, *Assessment of the Loss of Radioactive Isotopes from Waste Solids to the Environment. Part 1: Background and Theory*, ORNL-TM-4333, 1974.
37. O. U. Anders, J. F. Bartel, and S. J. Altschuler, "Determination of the Leachability of Solids," *Anal. Chem.*, 50, 4, pp. 564-569, 1978.
38. J. Crank, *The Mathematics of Diffusion*, 2nd Edition, Oxford: Clarendon Press, 1975.
39. H. W. Godbee et. al., "Application of Mass Transport Theory to the Leaching of Radionuclides from Waste Solids," *Nucl. and Chem. Waste Manag.*, 1, pp. 29-35, 1980.
40. C. W. Nestor, Jr., *Diffusion from Solid Cylinders*, ORNL/CSD/TM-84, January 1980.
41. D. Eastwood, *Properties of Solidified Decontamination Wastes, Quarterly Progress Report, July-September 1982*, BNL-NUREG-32072, November 1982.
42. C. W. Filby et al., *Environmental Analysis Procedure Manual*, Idaho National Engineering Laboratory, ENI-120, March 1980.
43. L. Rudling, "Simultaneous Determination of Nitrilotriacetic Acid, Ethylenediaminetetraacetic Acid, and Diethylenetriaminepentaacetic Acid as Their Methyl Ester Derivatives by GLC," *Water Research*, 6, pp. 871-876, 1972.
44. C. J. Eskall and M. E. Pick, *Simultaneous Determination of Oxalic, Citric, Nitrilotriacetic, and Ethylenediaminetetraacetic Acids by Gas Liquid Chromatography of Their Methyl Esters*, RD/B/N4763, Berkeley Nuclear Laboratories, April 1980.

**APPENDIX A**

**SUMMARY OF SOLIDIFIED RESIN WASTE LEACH STUDIES**



## **APPENDIX A**

### **SUMMARY OF SOLIDIFIED RESIN WASTE LEACH STUDIES**

A summary of solidified resin waste leach studies is presented in Table A-1.

**Table A-1. Summary of solidified resin waste leach studies**

Reference Number(s)	Sample Size Dia. x Hght. (cm)	Solidification Agent	Waste Loading <sup>a</sup> (W/W)	Cure Time (days)	Waste Type	Leachant <sup>b</sup>	Leach Test Duration (days)	Radionuclide, Element, Chelant
1	4.5 x 4.4	Portland C	0.18	28	Diaion SK IB cation	DIW	326 <sup>c</sup>	<sup>137</sup> Cs
	4.5 x 4.4	Polyethylene	0.12	— <sup>d</sup>	Diaion SK IB cation	DIW	326 <sup>c</sup>	<sup>137</sup> Cs
2	10.95 g	None	— <sup>d</sup>	— <sup>d</sup>	Rohm and Haas (R&H) IRN-77 (Na <sup>+</sup> )	DIW, SW, & GW	90 <sup>e</sup>	<sup>137</sup> Cs, <sup>85</sup> Sr
	0.97 g	None	— <sup>d</sup>	— <sup>d</sup>	Powdex PCH (Na <sup>+</sup> )	DIW, SW, & GW	90 <sup>e</sup>	<sup>137</sup> Cs, <sup>85</sup> Sr
	2.5 x 5.1	Urea-formaldehyde	2.6	— <sup>d</sup>	R&H IRN-150	DIW	120 <sup>c</sup>	<sup>137</sup> Cs, <sup>85</sup> Sr
3, 4	4.7 x 4.8	Portland I	0.13	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	260 <sup>c</sup>	<sup>137</sup> Cs
	4.7 x 9.2	Portland I	0.12	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	260	<sup>137</sup> Cs
	7.6 x 8.5	Portland I	0.13	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	260	<sup>137</sup> Cs
	15.4 x 14.6	Portland I	0.13	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	240	<sup>137</sup> Cs
	15.2 x 29.8	Portland I	0.13	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	119	<sup>137</sup> Cs
	31.2 x 29.2	Portland I	0.13	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	95	<sup>137</sup> Cs
	54.6 x 54.6	Portland I	0.13	28	R&H IRN-77 (Na <sup>+</sup> )	DIW	70	<sup>137</sup> Cs
5	4.7 x 8.9	Portland II	0.25	42	R&H IRN-77 (Na <sup>+</sup> )	DIW	95 <sup>c</sup>	<sup>137</sup> Cs, <sup>60</sup> Co, <sup>85</sup> Sr
	4.7 x 8.9	Portland II	0.36	42	R&H IRN-77 (Na <sup>+</sup> )	DIW	95	<sup>137</sup> Cs, <sup>60</sup> Co, <sup>85</sup> Sr
	4.7 x 8.9	Lumnite (HAC)	0.25	42	R&H IRN-77 (Na <sup>+</sup> )	DIW	95	<sup>137</sup> Cs, <sup>60</sup> Co, <sup>85</sup> Sr
	4.7 x 8.9	Lumnite (HAC)	0.36	42	R&H IRN-77 (Na <sup>+</sup> )	DIW	95	<sup>137</sup> Cs, <sup>60</sup> Co, <sup>85</sup> Sr
	— <sup>d</sup>	Pioneer 221						
	— <sup>d</sup>	(P 221) Bitumen	1.0	1	R&H IRN-77 (Na <sup>+</sup> )	DIW	95	<sup>137</sup> Cs
	— <sup>d</sup>	Bitumen (P 221)	1.0	1	R&H IRN-77 (Na <sup>+</sup> )	DIW	95	<sup>85</sup> Sr
	— <sup>d</sup>	Bitumen (P 221)	1.0	1	(1:1) R&H IRN-77 (Na <sup>+</sup> ) and IRN-78 (SO <sub>4</sub> <sup>-2</sup> )	DIW	95	<sup>137</sup> Cs
	— <sup>d</sup>	Bitumen (P 221)	1.0	1	Same	DIW	95	<sup>85</sup> Sr
6	54.6 x 54.6	Portland III	< 0.01 dry resin	300	Na <sub>2</sub> SO <sub>4</sub> conc. + mixed bed resin	DIW	316 <sup>f</sup>	<sup>134</sup> Cs, <sup>137</sup> Cs, <sup>60</sup> Co
7	4.7 x 6.8	Portland III	0.058 dry	55	R&H IRN-77 cation	DIW	21 <sup>f</sup>	<sup>137</sup> Cs, <sup>85</sup> Sr, <sup>60</sup> Co
	4.7 x 6.8	High alumina cement	0.058 dry	55	R&H IRN-77 cation	DIW	21	<sup>137</sup> Cs, <sup>85</sup> Sr, <sup>60</sup> Co
	4.7 x 6.5 <sup>g</sup>	Portland III	0.074 dry	14	(1:2) R&H IRN-77 and R&H IRN-78 anion	DC & SSW	5	<sup>137</sup> Cs, <sup>85</sup> Sr, <sup>60</sup> Co
	4.7 x 8.7	Envirostone	0.05 & 0.10 dry	7	Same	DIW & SSW	44	<sup>137</sup> Cs, <sup>85</sup> Sr, <sup>60</sup> Co
8	4.6 X 5.1 <sup>h</sup>	Portland I	0.33	19	D-mix	DIW	30 <sup>c</sup>	<sup>137</sup> Cs, <sup>85</sup> Sr

Table A-1. (continued)

Reference Number(s)	Sample Size Dia. x Hght. (cm)	Solidification Agent	Waste Loading <sup>a</sup> (W/W)	Cure Time (days)	Waste Type	Leachant <sup>b</sup>	Leach Test Duration (days)	Radionuclide, Element, Chelant
9	5 x 5 <sup>i</sup>	Portland I	0.13	28	R&H IRN-77 (Na + )	DIW	24 <sup>c</sup>	<sup>137</sup> Cs, <sup>85</sup> Sr
10	4.7 x 7.0 3.6 x 3.7	Vinyl ester-styrene Nuclear Binder-101	0.6 to 0.7 0.7	— <sup>d</sup> 3	DOW NS-1 conc. DOW NS-1 conc.	DIW, GW, & SW DIW, GW, & SW	64 50	Iron, nickel <sup>59</sup> Fe, <sup>60</sup> Co
11	4.7 x 9.3 (5 each)	Portland I	0.13	28	BWR waste resin	DIW	263	<sup>134</sup> Cs, <sup>137</sup> Cs, Sr-90
12	5 x 10 5 x 6 15 x 17 30 x 35 Same	Portland II    Bitumen (P 221)	0.23   0.40	28   — <sup>d</sup>	Incinerator ash: 31 W% cation resin, 11 W% anion resin, 58 W% general trash <sup>j</sup> Same	DIW, SBGW   DIW, SBGW	336 <sup>c</sup> 91 91 91 Same	Al, Ca, Cs, Co, Cu, Fe, Si, Na, and Sr  Same
13	4.7 x 9.6	Portland I	0.13	42 to 361	R&H IRN-78 anion	DIW	91 <sup>f</sup>	Oxalic, citric, picolinic, formic acids, EDTA, and LND-101A <sup>k</sup>
	4.7 x 9.6	Portland I	0.13	41 to 278	(1:2) R&H IRN-77 and R&H IRN-78 anion	DIW	92	Same
	4.7 x 9.6	Portland I	0.13	31	Ionac A-365 anion	DIW	88	Picolinic, formic
	4.7 x 9.6	Portland I	0.13	73	(1:2) R&H IRN-77 and Ionac A-365 anion	DIW	88	Picolinic, formic
	4.7 x 9.5	Vinyl ester-styrene	0.34	— <sup>d</sup>	R&H IRN-78 anion	DIW	88	EDTA, picolinic
	4.6 x 9.5	Vinyl ester-styrene	0.33	— <sup>d</sup>	(1:2) R&H IRN-77 and R&H IRN-78 anion	DIW	88	LND-101A, oxalic, and picolinic
	4.6 x 9.7	Vinyl ester-styrene	0.34	— <sup>d</sup>	Ionac A-365 anion	DIW	88	Picolinic
	4.6 x 9.6	Vinyl ester-styrene	0.33	— <sup>d</sup>	(1:2) R&H IRN-77 and Ionac A-365 anion	DIW	88	Picolinic
14	5 x 8	Portland I-II	0.24	600	PF-7 or PF-24 from EPICOR-II prefilters <sup>l</sup>	DIW, SSW	91 <sup>f</sup>	<sup>134</sup> Cs, <sup>137</sup> Cs
	5 x 8	Vinyl ester-styrene	0.61	600	Same	DIW, SSW	91	<sup>134</sup> Cs, <sup>137</sup> Cs

**Table A-1. (continued)**

Reference Number(s)	Sample Size Dia. x Hght. (cm)	Solidification Agent	Waste Loading <sup>a</sup> (W/W)	Cure Time (days)	Waste Type	Leachant <sup>b</sup>	Leach Test Duration (days)	Radionuclide, Element, Chelant
15	5 x 10	Portland I	0.13	28	(1:2) R&H IRN-77 and R&H IRN-78 anion	DIW	91 <sup>f</sup>	EDTA
	15 x 15	Portland I	0.13	28	Same	DIW	119	EDTA
	30 x 30	Portland I	0.13	28	Same	DIW	119	EDTA
	5 x 10	Portland I	0.13	28	(1:2) R&H IRN-77 and Ionac A-365 anion	DIW	474	Picolinic
	15 x 15	Portland I	0.13	28	Same	DIW	474	Picolinic
	30 x 30	Portland I	0.13	28	Same	DIW	308	Picolinic
	5 x 10	Vinyl ester-styrene	0.33	— <sup>d</sup>	(1:2) R&H IRN-77 and Ionac A-365 anion	DIW	203	Picolinic
	15 x 15	Vinyl ester-styrene	0.33	— <sup>d</sup>	Same	DIW	205	Picolinic
	4.6 x 9.5	Portland I	0.13	38	R&H IRN-77 (Na <sup>+</sup> )	DIW, 100 ppm picolinate, 100 ppm formate	60	<sup>60</sup> Co

a. Ratio of weight of resin(s), excluding excess water, to total weight of binder, water, and resin(s).

b. DIW = deionized water; SW = seawater; GW = Long Island aquifer groundwater; DC = demineralized water/cement leachant; SSW = simulated seawater; SBGW = simulated Barnwell groundwater.

c. Modified IAEA leach-test procedure.

d. Not applicable.

e. Static leach test.

f. ANS 16.1 leach-test procedure.

g. The 4.7 x 6.5 samples were irradiated to total doses of either 10<sup>4</sup> or 10<sup>6</sup> rads prior to static leach-testing.

h. The 4.7 x 5.1 samples were irradiated to total doses of either 10<sup>6</sup> or 10<sup>7</sup> rads prior to leach-testing.

i. One set of samples was irradiated to 9 x 10<sup>8</sup> rad to determine the effect of irradiation on leaching behavior.

j. Weights prior to incineration.

k. LND-101A = London Nuclear proprietary decontamination reagents.

l. PF-7 consists of phenolic cation, strong acid cation, and strong base anion. PF-24 consists of strong acid cation, strong base anion, and inorganic zeolite.

## References for Table A-1

1. N. Moriyama, S. Dojiri, and H. Matsuzuru, "Leaching of Cs-137 from the Ion-Exchange Resin Incorporated in Polyethylene or Cement Composite," *Health Physics*, 32, 1977, p. 549.
2. P. Colombo and R. M. Neilson, Jr., *Properties of Radioactive Wastes and Waste Containers*, NUREG/CR-0619, August 1979.
3. R. Dayal, H. Arora, and N. Morcos, *Estimation of Cesium-137 Release from Waste/Cement Composites Using Data from Small-Scale Specimens*, NUREG/CR-3382, July 1983.
4. H. Arora and R. Dayal, *Leaching Studies of Cement-Based Low-Level Radioactive Waste Forms*, NUREG/CR-4756, October 1986.
5. N. Morcos, R. Dayal, and A. J. Weiss, *Properties of Radioactive Wastes and Waste Containers, Status Report October 1980 - September 1981*, NUREG/CR-2617, April 1982.
6. R. M. Neilson, Jr., P. D. Kalb, and P. Colombo, *Lysimeter Study of Commercial Reactor Waste Forms: Waste Form Acquisition, Characterization and Full-Scale Leaching*, BNL 51613, September 1982.
7. R. M. Neilson, Jr. and P. Colombo, *Waste Form Development Program Annual Progress Report, October 1981 - September 1982*, BNL 51614, September 1982.
8. R. E. Barletta, K. J. Swyler, S. F. Chan, and R. E. Davis, *Leach Behavior and Mechanical-Integrity Studies of Irradiated EPICOR-II Waste Products*, BNL-NUREG-31929, 1982.
9. K. J. Swyler, C. J. Dodge, and R. Dayal, *Irradiation Effects on the Storage and Disposal of Radwaste Containing Organic Ion-Exchange Media*, NUREG/CR-3383, October 1983.
10. R. E. Barletta, J. W. Adams, and R. E. Davis, *Physical Tests on Solidified Decontamination Wastes from Dresden Unit 1*, NUREG/CR-3165, December 1983.
11. H. Arora and R. Dayal, *Solidification and Leaching of Boric Acid and Resin LWR Wastes*, NUREG/CR-3909, June 1984.
12. J. H. Westsik, Jr. (Comp.), *Characterization of Cement and Bitumen Waste Forms Containing Simulated Low-Level Waste Incinerator Ash*, NUREG/CR-3798, August 1984.
13. P. L. Piciulo, J. W. Adams, M. S. Davis, L. W. Milian, and C. I. Anderson, *Release of Organic Chelating Agents from Solidified Decontamination Wastes*, NUREG/CR-4709, July 1986.
14. R. M. Neilson, Jr., J. W. McConnell, Jr., and R. D. Rogers, "Performance Testing of High Specific Activity Waste Forms per 10 CFR Part 61," *Waste Management '87, Proceedings of the Symposium on Waste Management, Tucson, Arizona, March 1-5, 1987*.
15. P. Soo, L. W. Milian, and P. L. Piciulo, *The Leachability and Mechanical Integrity of Simulated Decontamination Resin Wastes Solidified in Cement and Vinyl Ester-Styrene*, NUREG/CR-5153, May 1988.





**APPENDIX B**

**ANS 16.1 LEACH-TEST PROCEDURE**



## APPENDIX B

### ANS 16.1 LEACH-TEST PROCEDURE

#### B.1 Requirements for Specimen Preparation

**B.1.1 Sampling.** Suitable precautions shall be taken to ensure that the specimen is representative of the solidified waste and that the homogeneity of the test specimen is the same as that of the material in the actual solidified waste form.

**B.1.2 Sample Preparation.** The test specimen shall be prepared in the same or similar manner as that established for the solidification process. The cement/waste mixture shall be cast in a form that has a well-defined shape, mass, and volume. Where possible, the specimen shall be a monolithic cylinder, parallelepiped, or sphere, with the cylindrical geometry being the preferred geometry. The casting shall conform to the sides of the specimen container and be performed in such a way that voids are eliminated and the sample is homogeneous. The thermal history of the test specimen is to be reported. The specimen container shall remain sealed during curing. The container shall be constructed of material(s) known to be chemically unreactive toward the specimen (e.g., polyethylene, polypropylene, stainless steel, ceramic, glass, etc.).

#### B.2 Requirements for Leach-Test Vessel

**B.2.1 Materials of Construction.** The vessel in which leaching takes place should be constructed of a material that:

1. Does not react chemically with the leachant or the specimen;
2. Does not sorb chemical species extracted from the specimen or those in the leachant itself;
3. Does not release interfering species which alter the composition of the leachant during the leaching process; and
4. Can withstand the conditions involved in leaching.

In addition, the leach-test vessel shall be constructed such that evaporation of the leachant is maintained below 2% per day.

**B.2.2 Dimensions and Form.** The dimensions of the vessel shall be such that greater than 98% of the external geometric surface area of the immersed specimen is in contact with the leachant during the entire leaching interval.

#### B.3 Requirements for Specimen Support

The specimen shall be supported in the leachant by any convenient device, made from unreactive material, that does not interfere with the leachate removal and replacement, does not impede leaching, does not damage the surface of the specimen, and does not preclude more than 2% of the specimen's external surface from exposure to the leachant.

#### B.4 Requirements for Leachant

The leachant shall be demineralized water with an electrical conductivity of less than 5  $\mu\text{mho}/\text{cm}$  at 298 K (25°C) and a total organic carbon content of less than 3 ppm. The leachant shall be kept in, and not exceed, the limits of the temperature range 291 to 301 K (17.5 to 27.5°C) during the test.

#### B.5 Leach Test Method

**B.5.1 Leachant Volume.** Sufficient leachant shall be used so that the ratio of the leachant volume,  $V_L$ , and the specimen external geometric surface area,  $S$ , is maintained such that

$$\frac{V_L(\text{cm}^3)}{S(\text{cm}^2)} = 10 \pm 0.2 (\text{cm})$$

**B.5.2 Procedure.** After removal from the specimen container and prior to the initiation of the leach test, the test specimen shall be rinsed by immersion in demineralized water for 30 s. The rinse water volume shall be the same as the required leachant volume.

The container used to store the specimen before leaching shall be rinsed with an amount of water equal to its volume. This water shall be combined with the specimen rinse water, and the combined rinse water shall be analyzed to determine the quantities of the species of interest that are present.

The specimen shall be supported in the leachate using a device of the type previously described in Section B.3. The leachate shall be sampled and the leachate completely replaced after cumulative leach times of 2, 7, and 24 h from the initiation of the test. Subsequent leachate sampling and leachant replacements shall be made at 24-h intervals for the next 4 days. Three additional leach intervals of 14, 28, and 43 days will be used to extend the entire test to 90 days. The standard requires that the leachant changeout time be short and insignificant relative to the duration of a leaching interval.

## B.6 Leachate Analysis

An aliquot of the leachate shall be taken at the end of each leaching interval to determine the amounts of the species of interest present in the leachate volume. The release of the species of interest will be determined by measuring the amount present in the leachate rather than the residual in the specimen.

Stirring of the leachate so as to suspend particulate solids prior to taking the leachate aliquot, or dissolution of such solids by the addition of chemicals to the leachate before sampling, may be necessary. If precipitation occurs in the leachate during the leaching interval, the amount of the extracted species of interest associated with the precipitate shall be determined and added to the amount of the dissolved species.

## B.7 Reporting of Results

**B.7.1 Experimental Information.** When reporting the results of leaching tests, the following information concerning the specimen tested and the leachant used during the test shall be reported:

1. The type of waste and the proportion of waste and binder incorporated in the waste-binder mixture prior to solidification.
2. The type and composition of the binder material, including additives.
3. The radionuclides and the total amount of each in the leaching specimen, both as-prepared and after the 30-s rinse.
4. Shape, mass, and dimensions of the leach-test specimen.
5. The history of the specimen between preparation and leaching, including time, temperature, and any other relevant information.
6. Electrical conductivity of leachant and total organic carbon.
7. Volume of leachant used during each leaching interval.

**B.7.2 Test Data.** The incremental fraction leached ( $a_n/A_o$ ), the incremental leaching rate  $[(a_n/A_o)/(t_n-t_{n-1})]$ , and the cumulative fraction leached ( $\Sigma a_n/A_o$ ) are recorded as functions of time where:

$a_n$  = quantity of a species released from the specimen during leaching interval  $n$ ;

$\Sigma a_n$  = cumulative quantity of a species released from the specimen from the beginning of the first leaching interval to the end of the leaching interval of concern;

$A_o$  = total quantity of a given species in the specimen at the beginning of the first leaching interval;

$t_n-t_{n-1}$  = duration of the  $n$ -th leaching interval, s.

**APPENDIX C**

**GLC CHROMATOGRAMS OF RESIN WASTE SAMPLES**



## APPENDIX C

### GLC CHROMATOGRAMS OF RESIN WASTE SAMPLES

Examples of chromatograms obtained by analyzing resin waste samples using the gas liquid chromatography (GLC) method are shown in Figures C-1 through C-6. In all cases, the volume of sample injected was 5  $\mu$ L. The retention times and identifications of the esters of the organic acids of interest are indicated in each figure. The ethyl benzoate internal standard has a retention time of about 14.3 min. The retention times of the methyl esters of the organic acids are: oxalic acid, 8.4 to 8.7 min; citric acid, 19.5 to 19.6 min; EDTA, 27.2 to 27.3 min; DTPA, 35.5 min; picolinic acid, 16.0 min. The chromatograms of the resins are identified below by plant and decontamination process.

- C-1. Indian Point-3 LOMI resin waste. Chloroform extract diluted by a factor of ten prior to injection.
- C-2. Peach Bottom-2 Can-Decon resin waste. Chloroform extract diluted by a factor of ten prior to injection.
- C-3. Cooper AP/Citrox mixed bed resin waste. Chloroform extract diluted by a factor of ten prior to analysis.
- C-4. Cooper AP/Citrox cation resin waste. Undiluted.
- C-5. Pilgrim Dow NS-1 resin waste. Undiluted.
- C-6. Pilgrim Dow NS-1 resin Waste. Chloroform extract diluted by a factor of ten prior to analysis.

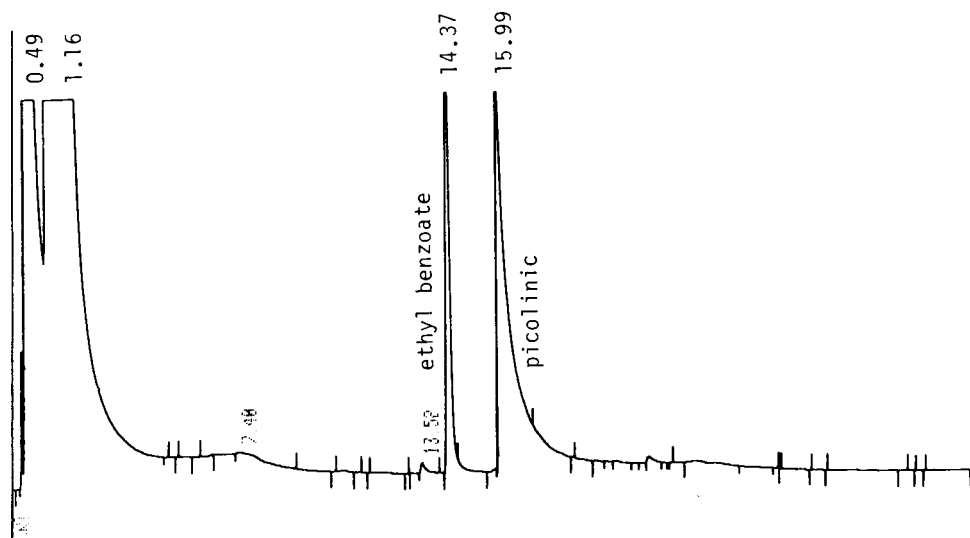
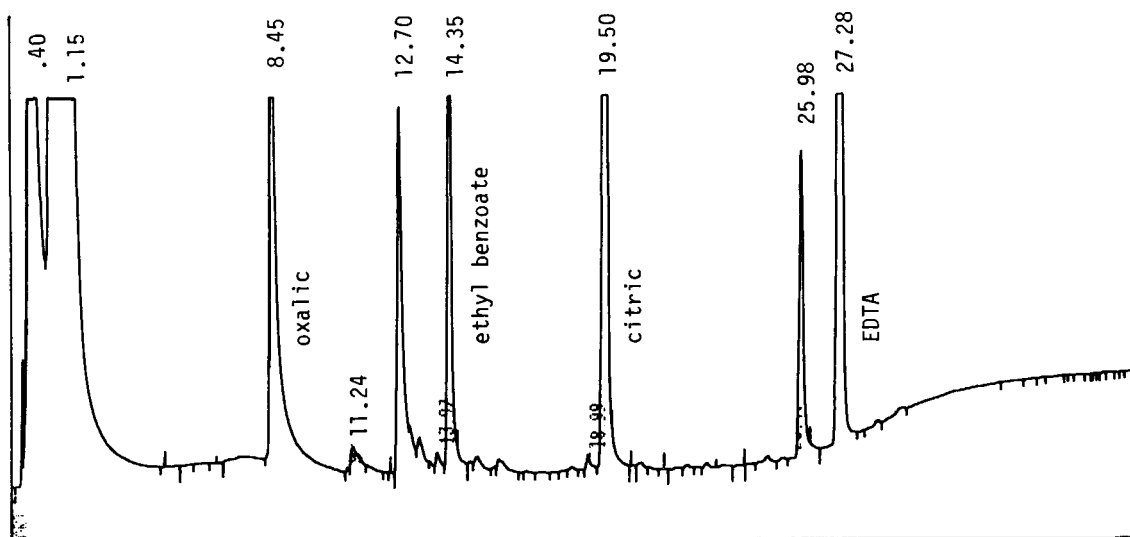
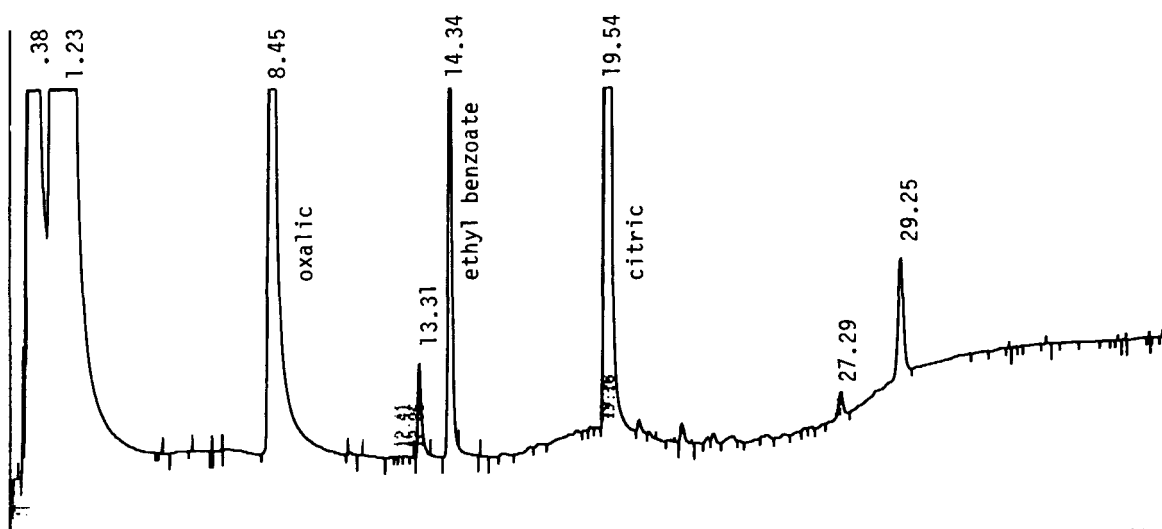


Figure C-1. Chromatogram of the Indian Point-3 waste resin.

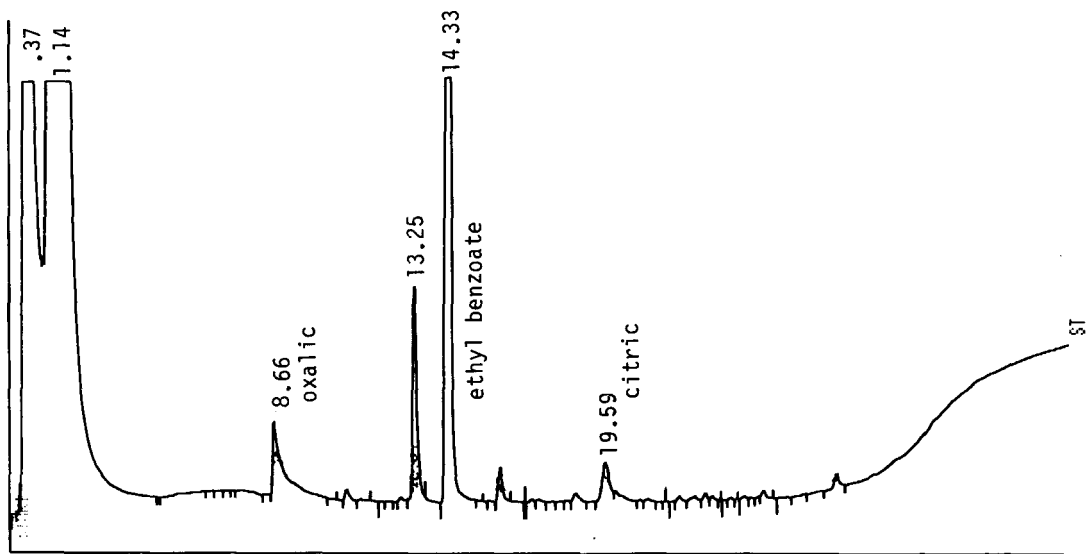




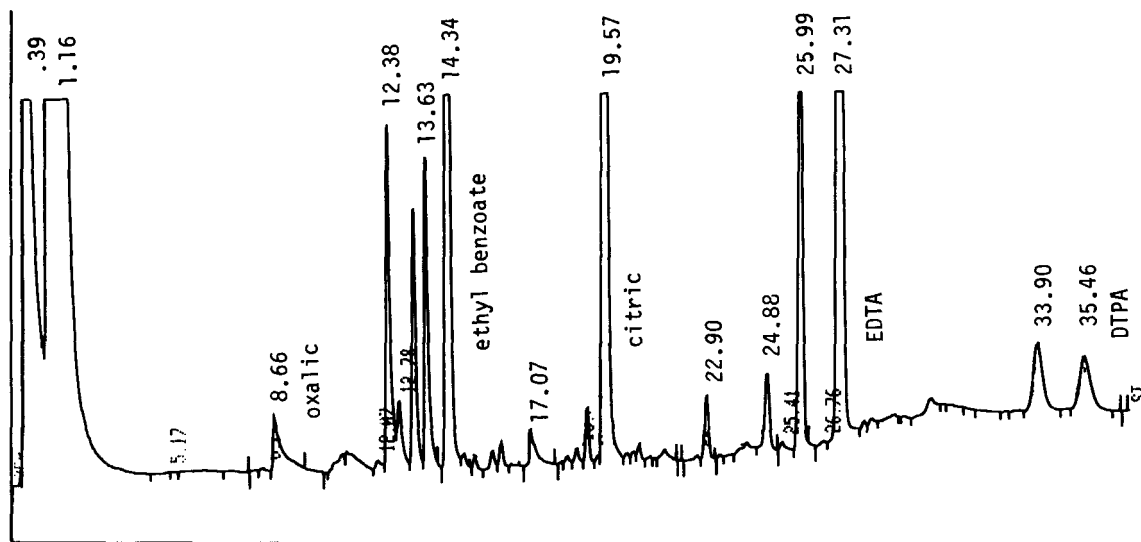
**Figure C-2.** Chromatogram of the Peach Bottom-2 waste resin.



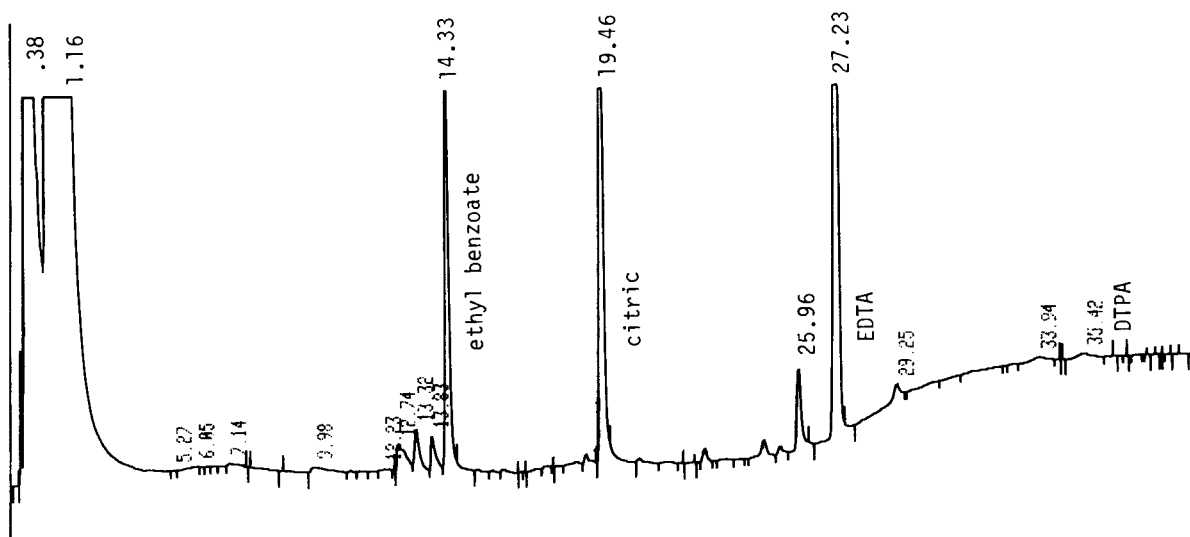
**Figure C-3.** Chromatogram of the Cooper mixed-bed waste resin.



**Figure C-4.** Chromatogram of the Cooper cation waste resin.



**Figure C-5.** Chromatogram of the Pilgrim waste resin (undiluted).



**Figure C-6.** Chromatogram of the Pilgrim waste resin (diluted).

**APPENDIX D**

**GRAPHS OF LEACHABILITY INDEX VERSUS LEACHING TIME**



## APPENDIX D

### GRAPHS OF LEACHABILITY INDEX VERSUS LEACHING TIME

Plots of leachability index versus leaching time for the resin waste samples are given in Figures D-1 through D-7.

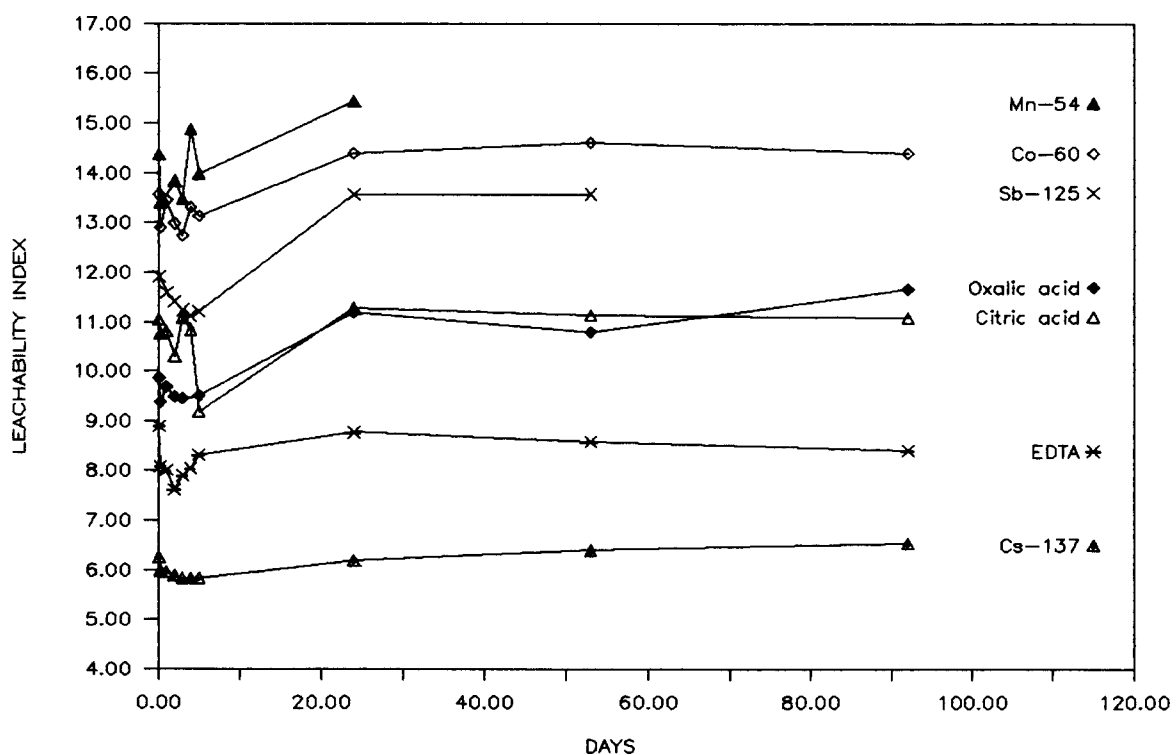


Figure D-1. Leachability index versus leaching time for Millstone-1 F33 waste form.

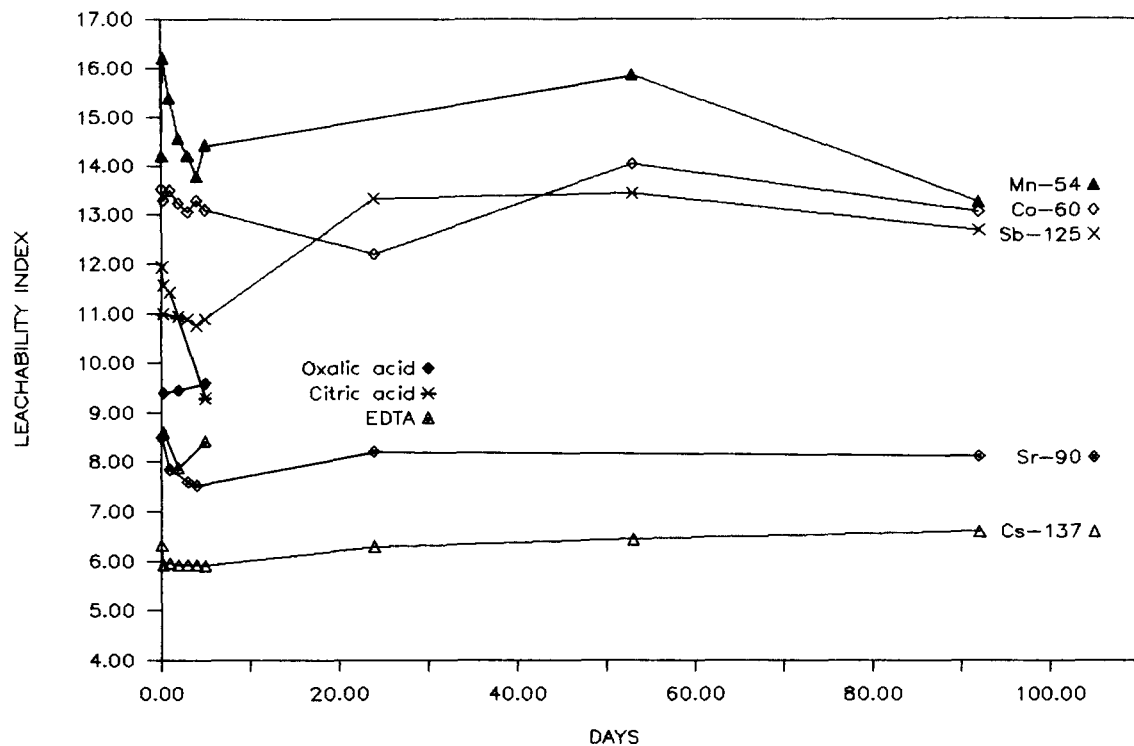


Figure D-2. Leachability index versus leaching time for Millstone-1 F201 waste form.

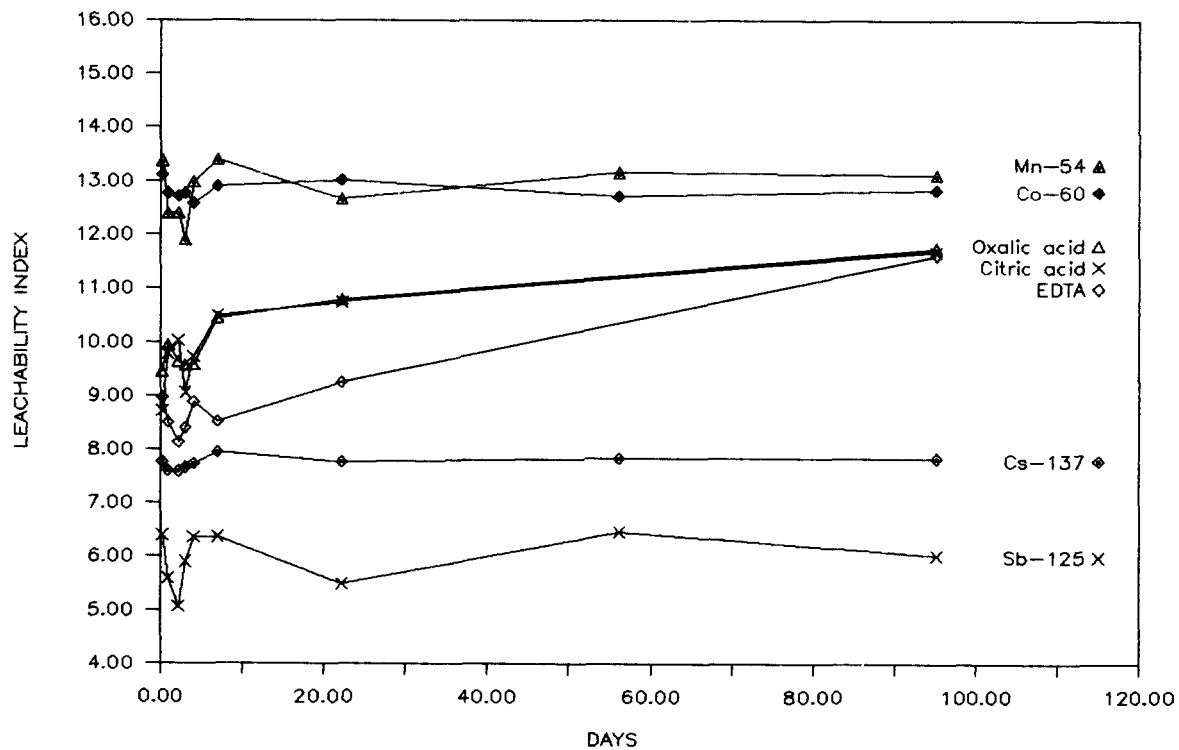


Figure D-3. Leachability index versus leaching time for Peach Bottom-2 waste form.

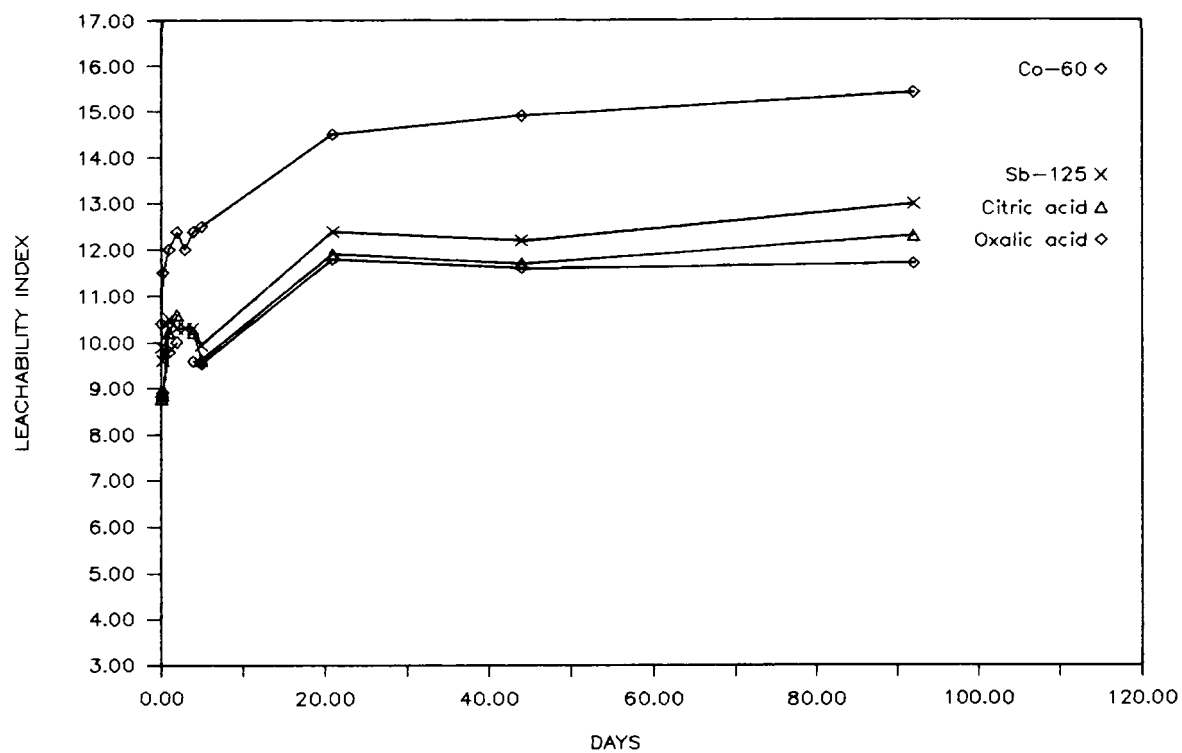


Figure D-4. Leachability index versus leaching time for Cooper mixed-bed waste form.

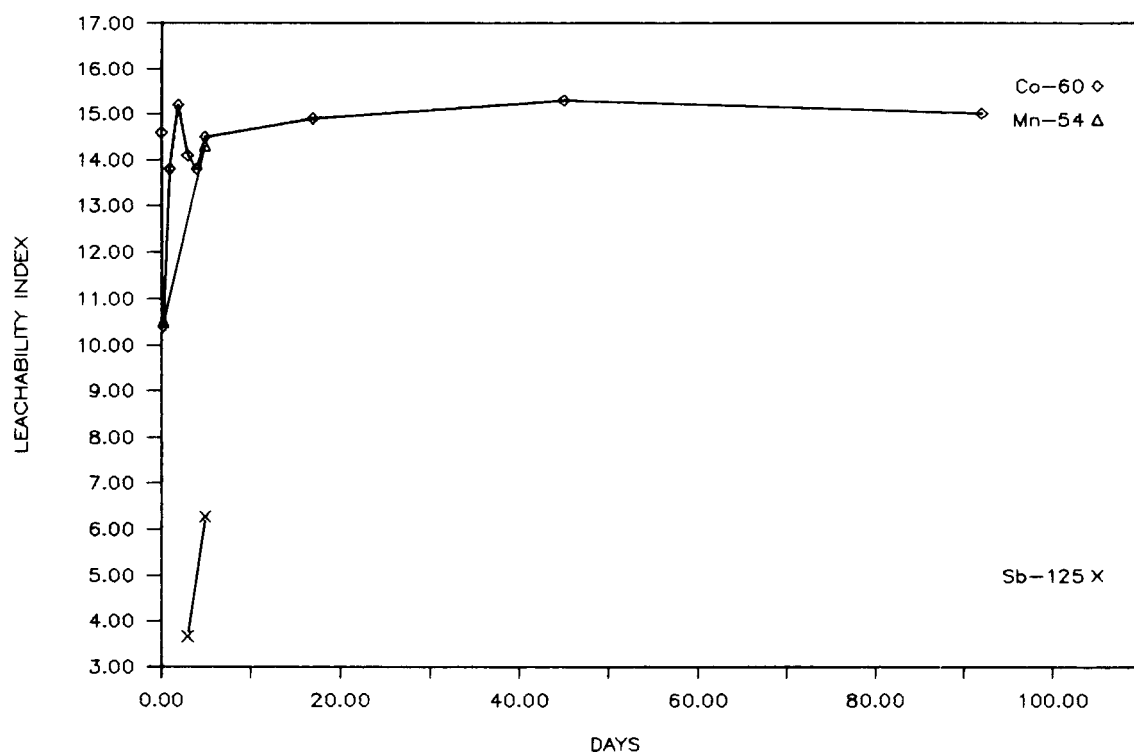


Figure D-5. Leachability index versus leaching time for Cooper cation waste form.



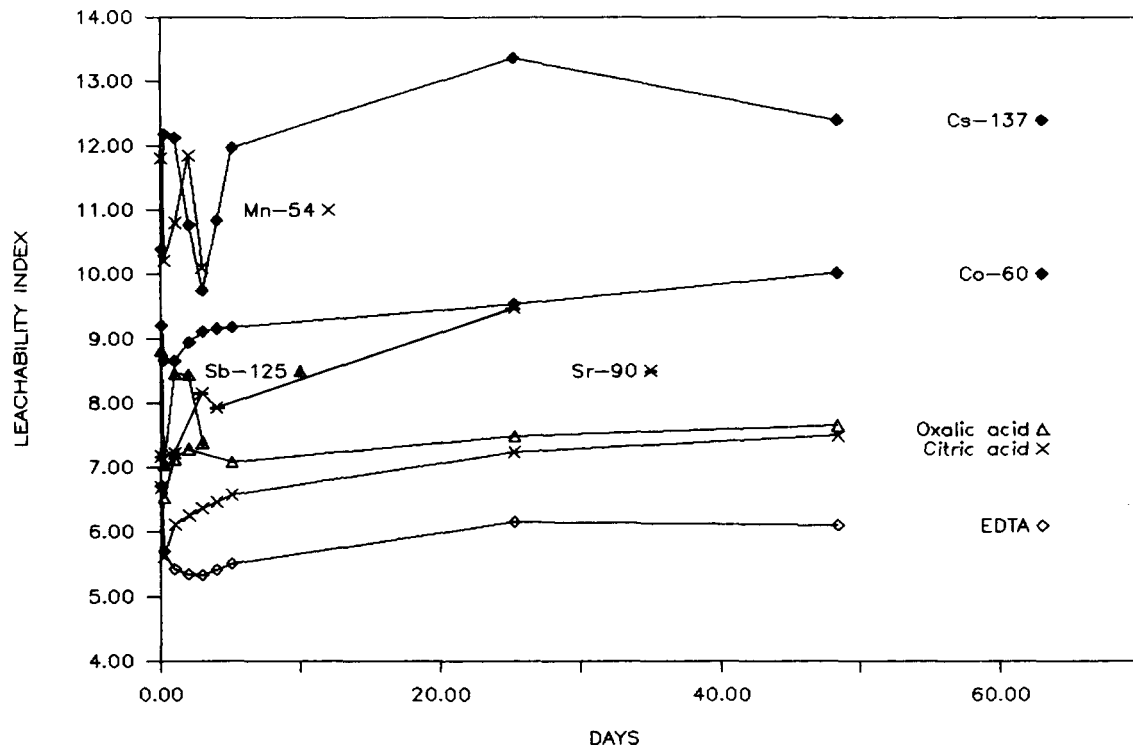


Figure D-6. Leachability index versus leaching time for Pilgrim waste form.

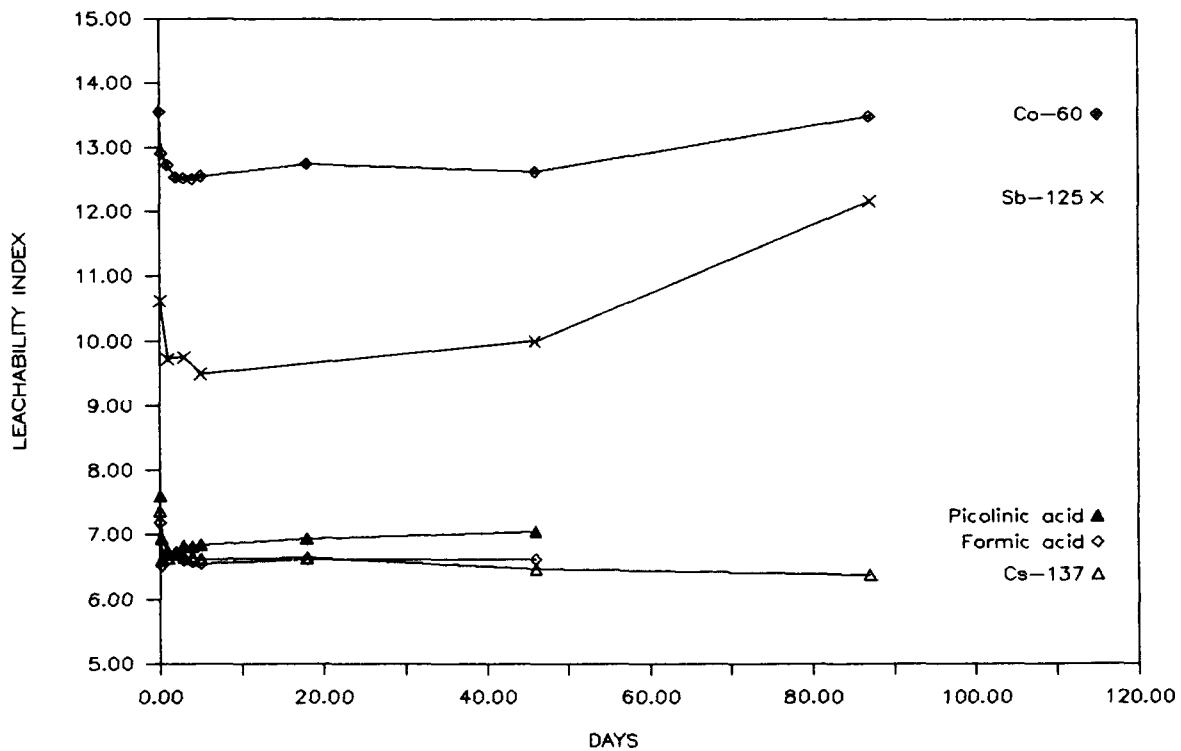


Figure D-7. Leachability index versus leaching time for Indian Point-3 waste form.

**APPENDIX E**

**RADIONUCLIDE LEACHING DATA FOR WASTE-FORM SAMPLES**



## **APPENDIX E**

### **RADIONUCLIDE LEACHING DATA FOR WASTE-FORM SAMPLES**

The radionuclide leaching data for waste-form samples are given in Tables E-1 through E-7.

TABLE E-1. RADIONUCLIDE LEACHING DATA FOR INDIAN POINT-3 SAMPLE

Mn-54

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.05E-04	5.73E-10	1.07E-11	1.32E-05	1.32E-05	1.63E-14	1.38E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	6.27E-04	5.99E-11	1.12E-12	1.17E-05	NA	5.09E-15	1.43E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	2.35E-04	1.59E-11	2.98E-13	4.40E-06	NA	1.50E-15	1.48E+01
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	1.41E-04	9.54E-12	1.79E-13	2.64E-06	NA	9.77E-16	1.50E+01
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	2.66E-04	6.43E-13	1.20E-14	4.98E-06	NA	3.01E-17	1.65E+01
10*	8.70E+01	2.43E-03	4.01E-12	7.50E-14	4.55E-05	NA	2.49E-15	1.46E+01

Mean:  
Standard Deviation:

Fe-55

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.08E-02	8.79E-09	4.68E-11	5.77E-05	5.77E-05	3.12E-13	1.25E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	3.61E-03	3.45E-10	1.84E-12	1.92E-05	NA	1.36E-14	1.39E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	4.99E-04	3.38E-11	1.80E-13	2.66E-06	NA	5.48E-16	1.53E+01
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	2.77E-04	1.88E-11	1.00E-13	1.48E-06	NA	3.07E-16	1.55E+01
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	1.28E-03	3.09E-12	1.64E-14	6.80E-06	NA	5.60E-17	1.63E+01
10*	8.70E+01	1.41E-02	2.34E-11	1.24E-13	7.54E-05	NA	6.86E-15	1.42E+01

Mean: 1.46E+01  
Standard Deviation: 1.23E+00

Co-58

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.71E-02	3.01E-08	1.20E-11	1.48E-05	1.48E-05	2.07E-14	1.37E+01
2*	2.92E-01	7.27E-02	2.36E-08	9.45E-12	2.91E-05	4.40E-05	1.05E-13	1.30E+01
3*	1.00E+00	1.76E-01	1.68E-08	6.72E-12	7.04E-05	1.14E-04	1.83E-13	1.27E+01
4*	2.00E+00	1.93E-01	1.31E-08	5.24E-12	7.75E-05	1.92E-04	2.73E-13	1.26E+01
5*	3.00E+00	1.42E-01	9.61E-09	3.85E-12	5.69E-05	2.49E-04	2.50E-13	1.26E+01
6*	4.00E+00	1.32E-01	8.95E-09	3.58E-12	5.29E-05	3.02E-04	3.05E-13	1.25E+01
7*	5.00E+00	9.85E-02	6.67E-09	2.67E-12	3.94E-05	3.41E-04	2.18E-13	1.27E+01
8*	1.80E+01	6.74E-01	3.51E-09	1.40E-12	2.70E-04	6.11E-04	1.41E-13	1.29E+01
9*	4.60E+01	1.05E+00	2.53E-09	1.01E-12	4.19E-04	1.03E-03	2.12E-13	1.27E+01
10*	8.70E+01	3.64E-01	6.01E-10	2.41E-13	1.46E-04	1.18E-03	2.57E-14	1.36E+01

Mean: 1.29E+01  
Standard Deviation: 3.98E-01

TABLE E-1. RADIONUCLIDE LEACHING DATA FOR INDIAN POINT-3 SAMPLE (Continued)

Co-60

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.54E-03	6.13E-09	1.40E-11	1.73E-05	1.73E-05	2.80E-14	1.36E+01
2*	2.92E-01	1.39E-02	4.50E-09	1.03E-11	3.18E-05	4.91E-05	1.25E-13	1.29E+01
3*	1.00E+00	3.16E-02	3.02E-09	6.92E-12	7.24E-05	1.22E-04	1.94E-13	1.27E+01
4*	2.00E+00	3.51E-02	2.38E-09	5.45E-12	8.06E-05	2.02E-04	2.95E-13	1.25E+01
5*	3.00E+00	2.76E-02	1.87E-09	4.27E-12	6.32E-05	2.65E-04	3.09E-13	1.25E+01
6*	4.00E+00	2.35E-02	1.59E-09	3.65E-12	5.40E-05	3.19E-04	3.17E-13	1.25E+01
7*	5.00E+00	1.97E-02	1.33E-09	3.05E-12	4.51E-05	3.64E-04	2.85E-13	1.25E+01
8*	1.80E+01	1.35E-01	7.01E-10	1.61E-12	3.09E-04	6.73E-04	1.85E-13	1.27E+01
9*	4.60E+01	1.97E-01	4.75E-10	1.09E-12	4.51E-04	1.12E-03	2.46E-13	1.26E+01
10*	8.70E+01	7.37E-02	1.22E-10	2.79E-13	1.69E-04	1.29E-03	3.44E-14	1.35E+01
Mean:								1.28E+01
Standard Deviation:								3.71E-01

Ni-63

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.12E-02	1.73E-08	3.24E-11	3.99E-05	3.99E-05	1.49E-13	1.28E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	1.80E-02	1.72E-09	3.22E-12	3.37E-05	NA	4.20E-14	1.34E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	1.31E-02	8.85E-10	1.66E-12	2.45E-05	NA	4.66E-14	1.33E+01
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	9.48E-03	6.42E-10	1.20E-12	1.78E-05	NA	4.44E-14	1.34E+01
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	7.52E-02	1.82E-10	3.41E-13	1.41E-04	NA	2.41E-14	1.36E+01
10*	8.70E+01	2.78E-02	4.59E-11	8.61E-14	5.22E-05	NA	3.28E-15	1.45E+01
Mean:								1.35E+01
Standard Deviation:								5.00E-01

Sr-90

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.38E-05	2.74E-11	1.90E-09	2.34E-03	2.34E-03	5.14E-10	9.29E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	1.82E-04	1.74E-11	1.21E-09	1.26E-02	NA	5.90E-09	8.23E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	1.55E-04	1.05E-11	7.29E-10	1.08E-02	NA	8.97E-09	8.05E+00
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	1.13E-04	7.66E-12	5.31E-10	7.84E-03	NA	8.62E-09	8.06E+00
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	1.06E-03	2.57E-12	1.78E-10	7.38E-02	NA	6.59E-09	8.18E+00
10*	8.70E+01	4.22E-04	6.97E-13	4.83E-11	2.93E-02	NA	1.03E-09	8.99E+00
Mean:								8.47E+00
Standard Deviation:								4.87E-01

TABLE E-1. RADIONUCLIDE LEACHING DATA FOR INDIAN POINT-3 SAMPLE (Continued)

Sb-125

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 5.25E-05	< 4.26E-11	< 4.13E-10	< 5.08E-04	< 5.08E-04	< 2.42E-11	> 1.06E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 2.36E-04	< 2.26E-11	< 2.18E-10	< 2.29E-03	NA	< 1.93E-10	> 9.71E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 1.57E-04	< 1.07E-11	< 1.03E-10	< 1.53E-03	NA	< 1.80E-10	> 9.74E+00
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	< 1.57E-04	< 1.07E-11	< 1.03E-10	< 1.53E-03	NA	< 3.26E-10	> 9.49E+00
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	9.45E-04	2.28E-12	2.21E-11	9.15E-03	NA	1.01E-10	9.99E+00
10*	8.70E+01	< 7.87E-05	< 1.30E-13	< 1.26E-12	< 7.63E-04	NA	< 7.01E-13	> 1.22E+01

Mean:

Standard Deviation:

Cs-137

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.18E-01	2.58E-07	1.73E-08	2.14E-02	2.14E-02	4.27E-08	7.37E+00
2*	2.92E-01	6.61E-01	2.15E-07	1.44E-08	4.44E-02	6.58E-02	2.44E-07	6.61E+00
3*	1.00E+00	1.19E+00	1.14E-07	7.63E-09	7.99E-02	1.46E-01	2.36E-07	6.63E+00
4*	2.00E+00	1.13E+00	7.65E-08	5.14E-09	7.60E-02	2.22E-01	1.99E-07	6.70E+00
5*	3.00E+00	8.21E-01	5.55E-08	3.73E-09	5.52E-02	2.77E-01	2.26E-07	6.65E+00
6*	4.00E+00	6.92E-01	4.68E-08	3.14E-09	4.65E-02	3.23E-01	2.27E-07	6.64E+00
7*	5.00E+00	5.51E-01	3.73E-08	2.51E-09	3.71E-02	3.60E-01	2.37E-07	6.62E+00
8*	1.80E+01	3.61E+00	1.88E-08	1.26E-09	2.43E-01	6.03E-01	2.28E-07	6.64E+00
9*	4.60E+01	4.37E+00	1.06E-08	7.10E-10	2.94E-01	8.97E-01	3.42E-07	6.47E+00
10*	8.70E+01	1.53E+00	2.53E-09	1.70E-10	1.03E-01	1.00E+00	4.13E-07	6.38E+00

Mean:

6.67E+00

Standard Deviation:

2.50E-01

Pu-241

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 1.60E-03	< 1.30E-09	< 2.39E-10	< 2.94E-04	< 2.94E-04	< 8.13E-12	> 1.11E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 7.12E-04	< 6.80E-11	< 1.25E-11	< 1.31E-04	NA	< 6.32E-13	> 1.22E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 8.90E-04	< 6.02E-11	< 1.11E-11	< 1.64E-04	NA	< 2.07E-12	> 1.17E+01
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	< 8.90E-04	< 6.02E-11	< 1.11E-11	< 1.64E-04	NA	< 3.75E-12	> 1.14E+01
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	< 7.12E-04	< 1.72E-12	< 3.16E-13	< 1.31E-04	NA	< 2.07E-14	> 1.37E+01
10*	8.70E+01	< 5.34E-04	< 8.81E-13	< 1.62E-13	< 9.82E-05	NA	< 1.16E-14	> 1.39E+01

Mean:

Standard Deviation:

TABLE E-1. RADIONUCLIDE LEACHING DATA FOR INDIAN POINT-3 SAMPLE (Continued)

Fe

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.22E+01	2.62E-05	3.57E-10	4.40E-04	4.40E-04	1.82E-11	1.07E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 1.29E+01	< 1.23E-06	< 1.68E-11	< 1.76E-04	NA	< 1.14E-12	> 1.19E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 1.29E+01	< 8.73E-07	< 1.19E-11	< 1.76E-04	NA	< 2.40E-12	> 1.16E+01
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	< 1.29E+01	< 8.73E-07	< 1.19E-11	< 1.76E-04	NA	< 4.34E-12	> 1.14E+01
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	< 1.29E+01	< 3.12E-08	< 4.25E-13	< 1.76E-04	NA	< 3.75E-14	> 1.34E+01
10*	8.70E+01	< 1.29E+01	< 2.13E-08	< 2.90E-13	< 1.76E-04	NA	< 3.74E-14	> 1.34E+01

Mean:  
Standard Deviation:

Ni

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.13E+02	9.17E-05	1.16E-09	1.43E-03	1.43E-03	1.92E-10	9.72E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	1.61E+02	1.54E-05	1.95E-10	2.05E-03	NA	1.54E-10	9.81E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	1.29E+02	8.73E-06	1.11E-10	1.64E-03	NA	2.07E-10	9.68E+00
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	1.13E+02	7.64E-06	9.68E-11	1.43E-03	NA	2.87E-10	9.54E+00
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	3.87E+02	9.35E-07	1.19E-11	4.91E-03	NA	2.92E-11	1.05E+01
10*	8.70E+01	1.93E+02	3.19E-07	4.05E-12	2.45E-03	NA	7.27E-12	1.11E+01

Mean: 1.01E+01  
Standard Deviation: 5.74E-01



TABLE E-1. RADIONUCLIDE LEACHING DATA FOR INDIAN POINT-3 SAMPLE (Continued)

Co

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 6.45E+01	< 5.24E-05	---	---	---	---	---
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 6.45E+01	< 6.16E-06	---	---	---	---	---
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 6.45E+01	< 4.36E-06	---	---	---	---	---
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	< 6.45E+01	< 4.36E-06	---	---	---	---	---
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	< 6.45E+01	< 1.56E-07	---	---	---	---	---
10*	8.70E+01	< 6.45E+01	< 1.06E-07	---	---	---	---	---
Mean:								NA
Standard Deviation:								NA

Cr

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 1.29E+01	< 1.05E-05	< 1.05E-09	< 1.29E-03	< 1.29E-03	< 1.56E-10	> 9.81E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 1.29E+01	< 1.23E-06	< 1.23E-10	< 1.29E-03	NA	< 6.15E-11	> 1.02E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 1.29E+01	< 8.73E-07	< 8.73E-11	< 1.29E-03	NA	< 1.29E-10	> 9.89E+00
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	< 1.29E+01	< 8.73E-07	< 8.73E-11	< 1.29E-03	NA	< 2.34E-10	> 9.63E+00
8*	1.80E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.60E+01	< 1.29E+01	< 3.12E-08	< 3.12E-12	< 1.29E-03	NA	< 2.02E-12	> 1.17E+01
10*	8.70E+01	< 1.29E+01	< 2.13E-08	< 2.13E-12	< 1.29E-03	NA	< 2.01E-12	> 1.17E+01
Mean:								
Standard Deviation:								

Table E-2. RADIONUCLIDE LEACHING DATA FOR COOPER MIXED BED RESIN SAMPLE

## Mn-54 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED ( $\mu\text{Ci}$ )	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ( $\text{cm}^2/\text{s}$ )	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA
3*	1.00E+00	6.37E-04	6.75E-10	4.13E-05	NA	2.05E-13	1.27E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.27E+01
Standard Deviation:							NA

## Fe-55 (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED ( $\mu\text{Ci}$ )	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ( $\text{cm}^2/\text{s}$ )	LEACH INDEX
1*	8.33E-02	1.81E-02	5.91E-08	4.25E-04	4.25E-04	5.50E-11	1.03E+01
2*	2.92E-01	5.26E-02	6.87E-08	1.24E-03	1.66E-03	6.14E-10	9.21E+00
3*	1.00E+00	1.97E-02	7.58E-09	4.63E-04	2.12E-03	2.58E-11	1.06E+01
4*	2.00E+00	3.29E-02	8.95E-09	7.72E-04	2.90E-03	8.84E-11	1.01E+01
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	2.96E-02	3.50E-10	6.95E-04	NA	2.92E-12	1.15E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.03E+01
Standard Deviation:							7.55E-01

## Co-60 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED ( $\mu\text{Ci}$ )	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ( $\text{cm}^2/\text{s}$ )	LEACH INDEX
1*	8.33E-02	2.90E-02	5.19E-08	3.73E-04	3.73E-04	4.25E-11	1.04E+01
2*	2.92E-01	7.13E-03	5.10E-09	9.16E-05	4.65E-04	3.38E-12	1.15E+01
3*	1.00E+00	7.01E-03	1.48E-09	9.02E-05	5.55E-04	9.77E-13	1.20E+01
4*	2.00E+00	4.04E-03	6.02E-10	5.20E-05	6.07E-04	4.00E-13	1.24E+01
5*	3.00E+00	4.65E-03	6.93E-10	5.98E-05	6.67E-04	9.01E-13	1.20E+01
6*	4.00E+00	2.66E-03	3.96E-10	3.42E-05	7.01E-04	4.13E-13	1.24E+01
7*	5.00E+00	2.15E-03	3.20E-10	2.76E-05	7.29E-04	3.47E-13	1.25E+01
8*	2.10E+01	2.15E-03	2.00E-11	2.76E-05	7.56E-04	3.52E-15	1.45E+01
9*	4.40E+01	1.07E-03	6.93E-12	1.37E-05	7.70E-04	1.14E-15	1.49E+01
10*	9.20E+01	9.44E-04	2.93E-12	1.21E-05	7.82E-04	4.28E-16	1.54E+01
Mean:							1.28E+01
Standard Deviation:							1.52E+00

Table E-2. RADIONUCLIDE LEACHING DATA FOR COOPER MIXED BED RESIN SAMPLE (Continued)

## Ni-63 (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.71E-04	1.35E-09	9.69E-06	9.69E-06	2.87E-14	1.35E+01
2*	2.92E-01	1.06E-03	1.54E-09	2.76E-05	3.73E-05	3.07E-13	1.25E+01
3*	1.00E+00	2.69E-04	1.15E-10	7.02E-06	4.43E-05	5.92E-15	1.42E+01
4*	2.00E+00	2.69E-04	8.13E-11	7.02E-06	5.14E-05	7.29E-15	1.41E+01
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	3.53E-04	4.64E-12	9.21E-06	NA	5.12E-16	1.53E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.39E+01
Standard Deviation:							9.10E-01

## Sr-90 (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.75E-06	1.15E-08	8.23E-05	8.23E-05	2.07E-12	1.17E+01
2*	2.92E-01	5.61E-05	1.47E-07	2.64E-03	2.72E-03	2.79E-09	8.55E+00
3*	1.00E+00	4.03E-06	3.10E-09	1.89E-04	2.91E-03	4.31E-12	1.14E+01
4*	2.00E+00	3.68E-05	2.00E-08	1.73E-03	4.64E-03	4.43E-10	9.35E+00
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	1.12E-05	2.66E-10	5.27E-04	NA	1.68E-12	1.18E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.05E+01
Standard Deviation:							1.33E+00

## Sb-125 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.87E-03	8.94E-08	6.42E-04	6.42E-04	1.26E-10	9.90E+00
2*	2.92E-01	3.51E-03	4.38E-08	7.86E-04	1.43E-03	2.49E-10	9.60E+00
3*	1.00E+00	2.39E-03	8.76E-09	5.35E-04	1.96E-03	3.44E-11	1.05E+01
4*	2.00E+00	2.62E-03	6.80E-09	5.87E-04	2.55E-03	5.11E-11	1.03E+01
5*	3.00E+00	2.00E-03	5.18E-09	4.47E-04	3.00E-03	5.02E-11	1.03E+01
6*	4.00E+00	1.59E-03	4.13E-09	3.57E-04	3.35E-03	4.50E-11	1.03E+01
7*	5.00E+00	2.24E-03	5.82E-09	5.02E-04	3.86E-03	1.15E-10	9.94E+00
8*	2.10E+01	1.26E-03	2.05E-10	2.82E-04	4.14E-03	3.68E-13	1.24E+01
9*	4.40E+01	1.49E-03	1.68E-10	3.34E-04	4.47E-03	6.73E-13	1.22E+01
10*	9.20E+01	8.04E-04	4.34E-11	1.80E-04	4.65E-03	9.39E-14	1.30E+01
Mean:							1.08E+01
Standard Deviation:							1.15E+00

Table E-2. RADIONUCLIDE LEACHING DATA FOR COOPER MIXED BED RESIN SAMPLE (Continued)

## Cs-137 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.24E-03	7.83E-06	5.63E-02	5.63E-02	9.66E-07	6.02E+00
2*	2.92E-01	6.47E-03	1.63E-05	2.94E-01	3.50E-01	3.82E-06	5.42E+00
3*	1.00E+00	3.33E-03	2.48E-06	1.51E-01	5.01E-01	2.59E-06	5.59E+00
4*	2.00E+00	6.90E-04	3.63E-07	3.13E-02	5.33E-01	1.49E-06	5.83E+00
5*	3.00E+00	2.26E-03	1.19E-06	1.03E-01	6.35E-01	1.64E-06	5.79E+00
6*	4.00E+00	1.67E-03	8.77E-07	7.57E-02	7.11E-01	1.65E-06	5.78E+00
7*	5.00E+00	4.32E-03	2.28E-06	1.96E-01	9.07E-01	3.32E-06	5.48E+00
8*	2.10E+01	5.30E-04	1.75E-08	2.41E-02	9.31E-01	8.98E-07	6.05E+00
9*	4.40E+01	1.09E-03	2.49E-08	4.94E-02	9.81E-01	6.83E-07	6.17E+00
10*	9.20E+01	4.24E-04	4.65E-09	1.93E-02	1.00E+00	3.91E-07	6.41E+00
Mean:							5.85E+00
Standard Deviation:							2.97E-01

## Pu-241 (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 9.36E-04	< 5.56E-08	< 4.00E-04	< 4.00E-04	< 4.87E-11	< 1.03E+01
2*	2.92E-01	< 7.48E-04	< 1.78E-08	< 3.20E-04	< 7.19E-04	< 4.11E-11	< 1.04E+01
3*	1.00E+00	< 9.36E-04	< 6.54E-09	< 4.00E-04	< 1.12E-03	< 1.92E-11	< 1.07E+01
4*	2.00E+00	< 3.74E-04	< 1.85E-09	< 1.60E-04	< 1.28E-03	< 3.79E-12	< 1.14E+01
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	< 3.74E-04	< 8.06E-11	< 1.60E-04	NA	< 1.54E-13	< 1.28E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

## Fe (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 1.32E+01	< 3.32E-08	< 2.39E-04	< 2.39E-04	< 1.74E-11	< 1.08E+01
2*	2.92E-01	< 1.32E+01	< 1.33E-08	< 2.39E-04	< 4.77E-04	< 2.29E-11	< 1.06E+01
3*	1.00E+00	< 1.32E+01	< 3.90E-09	< 2.39E-04	< 7.16E-04	< 6.84E-12	< 1.12E+01
4*	2.00E+00	< 1.32E+01	< 2.77E-09	< 2.39E-04	< 9.54E-04	< 8.43E-12	< 1.11E+01
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	< 1.32E+01	< 1.20E-10	< 2.39E-04	NA	< 3.44E-13	< 1.25E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

Table E-2. RADIONUCLIDE LEACHING DATA FOR COOPER MIXED BED RESIN SAMPLE (Continued)

Ni (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.09E+01	1.25E-06	9.01E-03	9.01E-03	2.48E-08	7.61E+00
2*	2.92E-01	< 6.60E+01	< 4.66E-07	< 8.38E-03	< 1.74E-02	< 2.83E-08	> 7.55E+00
3*	1.00E+00	< 6.60E+01	< 1.37E-07	< 8.38E-03	< 2.58E-02	< 8.44E-09	> 8.07E+00
4*	2.00E+00	7.42E+01	1.09E-07	9.43E-03	< 3.52E-02	1.32E-08	7.88E+00
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	< 6.60E+01	< 4.22E-09	< 8.38E-03	NA	< 4.24E-10	> 9.37E+00
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

Co (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 6.60E+01	---	---	---	---	---
2*	2.92E-01	< 6.60E+01	---	---	---	---	---
3*	1.00E+00	< 6.60E+01	---	---	---	---	---
4*	2.00E+00	< 6.60E+01	---	---	---	---	---
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	< 6.60E+01	---	---	---	---	---
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

Cr (UNFILTERED)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.48E+01	NA	NA	NA	NA	NA
2*	2.92E-01	< 1.32E+01	NA	NA	NA	NA	NA
3*	1.00E+00	< 1.32E+01	NA	NA	NA	NA	NA
4*	2.00E+00	< 1.32E+01	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	2.10E+01	NA	NA	NA	NA	NA	NA
9*	4.40E+01	< 1.32E+01	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							NA
Standard Deviation:							NA

Table E-3. RADIONUCLIDE LEACHING DATA FOR COOPER CATION RESIN SAMPLE

## Mn-54 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA
2*	2.92E-01	3.24E-02	1.43E-08	2.57E-04	NA	3.00E-11	1.05E+01
3*	1.00E+00	NA	NA	NA	NA	NA	NA
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	3.91E-04	3.58E-11	3.10E-06	NA	4.96E-15	1.43E+01
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.24E+01
Standard Deviation:							1.89E+00

## Fe-55 (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	4.04E+01	8.10E-06	5.83E-02	5.83E-02	1.18E-06	5.93E+00
2*	2.92E-01	5.65E-02	4.54E-09	8.17E-05	5.84E-02	3.04E-12	1.15E+01
3*	1.00E+00	2.22E-01	5.24E-09	3.21E-04	5.87E-02	1.40E-11	1.09E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	3.23E-01	5.40E-09	4.67E-04	NA	8.73E-11	1.01E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	1.09E-01	6.51E-11	1.57E-04	NA	1.07E-13	1.30E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.03E+01
Standard Deviation:							2.37E+00

## Co-60 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.09E-03	3.57E-10	2.57E-06	2.57E-06	2.28E-15	1.46E+01
2*	2.92E-01	2.27E-01	1.55E-08	2.79E-04	2.82E-04	3.56E-11	1.04E+01
3*	1.00E+00	9.02E-03	1.81E-10	1.11E-05	2.93E-04	1.68E-14	1.38E+01
4*	2.00E+00	1.55E-03	2.21E-11	1.91E-06	2.95E-04	6.14E-16	1.52E+01
5*	3.00E+00	4.21E-03	6.01E-11	5.19E-06	3.00E-04	7.67E-15	1.41E+01
6*	4.00E+00	5.11E-03	7.28E-11	6.29E-06	3.06E-04	1.58E-14	1.38E+01
7*	5.00E+00	2.05E-03	2.92E-11	2.52E-06	3.09E-04	3.28E-15	1.45E+01
8*	1.70E+01	9.72E-03	1.15E-11	1.20E-05	3.21E-04	1.16E-15	1.49E+01
9*	4.50E+01	8.45E-03	4.30E-12	1.04E-05	3.31E-04	4.66E-16	1.53E+01
10*	9.20E+01	1.31E-02	3.99E-12	1.62E-05	3.48E-04	9.08E-16	1.50E+01
Mean:							1.42E+01
Standard Deviation:							1.35E+00

Table E-3. RADIONUCLIDE LEACHING DATA FOR COOPER CATION RESIN SAMPLE

(Continued)

## Ni-63 (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.44E+00	1.60E-07	1.15E-03	1.15E-03	4.61E-10	9.34E+00
2*	2.92E-01	2.47E-03	1.10E-10	1.98E-06	1.16E-03	1.78E-15	1.47E+01
3*	1.00E+00	4.54E-03	5.93E-11	3.63E-06	1.16E-03	1.79E-15	1.47E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	5.16E-03	4.77E-11	4.12E-06	NA	6.82E-15	1.42E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	2.06E-03	6.82E-13	1.65E-06	NA	1.17E-17	1.69E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							1.40E+01
Standard Deviation:							2.51E+00

## Sr-90 (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.75E-03	1.27E-04	9.11E-01	9.11E-01	2.87E-04	3.54E+00
2*	2.92E-01	1.57E-05	1.03E-07	1.85E-03	9.13E-01	1.56E-09	8.81E+00
3*	1.00E+00	2.15E-05	4.14E-08	2.53E-03	9.16E-01	8.72E-10	9.06E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	6.24E-05	8.50E-08	7.34E-03	NA	2.16E-08	7.67E+00
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	1.94E-05	9.42E-10	2.28E-03	NA	2.24E-11	1.07E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							7.94E+00
Standard Deviation:							2.40E+00

## Sb-125 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA
3*	1.00E+00	NA	NA	NA	NA	NA	NA
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	2.09E-02	9.95E-06	8.59E-01	NA	2.11E-04	3.68E+00
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	7.82E-04	3.73E-07	3.22E-02	NA	5.36E-07	6.27E+00
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

Table E-3. RADIONUCLIDE LEACHING DATA FOR COOPER CATION RESIN SAMPLE

(Continued)

## Cs-137 (FILTRATE)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.69E-04	> 2.27E-06	> 1.63E-02	> 1.63E-02	> 9.20E-08	< 7.04E+00
2*	2.92E-01	5.43E-04	> 1.83E-06	> 3.29E-02	> 4.92E-02	> 4.93E-07	< 6.31E+00
3*	1.00E+00	9.85E-04	> 9.75E-07	> 5.96E-02	> 1.09E-01	> 4.84E-07	< 6.32E+00
4*	2.00E+00	1.37E-04	> 9.59E-08	> 8.28E-03	> 1.17E-01	> 1.15E-08	< 7.94E+00
5*	3.00E+00	2.57E-04	> 1.80E-07	> 1.56E-02	> 1.33E-01	> 6.92E-08	< 7.16E+00
6*	4.00E+00	1.62E-04	> 1.14E-07	> 9.84E-03	> 1.43E-01	> 3.88E-08	< 7.41E+00
7*	5.00E+00	6.60E-05	> 4.63E-08	> 4.00E-03	> 1.47E-01	> 8.27E-09	< 8.08E+00
8*	1.70E+01	1.58E-03	> 9.22E-08	> 9.56E-02	> 2.42E-01	> 2.84E-08	< 7.55E+00
9*	4.50E+01	9.00E-03	> 2.25E-07	> 5.45E-01	> 7.87E-01	> 2.06E-07	< 6.69E+00
10*	9.20E+01	3.55E-03	> 5.29E-08	> 2.15E-01	> 1.00E+00	> 3.91E-07	< 6.41E+00

Mean:

Standard Deviation:

## Pu-241 (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.30E-02	2.60E-06	1.87E-02	1.87E-02	1.21E-07	6.92E+00
2*	2.92E-01	< 4.59E-04	< 2.08E-08	< 3.75E-04	< 1.91E-02	< 6.40E-11	> 1.02E+01
3*	1.00E+00	< 2.30E-04	< 3.06E-09	< 1.87E-04	< 1.93E-02	< 4.78E-12	> 1.13E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	< 2.30E-04	< 2.17E-09	< 1.87E-04	NA	< 1.41E-11	> 1.09E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	< 2.30E-04	< 7.75E-11	< 1.87E-04	NA	< 1.51E-13	> 1.28E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA

Mean:

Standard Deviation:

## Fe (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.85E+05	1.92E-05	1.39E-01	1.39E-01	6.63E-06	5.18E+00
2*	2.92E-01	< 1.62E+01	< 6.74E-10	< 1.21E-05	< 1.39E-01	< 6.71E-14	> 1.32E+01
3*	1.00E+00	7.50E+02	9.17E-09	5.61E-04	< 1.39E-01	4.29E-11	1.04E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	1.70E+03	1.48E-08	1.27E-03	NA	6.51E-10	9.19E+00
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	5.88E+02	1.82E-10	4.40E-04	NA	8.34E-13	1.21E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA

Mean:

Standard Deviation:



Table E-3. RADIONUCLIDE LEACHING DATA FOR COOPER CATION RESIN SAMPLE

(Continued)

Ni (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.09E+04	1.04E-05	7.51E-02	7.51E-02	1.95E-06	5.71E+00
2*	2.92E-01	1.22E+02	4.64E-08	8.34E-04	7.59E-02	3.17E-10	9.50E+00
3*	1.00E+00	1.22E+02	1.36E-08	8.34E-04	7.68E-02	9.48E-11	1.00E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	1.22E+02	9.66E-09	8.34E-04	NA	2.79E-10	9.55E+00
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	1.01E+02	2.87E-10	6.95E-04	NA	2.08E-12	1.17E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							9.29E+00
Standard Deviation:							1.96E+00

Co (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.24E+02	6.62E-06	4.77E-02	4.77E-02	7.85E-07	6.10E+00
2*	2.92E-01	< 8.10E+01	< 6.62E-07	< 1.19E-02	< 5.96E-02	< 6.47E-08	> 7.19E+00
3*	1.00E+00	< 8.10E+01	< 1.95E-07	< 1.19E-02	< 7.15E-02	< 1.93E-08	> 7.71E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	< 8.10E+01	< 1.38E-07	< 1.19E-02	NA	< 5.70E-08	> 7.24E+00
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	< 8.10E+01	< 4.93E-09	< 1.19E-02	NA	< 6.12E-10	> 9.21E+00
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

Cr (FILTRATE + FILTER)

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.13E+03	7.61E-06	5.48E-02	5.48E-02	1.04E-06	5.98E+00
2*	2.92E-01	< 1.62E+01	< 2.32E-08	< 4.17E-04	< 5.52E-02	< 7.93E-11	> 1.01E+01
3*	1.00E+00	< 1.62E+01	< 6.82E-09	< 4.17E-04	< 5.56E-02	< 2.37E-11	> 1.06E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	< 1.62E+01	< 4.83E-09	< 4.17E-04	NA	< 6.98E-11	> 1.02E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	6.08E+01	6.47E-10	1.56E-03	NA	1.05E-11	1.10E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							
Standard Deviation:							

Table E-4. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F33 SAMPLE

Mn-54

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.18E-03	1.53E-09	1.02E-11	7.89E-06	7.89E-06	4.26E-15	1.44E+01
2*	2.92E-01	3.13E-03	1.63E-09	1.09E-11	2.10E-05	2.89E-05	3.98E-14	1.34E+01
3*	1.00E+00	2.19E-03	3.35E-10	2.24E-12	1.47E-05	4.36E-05	5.82E-15	1.42E+01
4*	2.00E+00	3.09E-03	3.34E-10	2.24E-12	2.07E-05	6.43E-05	1.43E-14	1.38E+01
5*	3.00E+00	3.61E-03	3.91E-10	2.62E-12	2.42E-05	8.86E-05	3.32E-14	1.35E+01
6*	4.00E+00	6.06E-04	6.56E-11	4.39E-13	4.07E-06	9.26E-05	1.31E-15	1.49E+01
7*	5.00E+00	1.51E-03	1.63E-10	1.09E-12	1.01E-05	1.03E-04	1.05E-14	1.40E+01
8*	2.40E+01	3.17E-03	1.81E-11	1.21E-13	2.13E-05	1.24E-04	3.65E-16	1.54E+01
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Co-58

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.48E-02	9.71E-08	3.17E-09	2.45E-03	2.45E-03	4.10E-10	9.39E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	NA	NA	NA	NA	NA	NA	NA
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA	NA
6*	4.00E+00	1.08E-01	1.17E-08	3.82E-10	3.53E-03	NA	9.92E-10	9.00E+00
7*	5.00E+00	2.04E-01	2.21E-08	7.22E-10	6.68E-03	NA	4.57E-09	8.34E+00
8*	2.40E+01	NA	NA	NA	NA	NA	NA	NA
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Co-60

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	9.38E-03	1.22E-08	2.55E-11	1.96E-05	1.96E-05	2.64E-14	1.36E+01
2*	2.92E-01	1.79E-02	9.27E-09	1.94E-11	3.74E-05	5.71E-05	1.26E-13	1.29E+01
3*	1.00E+00	1.73E-02	2.64E-09	5.53E-12	3.63E-05	9.33E-05	3.55E-14	1.34E+01
4*	2.00E+00	2.66E-02	2.88E-09	6.02E-12	5.58E-05	1.49E-04	1.03E-13	1.30E+01
5*	3.00E+00	2.73E-02	2.95E-09	6.17E-12	5.71E-05	2.06E-04	1.84E-13	1.27E+01
6*	4.00E+00	1.18E-02	1.27E-09	2.66E-12	2.46E-05	2.31E-04	4.82E-14	1.33E+01
7*	5.00E+00	1.28E-02	1.39E-09	2.90E-12	2.69E-05	2.58E-04	7.39E-14	1.31E+01
8*	2.40E+01	3.35E-02	1.90E-10	3.98E-13	7.01E-05	3.28E-04	3.95E-15	1.44E+01
9*	5.30E+01	2.35E-02	8.77E-11	1.83E-13	4.93E-05	3.77E-04	2.44E-15	1.46E+01
10*	9.20E+01	2.94E-02	8.15E-11	1.71E-13	6.16E-05	4.39E-04	4.05E-15	1.44E+01
=====								
Mean:								1.36E+01
Standard Deviation:								6.49E-01

Table E-4. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F33 SAMPLE (Continued)

## Sb-125

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.31E-03	1.71E-09	1.74E-10	1.34E-04	1.34E-04	1.24E-12	1.19E+01
2*	2.92E-01	2.16E-03	1.12E-09	1.14E-10	2.21E-04	3.55E-04	4.40E-12	1.14E+01
3*	1.00E+00	3.00E-03	4.59E-10	4.68E-11	3.07E-04	6.63E-04	2.54E-12	1.16E+01
4*	2.00E+00	3.32E-03	3.60E-10	3.67E-11	3.40E-04	1.00E-03	3.84E-12	1.14E+01
5*	3.00E+00	3.12E-03	3.38E-10	3.45E-11	3.19E-04	1.32E-03	5.75E-12	1.12E+01
6*	4.00E+00	3.00E-03	3.25E-10	3.32E-11	3.07E-04	1.63E-03	7.50E-12	1.11E+01
7*	5.00E+00	2.41E-03	2.60E-10	2.66E-11	2.46E-04	1.88E-03	6.19E-12	1.12E+01
8*	2.40E+01	1.80E-03	1.02E-11	1.04E-12	1.84E-04	2.06E-03	2.71E-14	1.36E+01
9*	5.30E+01	1.59E-03	5.94E-12	6.07E-13	1.63E-04	2.22E-03	2.67E-14	1.36E+01
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								1.19E+01
Standard Deviation:								9.26E-01

## Cs-137

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.18E-02	1.53E-08	1.16E-07	8.98E-02	8.98E-02	5.52E-07	6.26E+00
2*	2.92E-01	1.41E-02	7.33E-09	5.59E-08	1.08E-01	1.98E-01	1.05E-06	5.98E+00
3*	1.00E+00	2.41E-02	3.67E-09	2.80E-08	1.84E-01	3.81E-01	1.09E-06	5.96E+00
4*	2.00E+00	2.13E-02	2.30E-09	1.75E-08	1.63E-01	5.44E-01	1.29E-06	5.89E+00
5*	3.00E+00	1.40E-02	1.51E-09	1.16E-08	1.07E-01	6.51E-01	1.44E-06	5.84E+00
6*	4.00E+00	8.65E-03	9.35E-10	7.13E-09	6.61E-02	7.17E-01	1.46E-06	5.84E+00
7*	5.00E+00	6.55E-03	7.09E-10	5.41E-09	5.01E-02	7.67E-01	1.45E-06	5.84E+00
8*	2.40E+01	1.97E-02	1.12E-10	8.54E-10	1.50E-01	9.17E-01	6.46E-07	6.19E+00
9*	5.30E+01	6.16E-03	2.30E-11	1.75E-10	4.70E-02	9.64E-01	3.96E-07	6.40E+00
10*	9.20E+01	1.81E-03	5.01E-12	3.82E-11	1.38E-02	9.78E-01	2.88E-07	6.54E+00
Mean:								6.07E+00
Standard Deviation:								2.44E-01

Table E-5. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F201 SAMPLE

## Mn-54

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.53E-03	1.85E-09	1.12E-11	9.32E-06	9.32E-06	6.24E-15	1.42E+01
2*	2.92E-01	1.31E-04	6.33E-11	3.85E-13	7.98E-07	1.01E-05	6.03E-17	1.62E+01
3*	1.00E+00	6.19E-04	8.80E-11	5.35E-13	3.77E-06	1.39E-05	4.03E-16	1.54E+01
4*	2.00E+00	1.43E-03	1.44E-10	8.76E-13	8.72E-06	2.26E-05	2.65E-15	1.46E+01
5*	3.00E+00	1.68E-03	1.69E-10	1.03E-12	1.02E-05	3.28E-05	6.18E-15	1.42E+01
6*	4.00E+00	2.31E-03	2.33E-10	1.42E-12	1.41E-05	4.69E-05	1.66E-14	1.38E+01
7*	5.00E+00	9.56E-04	9.62E-11	5.85E-13	5.82E-06	5.27E-05	3.64E-15	1.44E+01
8*	2.40E+01	NA	NA	NA	NA	NA	NA	NA
9*	5.30E+01	1.88E-03	6.52E-12	3.96E-14	1.14E-05	NA	1.38E-16	1.59E+01
10*	9.20E+01	3.65E-02	9.43E-11	5.73E-13	2.23E-04	NA	5.55E-14	1.33E+01
=====								
Mean:								
Standard Deviation:								

## Fe-55

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.56E-02	1.88E-08	1.40E-11	1.16E-05	1.16E-05	9.64E-15	1.40E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	2.19E-02	3.11E-09	2.31E-12	1.63E-05	NA	7.48E-15	1.41E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	2.74E-02	2.75E-09	2.04E-12	2.03E-05	NA	2.45E-14	1.36E+01
6*	4.00E+00	1.91E-02	1.93E-09	1.43E-12	1.42E-05	NA	1.69E-14	1.38E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	4.92E-03	2.61E-11	1.93E-14	3.66E-06	NA	1.13E-17	1.69E+01
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	4.65E-03	1.20E-11	8.90E-15	3.46E-06	NA	1.34E-17	1.69E+01
=====								
Mean: 1.49E+01								
Standard Deviation: 1.44E+00								

## Co-58

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.32E-01	1.60E-07	4.74E-09	3.93E-03	3.93E-03	1.11E-09	8.96E+00
2*	2.92E-01	7.58E-02	3.66E-08	1.09E-09	2.25E-03	6.18E-03	4.80E-10	9.32E+00
3*	1.00E+00	9.40E-02	1.34E-08	3.96E-10	2.79E-03	8.97E-03	2.21E-10	9.66E+00
4*	2.00E+00	1.70E-01	1.71E-08	5.08E-10	5.06E-03	1.40E-02	8.92E-10	9.05E+00
5*	3.00E+00	2.58E-01	2.60E-08	7.71E-10	7.68E-03	2.17E-02	3.49E-09	8.46E+00
6*	4.00E+00	3.28E-01	3.30E-08	9.79E-10	9.75E-03	3.15E-02	7.93E-09	8.10E+00
7*	5.00E+00	1.48E-01	1.49E-08	4.42E-10	4.40E-03	3.59E-02	2.08E-09	8.68E+00
8*	2.40E+01	NA	NA	NA	NA	NA	NA	NA
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Table E-5. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F201 SAMPLE (Continued)

## Co-60

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.06E-02	1.28E-08	2.43E-11	2.02E-05	2.02E-05	2.92E-14	1.35E+01
2*	2.92E-01	1.21E-02	5.83E-09	1.11E-11	2.30E-05	4.31E-05	5.00E-14	1.33E+01
3*	1.00E+00	1.75E-02	2.49E-09	4.72E-12	3.33E-05	7.64E-05	3.14E-14	1.35E+01
4*	2.00E+00	2.13E-02	2.14E-09	4.07E-12	4.05E-05	1.17E-04	5.73E-14	1.32E+01
5*	3.00E+00	2.01E-02	2.02E-09	3.84E-12	3.82E-05	1.55E-04	8.65E-14	1.31E+01
6*	4.00E+00	1.30E-02	1.31E-09	2.48E-12	2.47E-05	1.80E-04	5.08E-14	1.33E+01
7*	5.00E+00	1.42E-02	1.43E-09	2.71E-12	2.70E-05	2.07E-04	7.84E-14	1.31E+01
8*	2.40E+01	4.58E-01	2.43E-09	4.61E-12	8.71E-04	1.08E-03	6.41E-13	1.22E+01
9*	5.30E+01	4.93E-02	1.71E-10	3.25E-13	9.37E-05	1.17E-03	9.27E-15	1.40E+01
10*	9.20E+01	1.48E-01	3.81E-10	7.24E-13	2.81E-04	1.45E-03	8.85E-14	1.31E+01
Mean:								1.32E+01
Standard Deviation:								4.44E-01

## Ni-63

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.12E-02	1.36E-08	2.32E-10	1.92E-04	1.92E-04	2.65E-12	1.16E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	4.37E-02	6.21E-09	1.06E-10	7.47E-04	NA	1.58E-11	1.08E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	4.50E-02	4.53E-09	7.72E-11	7.68E-04	NA	3.50E-11	1.05E+01
6*	4.00E+00	3.12E-02	3.14E-09	5.36E-11	5.34E-04	NA	2.37E-11	1.06E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	1.37E-01	7.28E-10	1.24E-11	2.35E-03	NA	4.65E-12	1.13E+01
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	7.00E-02	1.81E-10	3.08E-12	1.20E-03	NA	1.60E-12	1.18E+01
Mean:								1.11E+01
Standard Deviation:								4.99E-01

## Sr-90

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	2.63E-04	3.17E-10	7.98E-09	6.62E-03	6.62E-03	3.14E-09	8.50E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	8.80E-04	1.25E-10	3.14E-09	2.22E-02	NA	1.39E-08	7.86E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	8.14E-04	8.20E-11	2.06E-09	2.05E-02	NA	2.49E-08	7.60E+00
6*	4.00E+00	7.49E-04	7.53E-11	1.89E-09	1.89E-02	NA	2.96E-08	7.53E+00
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	3.41E-03	1.81E-11	4.55E-10	8.60E-02	NA	6.25E-09	8.20E+00
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	3.28E-03	8.47E-12	2.13E-10	8.27E-02	NA	7.66E-09	8.12E+00
Mean:								7.97E+00
Standard Deviation:								3.42E-01

Table E-5. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F201 SAMPLE (Continued)

Sb-125

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.36E-03	1.64E-09	1.52E-10	1.26E-04	1.26E-04	1.14E-12	1.19E+01
2*	2.92E-01	1.81E-03	8.75E-10	8.11E-11	1.68E-04	2.94E-04	2.68E-12	1.16E+01
3*	1.00E+00	3.92E-03	5.57E-10	5.16E-11	3.64E-04	6.58E-04	3.75E-12	1.14E+01
4*	2.00E+00	6.16E-03	6.20E-10	5.74E-11	5.72E-04	1.23E-03	1.14E-11	1.09E+01
5*	3.00E+00	5.03E-03	5.06E-10	4.69E-11	4.67E-04	1.70E-03	1.29E-11	1.09E+01
6*	4.00E+00	4.92E-03	4.95E-10	4.59E-11	4.57E-04	2.15E-03	1.74E-11	1.08E+01
7*	5.00E+00	3.72E-03	3.74E-10	3.47E-11	3.46E-04	2.50E-03	1.28E-11	1.09E+01
8*	2.40E+01	2.52E-03	1.34E-11	1.24E-12	2.34E-04	2.73E-03	4.64E-14	1.33E+01
9*	5.30E+01	1.99E-03	6.89E-12	6.39E-13	1.84E-04	2.92E-03	3.59E-14	1.34E+01
10*	9.20E+01	4.58E-03	1.18E-11	1.10E-12	4.26E-04	3.34E-03	2.03E-13	1.27E+01
Mean:								1.18E+01
Standard Deviation:								9.77E-01

Cs-137

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.19E-02	1.43E-08	9.93E-08	8.24E-02	8.24E-02	4.87E-07	6.31E+00
2*	2.92E-01	1.62E-02	7.81E-09	5.41E-08	1.12E-01	1.94E-01	1.19E-06	5.92E+00
3*	1.00E+00	2.57E-02	3.65E-09	2.53E-08	1.78E-01	3.73E-01	1.09E-06	5.96E+00
4*	2.00E+00	2.07E-02	2.08E-09	1.44E-08	1.43E-01	5.15E-01	1.17E-06	5.93E+00
5*	3.00E+00	1.32E-02	1.33E-09	9.21E-09	9.17E-02	6.06E-01	1.20E-06	5.92E+00
6*	4.00E+00	8.38E-03	8.44E-10	5.84E-09	5.82E-02	6.66E-01	1.17E-06	5.93E+00
7*	5.00E+00	8.10E-03	8.16E-10	5.65E-09	5.62E-02	7.22E-01	1.22E-06	5.91E+00
8*	2.40E+01	2.18E-02	1.16E-10	8.01E-10	1.52E-01	8.74E-01	5.13E-07	6.29E+00
9*	5.30E+01	9.81E-03	3.40E-11	2.36E-10	6.81E-02	9.42E-01	3.63E-07	6.44E+00
10*	9.20E+01	3.09E-03	7.97E-12	5.52E-11	2.14E-02	9.63E-01	2.53E-07	6.60E+00
Mean:								6.12E+00
Standard Deviation:								2.48E-01

Pu-241

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	7.20E+03	< 5.67E-04	< 6.85E-10	< 2.90E-11	< 2.41E-05	< 2.41E-05	< 4.17E-14	> 1.34E+01
2*	2.52E+04	NA	NA	NA	NA	NA	NA	NA
3*	8.64E+04	< 2.84E-04	< 4.03E-11	< 1.71E-12	< 1.20E-05	NA	< 4.11E-15	> 1.44E+01
4*	1.73E+05	NA	NA	NA	NA	NA	NA	NA
5*	2.59E+05	< 1.13E-03	< 1.14E-10	< 4.84E-12	< 4.82E-05	NA	< 1.38E-13	> 1.29E+01
6*	3.46E+05	< 7.09E-04	< 7.14E-11	< 3.03E-12	< 3.01E-05	NA	< 7.56E-14	> 1.31E+01
7*	4.32E+05	NA	NA	NA	NA	NA	NA	NA
8*	2.07E+06	< 4.26E-04	< 2.25E-12	< 9.55E-14	< 1.81E-05	NA	< 2.76E-16	> 1.56E+01
9*	4.58E+06	NA	NA	NA	NA	NA	NA	NA
10*	7.95E+06	< 1.42E-04	< 3.66E-13	< 1.55E-14	< 5.02E-06	NA	< 4.06E-17	> 1.64E+01
Mean:								
Standard Deviation:								

Table E-5. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F201 SAMPLE (Continued)

Fe

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 9.79E+00	< 1.18E-05	< 3.02E-11	< 2.51E-05	< 2.51E-05	< 4.52E-14	> 1.33E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 9.79E+00	< 1.39E-06	< 3.56E-12	< 2.51E-05	NA	< 1.78E-14	> 1.37E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 9.79E+00	< 9.86E-07	< 2.52E-12	< 2.51E-05	NA	< 3.73E-14	> 1.34E+01
6*	4.00E+00	< 9.79E+00	< 9.86E-07	< 2.52E-12	< 2.51E-05	NA	< 5.25E-14	> 1.33E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	< 9.79E+00	< 5.19E-08	< 1.33E-13	< 2.51E-05	NA	< 5.31E-16	> 1.53E+01
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	< 9.79E+00	< 2.53E-08	< 6.46E-14	< 2.51E-05	NA	< 7.05E-16	> 1.52E+01
=====								
Mean:								
Standard Deviation:								

Ni

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.34E+01	8.87E-05	6.26E-10	5.20E-04	5.20E-04	1.94E-11	1.07E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	1.59E+02	2.26E-05	1.60E-10	1.13E-03	NA	3.59E-11	1.04E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	1.47E+02	1.48E-05	1.04E-10	1.04E-03	NA	6.40E-11	1.02E+01
6*	4.00E+00	1.35E+02	1.36E-05	9.57E-11	9.53E-04	NA	7.57E-11	1.01E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	4.04E+02	2.14E-06	1.51E-11	2.86E-03	NA	6.90E-12	1.12E+01
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	2.20E+02	5.69E-07	4.02E-12	1.56E-03	NA	2.72E-12	1.16E+01
=====								
Mean: 1.07E+01								
Standard Deviation: 5.19E-01								

Table E-5. RADIONUCLIDE LEACHING DATA FOR MILLSTONE-1 F201 SAMPLE (Continued)

Co

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 4.90E+01	< 5.91E-05	< 5.48E-08	< 4.55E-02	< 4.55E-02	< 1.49E-07	> 6.83E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 4.90E+01	< 6.96E-06	< 6.45E-09	< 4.55E-02	NA	< 5.85E-08	> 7.23E+00
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 4.90E+01	< 4.93E-06	< 4.57E-09	< 4.55E-02	NA	< 1.23E-07	> 6.91E+00
6*	4.00E+00	< 4.90E+01	< 4.93E-06	< 4.57E-09	< 4.55E-02	NA	< 1.72E-07	> 6.76E+00
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	< 4.90E+01	< 2.59E-07	< 2.40E-10	< 4.55E-02	NA	< 1.75E-09	> 8.76E+00
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	< 4.90E+01	< 1.26E-07	< 1.17E-10	< 4.55E-02	NA	< 2.32E-09	> 8.64E+00
=====								
Mean:								
Standard Deviation:								

Cr

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 9.79E+00	< 1.18E-05	< 5.01E-10	< 4.16E-04	< 4.16E-04	< 1.24E-11	> 1.09E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.00E+00	< 9.79E+00	< 1.39E-06	< 5.90E-11	< 4.16E-04	NA	< 4.89E-12	> 1.13E+01
4*	2.00E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 9.79E+00	< 9.86E-07	< 4.18E-11	< 4.16E-04	NA	< 1.02E-11	> 1.10E+01
6*	4.00E+00	< 9.79E+00	< 9.86E-07	< 4.18E-11	< 4.16E-04	NA	< 1.44E-11	> 1.08E+01
7*	5.00E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.40E+01	< 9.79E+00	< 5.19E-08	< 2.20E-12	< 4.16E-04	NA	< 1.46E-13	> 1.28E+01
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	< 9.79E+00	< 2.53E-08	< 1.07E-12	< 4.16E-04	NA	< 1.94E-13	> 1.27E+01
=====								
Mean:								
Standard Deviation:								



Table E-6. RADIONUCLIDE LEACHING DATA FOR PILGRIM SAMPLE

Mn-54

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 5.65E-03	< 4.53E-09	< 1.02E-10	< 1.28E-04	< 1.28E-04	< 1.59E-12	> 1.18E+01
2*	2.92E-01	< 3.09E-03	< 9.92E-10	< 2.24E-11	< 6.98E-05	< 1.97E-04	< 6.28E-13	> 1.22E+01
3*	1.04E+00	< 2.96E-02	< 2.65E-09	< 5.97E-11	< 6.69E-04	< 8.66E-04	< 1.58E-11	> 1.08E+01
4*	2.04E+00	< 7.61E-03	< 5.09E-10	< 1.15E-11	< 1.72E-04	< 1.04E-03	< 1.44E-12	> 1.18E+01
5*	3.00E+00	< 4.21E-02	< 2.93E-09	< 6.61E-11	< 9.51E-04	< 1.99E-03	< 7.98E-11	> 1.01E+01
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.08E+00	9.77E-04	6.05E-11	1.36E-12	2.21E-05	NA	6.16E-14	1.32E+01
8*	2.52E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Fe-55

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.12E-03	8.96E-10	2.84E-12	3.55E-06	3.55E-06	1.23E-15	1.49E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	< 2.30E-04	< 2.05E-11	< 6.51E-14	< 7.30E-07	NA	< 1.89E-17	> 1.67E+01
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	3.28E-04	2.29E-11	7.25E-14	1.04E-06	NA	9.61E-17	1.60E+01
6*	4.00E+00	< 6.56E-04	< 4.39E-11	< 1.39E-13	< 2.09E-06	NA	< 4.94E-16	> 1.53E+01
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	5.58E-04	1.85E-12	5.88E-15	1.77E-06	NA	3.34E-18	1.75E+01
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Co-58

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 1.27E+00	< 1.02E-06	---	---	---	---	---
2*	2.92E-01	< 5.03E+00	< 1.62E-06	---	---	---	---	---
3*	1.04E+00	< 3.18E+00	< 2.84E-07	---	---	---	---	---
4*	2.04E+00	< 1.77E+00	< 1.19E-07	---	---	---	---	---
5*	3.00E+00	< 5.92E+00	< 4.13E-07	---	---	---	---	---
6*	4.00E+00	ND	ND	ND	ND	ND	ND	ND
7*	5.08E+00	ND	ND	ND	ND	ND	ND	ND
8*	2.52E+01	ND	ND	ND	ND	ND	ND	ND
9*	4.84E+01	ND	ND	ND	ND	ND	ND	ND
=====								
Mean:								
Standard Deviation:								

Table E-6. RADIONUCLIDE LEACHING DATA FOR PILGRIM SAMPLE

(Continued)

## Co-60

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED ( $\mu\text{Ci}$ )	RELEASE RATE ( $\mu\text{Ci}/\text{cm}^2/\text{s}$ )	RELEASE RATE ( $\text{F}/\text{cm}^2/\text{s}$ )	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ( $\text{cm}^2/\text{s}$ )	LEACH INDEX
1*	8.33E-02	1.54E+00	1.24E-06	2.05E-09	2.55E-03	2.55E-03	6.38E-10	9.20E+00
2*	2.92E-01	2.50E+00	8.04E-07	1.33E-09	4.14E-03	6.70E-03	2.21E-09	8.66E+00
3*	1.04E+00	4.81E+00	4.30E-07	7.09E-10	7.95E-03	1.46E-02	2.24E-09	8.65E+00
4*	2.04E+00	2.93E+00	1.96E-07	3.24E-10	4.85E-03	1.95E-02	1.15E-09	8.94E+00
5*	3.00E+00	1.79E+00	1.25E-07	2.06E-10	2.97E-03	2.25E-02	7.77E-10	9.11E+00
6*	4.00E+00	1.51E+00	1.01E-07	1.67E-10	2.50E-03	2.50E-02	7.07E-10	9.15E+00
7*	5.08E+00	1.40E+00	8.65E-08	1.43E-10	2.31E-03	2.73E-02	6.74E-10	9.17E+00
8*	2.52E+01	1.01E+01	3.34E-08	5.52E-11	1.67E-02	4.39E-02	2.94E-10	9.53E+00
9*	4.84E+01	4.05E+00	1.17E-08	1.93E-11	6.71E-03	5.06E-02	9.80E-11	1.00E+01
Mean:								9.16E+00
Standard Deviation:								3.98E-01

## Ni-63

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED ( $\mu\text{Ci}$ )	RELEASE RATE ( $\mu\text{Ci}/\text{cm}^2/\text{s}$ )	RELEASE RATE ( $\text{F}/\text{cm}^2/\text{s}$ )	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ( $\text{cm}^2/\text{s}$ )	LEACH INDEX
1*	8.33E-02	8.58E-01	6.89E-07	2.11E-08	2.63E-02	2.63E-02	6.76E-08	7.17E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	3.28E+00	2.93E-07	8.96E-09	1.00E-01	NA	3.57E-07	6.45E+00
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	1.09E+00	7.61E-08	2.33E-09	3.35E-02	NA	9.90E-08	7.00E+00
6*	4.00E+00	7.80E-01	5.22E-08	1.60E-09	2.39E-02	NA	6.49E-08	7.19E+00
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	4.68E+00	1.55E-08	4.75E-10	1.44E-01	NA	2.19E-08	7.66E+00
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								7.09E+00
Standard Deviation:								3.90E-01

## Sr-90

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED ( $\mu\text{Ci}$ )	RELEASE RATE ( $\mu\text{Ci}/\text{cm}^2/\text{s}$ )	RELEASE RATE ( $\text{F}/\text{cm}^2/\text{s}$ )	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY ( $\text{cm}^2/\text{s}$ )	LEACH INDEX
1*	8.33E-02	4.58E-04	3.68E-10	2.10E-08	2.62E-02	2.62E-02	6.71E-08	7.17E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	7.20E-04	6.43E-11	3.67E-09	4.12E-02	NA	6.00E-08	7.22E+00
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	1.57E-04	1.09E-11	6.25E-10	8.99E-03	NA	7.12E-09	8.15E+00
6*	4.00E+00	1.78E-04	1.19E-11	6.81E-10	1.02E-02	NA	1.18E-08	7.93E+00
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	3.07E-04	1.02E-12	5.83E-11	1.76E-02	NA	3.29E-10	9.48E+00
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								7.99E+00
Standard Deviation:								8.38E-01

Table E-6. RADIONUCLIDE LEACHING DATA FOR PILGRIM SAMPLE

(Continued)

## Sb-125

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 3.87E-03	< 3.11E-09	< 3.17E-09	< 3.96E-03	< 3.96E-03	< 1.53E-09	> 8.82E+00
2*	2.92E-01	< 2.59E-02	< 8.30E-09	< 8.47E-09	< 2.64E-02	< 3.04E-02	< 8.99E-08	> 7.05E+00
3*	1.04E+00	9.82E-03	8.78E-10	8.95E-10	1.00E-02	4.04E-02	3.56E-09	8.45E+00
4*	2.04E+00	< 8.48E-03	< 5.67E-10	< 5.78E-10	< 8.66E-03	< 4.91E-02	< 3.66E-09	> 8.44E+00
5*	3.00E+00	< 2.13E-02	< 1.48E-09	< 1.51E-09	< 2.17E-02	< 7.08E-02	< 4.17E-08	> 7.38E+00
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	NA	NA	NA	NA	NA	NA	NA
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								
Standard Deviation:								

## Cs-137

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 2.25E-03	< 1.81E-09	< 5.16E-10	< 6.45E-04	< 6.45E-04	< 4.06E-11	> 1.04E+01
2*	2.92E-01	< 2.51E-04	< 8.06E-11	< 2.30E-11	< 7.18E-05	< 7.16E-04	< 6.64E-13	> 1.22E+01
3*	1.04E+00	5.13E-04	4.58E-11	1.31E-11	1.47E-04	< 8.63E-04	7.61E-13	1.21E+01
4*	2.04E+00	< 2.09E-03	< 1.40E-10	< 3.98E-11	< 5.97E-04	< 1.46E-03	< 1.74E-11	> 1.08E+01
5*	3.00E+00	< 4.97E-03	< 3.47E-10	< 9.89E-11	< 1.42E-03	< 2.88E-03	< 1.79E-10	> 9.75E+00
6*	4.00E+00	1.26E-03	8.45E-11	2.41E-11	3.62E-04	< 3.24E-03	1.48E-11	1.08E+01
7*	5.08E+00	3.26E-04	2.02E-11	5.76E-12	9.32E-05	< 3.34E-03	1.10E-12	1.20E+01
8*	2.52E+01	7.05E-04	2.34E-12	6.68E-13	2.02E-04	< 3.54E-03	4.32E-14	1.34E+01
9*	4.84E+01	1.52E-03	4.39E-12	1.25E-12	4.34E-04	< 3.97E-03	4.11E-13	1.24E+01
Mean:								
Standard Deviation:								

## Pu-241

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	< 7.04E-04	< 5.65E-10	< 1.38E-10	< 1.73E-04	< 1.73E-04	< 2.91E-12	> 1.15E+01
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	< 8.80E-04	< 7.86E-11	< 1.92E-11	< 2.16E-04	NA	< 1.65E-12	> 1.18E+01
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 7.04E-04	< 4.91E-11	< 1.20E-11	< 1.73E-04	NA	< 2.63E-12	> 1.16E+01
6*	4.00E+00	< 5.28E-04	< 3.53E-11	< 8.64E-12	< 1.29E-04	NA	< 1.90E-12	> 1.17E+01
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	< 7.04E-04	< 2.34E-12	< 5.72E-13	< 1.73E-04	NA	< 3.16E-14	> 1.35E+01
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								
Standard Deviation:								

Table E-6. RADIONUCLIDE LEACHING DATA FOR PILGRIM SAMPLE

(Continued)

Fe

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	< 1.22E+01	< 1.09E-06	< 8.52E-12	< 9.55E-05	NA	< 3.23E-13	> 1.25E+01
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 1.22E+01	< 8.53E-07	< 6.64E-12	< 9.55E-05	NA	< 8.05E-13	> 1.21E+01
6*	4.00E+00	< 1.22E+01	< 8.19E-07	< 6.38E-12	< 9.55E-05	NA	< 1.03E-12	> 1.20E+01
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	< 1.22E+01	< 4.06E-08	< 3.16E-13	< 9.55E-05	NA	< 9.69E-15	> 1.40E+01
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Ni

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	1.85E+03	1.65E-04	1.01E-08	1.14E-01	NA	4.56E-07	6.34E+00
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	6.43E+02	4.48E-05	2.74E-09	3.94E-02	NA	1.37E-07	6.86E+00
6*	4.00E+00	5.97E+02	3.99E-05	2.44E-09	3.66E-02	NA	1.52E-07	6.82E+00
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	2.60E+03	8.64E-06	5.28E-10	1.59E-01	NA	2.70E-08	7.57E+00
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean: 6.90E+00								
Standard Deviation: 4.38E-01								

Table E-6. RADIONUCLIDE LEACHING DATA FOR PILGRIM SAMPLE

(Continued)

Cr

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	< 1.22E+01	< 1.09E-06	NA	NA	NA	NA	NA
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 1.22E+01	< 8.53E-07	NA	NA	NA	NA	NA
6*	4.00E+00	< 1.22E+01	< 8.19E-07	NA	NA	NA	NA	NA
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	< 1.22E+01	< 4.06E-08	NA	NA	NA	NA	NA
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								NA
Standard Deviation:								NA

Co

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA	NA
3*	1.04E+00	< 6.12E+01	< 5.47E-06	< 4.88E-09	< 5.47E-02	NA < 1.06E-07	> 6.98E+00	NA
4*	2.04E+00	NA	NA	NA	NA	NA	NA	NA
5*	3.00E+00	< 6.12E+01	< 4.27E-06	< 3.80E-09	< 5.47E-02	NA < 2.64E-07	> 6.58E+00	NA
6*	4.00E+00	< 6.12E+01	< 4.09E-06	< 3.65E-09	< 5.47E-02	NA < 3.40E-07	> 6.47E+00	NA
7*	5.08E+00	NA	NA	NA	NA	NA	NA	NA
8*	2.52E+01	< 6.12E+01	< 2.03E-07	< 1.81E-10	< 5.47E-02	NA < 3.18E-09	> 8.50E+00	NA
9*	4.84E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								
Standard Deviation:								

Table E-7. RADIONUCLIDE LEACHING DATA FOR PEACH BOTTOM-2 SAMPLE

Mn-54

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	< 7.54E-04	< 1.77E-10	< 9.26E-12	< 3.94E-05	< 3.94E-05	< 4.26E-14	> 1.34E+01
2*	9.88E-01	< 1.94E-03	< 1.91E-10	< 9.99E-12	< 1.01E-04	< 1.41E-04	< 4.00E-13	> 1.24E+01
3*	2.23E+00	< 2.14E-03	< 1.18E-10	< 6.18E-12	< 1.12E-04	< 2.52E-04	< 4.02E-13	> 1.24E+01
4*	3.06E+00	1.96E-03	1.61E-10	8.41E-12	1.02E-04	3.55E-04	1.27E-12	1.19E+01
5*	4.04E+00	< 5.70E-04	< 3.98E-11	< 2.08E-12	< 2.98E-05	< 3.84E-04	< 1.05E-13	> 1.30E+01
6*	7.02E+00	< 8.63E-04	< 1.98E-11	< 1.04E-12	< 4.51E-05	< 4.29E-04	< 3.98E-14	> 1.34E+01
7*	2.23E+01	< 6.53E-03	< 2.93E-11	< 1.53E-12	< 3.41E-04	< 7.70E-04	< 2.18E-13	> 1.27E+01
8*	5.61E+01	4.97E-03	1.00E-11	5.26E-13	2.59E-04	1.03E-03	7.01E-14	1.32E+01
9*	9.51E+01	< 4.24E-03	< 7.44E-12	< 3.89E-13	< 2.21E-04	< 1.25E-03	< 7.66E-14	> 1.31E+01
=====								
Mean:								
Standard Deviation:								

Co-60

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	7.11E-02	1.67E-08	1.24E-11	5.29E-05	5.29E-05	7.68E-14	1.31E+01
2*	9.88E-01	8.85E-02	8.71E-09	6.49E-12	6.58E-05	1.19E-04	1.69E-13	1.28E+01
3*	2.23E+00	1.05E-01	5.80E-09	4.32E-12	7.79E-05	1.97E-04	1.96E-13	1.27E+01
4*	3.06E+00	5.02E-02	4.13E-09	3.07E-12	3.74E-05	2.34E-04	1.69E-13	1.28E+01
5*	4.04E+00	6.33E-02	4.43E-09	3.30E-12	4.71E-05	2.81E-04	2.61E-13	1.26E+01
6*	7.02E+00	1.08E-01	2.48E-09	1.85E-12	8.03E-05	3.61E-04	1.26E-13	1.29E+01
7*	2.23E+01	3.09E-01	1.39E-09	1.04E-12	2.30E-04	5.91E-04	9.91E-14	1.30E+01
8*	5.61E+01	5.80E-01	1.17E-09	8.73E-13	4.31E-04	1.02E-03	1.93E-13	1.27E+01
9*	9.51E+01	4.17E-01	7.33E-10	5.46E-13	3.10E-04	1.33E-03	1.51E-13	1.28E+01
=====								
Mean:								1.28E+01
Standard Deviation:								1.53E-01

Sr-90

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	1.39E-05	3.27E-12	1.79E-09	7.63E-03	7.63E-03	1.60E-09	8.80E+00
2*	9.88E-01	NA	NA	NA	NA	NA	NA	NA
3*	2.23E+00	8.35E-06	4.62E-13	2.53E-10	4.58E-03	NA	6.76E-10	9.17E+00
4*	3.06E+00	NA	NA	NA	NA	NA	NA	NA
5*	4.04E+00	< 8.35E-06	< 5.83E-13	< 3.20E-10	< 4.58E-03	NA	< 2.47E-09	> 8.61E+00
6*	7.02E+00	< 8.35E-06	< 1.92E-13	< 1.05E-10	< 4.58E-03	NA	< 4.10E-10	> 9.39E+00
7*	2.23E+01	1.67E-05	7.50E-14	4.12E-11	9.15E-03	NA	1.57E-10	9.80E+00
8*	5.61E+01	NA	NA	NA	NA	NA	NA	NA
9*	9.51E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

Table E-7. RADIONUCLIDE LEACHING DATA FOR PEACH BOTTOM-2 SAMPLE (Continued)

Sb-125

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	< 8.21E-04	< 1.93E-10	---	---	---	---	---
2*	9.88E-01	< 1.74E-03	< 1.71E-10	---	---	---	---	---
3*	2.23E+00	< 3.52E-03	< 1.95E-10	---	---	---	---	---
4*	3.06E+00	< 6.98E-04	< 5.73E-11	---	---	---	---	---
5*	4.04E+00	< 4.19E-04	< 2.93E-11	---	---	---	---	---
6*	7.02E+00	< 1.01E-03	< 2.33E-11	---	---	---	---	---
7*	2.23E+01	< 8.90E-03	< 4.00E-11	---	---	---	---	---
8*	5.61E+01	< 3.94E-03	< 7.97E-12	---	---	---	---	---
9*	9.51E+01	< 5.30E-03	< 9.30E-12	---	---	---	---	---
=====								
Mean:								
Standard Deviation:								

Cs-137

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	8.03E-03	1.88E-09	5.85E-09	2.49E-02	2.49E-02	1.70E-08	7.77E+00
2*	9.88E-01	8.22E-03	8.08E-10	2.51E-09	2.55E-02	5.03E-02	2.52E-08	7.60E+00
3*	2.23E+00	9.05E-03	5.00E-10	1.55E-09	2.81E-02	7.84E-02	2.54E-08	7.59E+00
4*	3.06E+00	4.32E-03	3.55E-10	1.10E-09	1.34E-02	9.18E-02	2.17E-08	7.66E+00
5*	4.04E+00	4.04E-03	2.82E-10	8.77E-10	1.25E-02	1.04E-01	1.85E-08	7.73E+00
6*	7.02E+00	7.69E-03	1.77E-10	5.49E-10	2.38E-02	1.28E-01	1.11E-08	7.95E+00
7*	2.23E+01	2.49E-02	1.12E-10	3.48E-10	7.72E-02	2.05E-01	1.68E-08	7.78E+00
8*	5.61E+01	2.89E-02	5.84E-11	1.81E-10	8.96E-02	2.95E-01	1.49E-08	7.83E+00
9*	9.51E+01	2.11E-02	3.71E-11	1.15E-10	6.55E-02	3.60E-01	1.43E-08	7.85E+00
=====								
Mean:								7.75E+00
Standard Deviation:								1.12E-01

Pu-241

SAMPLE NUMBER	TOTAL TIME (days)	ACTIVITY LEACHED (uCi)	RELEASE RATE (uCi/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	< 1.05E-03	< 2.46E-10	< 4.24E-10	< 1.80E-03	< 1.80E-03	< 8.92E-11	> 1.00E+01
2*	9.88E-01	NA	NA	NA	NA	NA	NA	NA
3*	2.23E+00	< 1.50E-03	< 8.27E-11	< 1.43E-10	< 2.57E-03	NA	< 2.14E-10	> 9.67E+00
4*	3.06E+00	NA	NA	NA	NA	NA	NA	NA
5*	4.04E+00	< 8.98E-04	< 6.27E-11	< 1.08E-10	< 1.54E-03	NA	< 2.81E-10	> 9.55E+00
6*	7.02E+00	< 8.98E-04	< 2.06E-11	< 3.55E-11	< 1.54E-03	NA	< 4.67E-11	> 1.03E+01
7*	2.23E+01	< 1.20E-03	< 5.38E-12	< 9.27E-12	< 2.06E-03	NA	< 7.94E-12	> 1.11E+01
8*	5.61E+01	NA	NA	NA	NA	NA	NA	NA
9*	9.51E+01	NA	NA	NA	NA	NA	NA	NA
=====								
Mean:								
Standard Deviation:								

## **APPENDIX F**

### **CHELATING AGENT LEACHING DATA FOR WASTE-FORM SAMPLES**





## **APPENDIX F**

### **CHELATING AGENT LEACHING DATA FOR WASTE-FORM SAMPLES**

Chelating agent leaching data for waste-form samples are given in Tables F-1 through F-7.

Table F-1. CHELATING AGENT LEACHING DATA FOR INDIAN POINT-3 SAMPLE

## Formic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.16E+04	9.43E-03	2.12E-08	2.61E-02	2.61E-02	6.40E-08	7.19E+00
2*	2.92E-01	2.18E+04	7.07E-03	1.59E-08	4.90E-02	7.52E-02	2.97E-07	6.53E+00
3*	1.00E+00	3.27E+04	3.13E-03	7.04E-09	7.37E-02	1.49E-01	2.01E-07	6.70E+00
4*	2.00E+00	3.27E+04	2.21E-03	4.99E-09	7.37E-02	2.23E-01	1.99E-07	6.70E+00
5*	3.00E+00	3.01E+04	2.04E-03	4.59E-09	6.79E-02	2.90E-01	2.44E-07	6.61E+00
6*	4.00E+00	2.39E+04	1.61E-03	3.63E-09	5.37E-02	3.44E-01	2.60E-07	6.58E+00
7*	5.00E+00	2.10E+04	1.42E-03	3.19E-09	4.72E-02	3.91E-01	2.86E-07	6.54E+00
8*	1.80E+01	9.70E+04	5.05E-04	1.14E-09	2.17E-01	6.10E-01	2.39E-07	6.62E+00
9*	4.60E+01	9.58E+04	2.31E-04	5.21E-10	2.16E-01	8.26E-01	2.41E-07	6.62E+00
10*	8.70E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								6.68E+00
Standard Deviation:								1.91E-01

## Picolinic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	7.43E+03	6.04E-03	1.32E-08	1.62E-02	1.62E-02	2.47E-08	7.61E+00
2*	2.92E-01	1.40E+04	4.54E-03	9.92E-09	3.06E-02	4.68E-02	1.15E-07	6.94E+00
3*	1.00E+00	3.21E+04	3.07E-03	6.70E-09	7.01E-02	1.17E-01	1.82E-07	6.74E+00
4*	2.00E+00	2.92E+04	1.97E-03	4.32E-09	6.38E-02	1.81E-01	1.85E-07	6.73E+00
5*	3.00E+00	2.19E+04	1.48E-03	3.24E-09	4.79E-02	2.29E-01	1.47E-07	6.83E+00
6*	4.00E+00	1.76E+04	1.19E-03	2.60E-09	3.84E-02	2.67E-01	1.56E-07	6.81E+00
7*	5.00E+00	1.17E+04	7.94E-04	1.74E-09	2.57E-02	2.93E-01	1.46E-07	6.84E+00
8*	1.80E+01	7.66E+04	3.99E-04	8.71E-10	1.67E-01	4.60E-01	1.17E-07	6.93E+00
9*	4.60E+01	6.43E+04	1.55E-04	3.40E-10	1.41E-01	6.01E-01	8.98E-08	7.05E+00
10*	8.70E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								6.94E+00
Standard Deviation:								2.54E-01

Table F-2. CHELATING AGENT LEACHING DATA FOR COOPER MIXED BED RESIN SAMPLE

## Oxalic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.85E+03	3.11E-07	2.23E-03	2.23E-03	1.52E-09	8.82E+00
2*	2.92E-01	3.02E+03	9.74E-08	1.75E-03	3.99E-03	1.23E-09	8.91E+00
3*	1.00E+00	2.00E+03	1.90E-08	1.16E-03	5.14E-03	1.61E-10	9.79E+00
4*	2.00E+00	1.42E+03	9.55E-09	8.24E-04	5.97E-03	1.01E-10	1.00E+01
5*	3.00E+00	1.44E+03	9.65E-09	8.33E-04	6.80E-03	1.75E-10	NA
6*	4.00E+00	1.46E+03	9.82E-09	8.47E-04	7.65E-03	2.54E-10	9.59E+00
7*	5.00E+00	1.38E+03	9.28E-09	8.01E-04	8.45E-03	2.92E-10	9.53E+00
8*	2.10E+01	1.02E+03	4.29E-10	5.91E-04	9.04E-03	1.62E-12	1.18E+01
9*	4.40E+01	1.10E+03	3.20E-10	6.36E-04	9.68E-03	2.44E-12	1.16E+01
10*	9.20E+01	1.43E+03	2.00E-10	8.28E-04	1.05E-02	1.99E-12	1.17E+01
Mean:							1.02E+01
Standard Deviation:							1.12E+00

## Citric Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	6.21E+03	3.26E-07	2.35E-03	2.35E-03	1.68E-09	8.78E+00
2*	2.92E-01	5.01E+03	1.05E-07	1.89E-03	4.24E-03	1.44E-09	8.84E+00
3*	1.00E+00	1.98E+03	1.22E-08	7.47E-04	4.98E-03	6.71E-11	1.02E+01
4*	2.00E+00	1.11E+03	4.86E-09	4.19E-04	5.40E-03	2.61E-11	1.06E+01
5*	3.00E+00	1.11E+03	4.85E-09	4.19E-04	5.82E-03	4.41E-11	NA
6*	4.00E+00	1.11E+03	4.85E-09	4.18E-04	6.24E-03	6.20E-11	1.02E+01
7*	5.00E+00	1.93E+03	8.46E-09	7.30E-04	6.97E-03	2.43E-10	9.61E+00
8*	2.10E+01	1.32E+03	3.60E-10	4.97E-04	7.47E-03	1.14E-12	1.19E+01
9*	4.40E+01	1.50E+03	2.85E-10	5.65E-04	8.03E-03	1.93E-12	1.17E+01
10*	9.20E+01	1.07E+03	9.77E-11	4.05E-04	8.44E-03	4.76E-13	1.23E+01
Mean:							1.05E+01
Standard Deviation:							1.23E+00

Table F-3. CHELATING AGENT LEACHING DATA FOR COOPER CATION RESIN SAMPLE

## Oxalic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA
2*	2.92E-01	NA	NA	NA	NA	NA	NA
3*	1.00E+00	NA	NA	NA	NA	NA	NA
4*	2.00E+00	NA	NA	NA	NA	NA	NA
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							NA
Standard Deviation:							NA

## Citric Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (F/s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.35E+02	3.99E-06	2.87E-02	2.87E-02	2.85E-07	6.55E+00
2*	2.92E-01	NA	NA	NA	NA	NA	NA
3*	1.00E+00	NA	NA	NA	NA	NA	NA
4*	2.00E+00	2.01E+02	1.99E-07	1.72E-02	NA	4.98E-08	7.30E+00
5*	3.00E+00	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA
7*	5.00E+00	NA	NA	NA	NA	NA	NA
8*	1.70E+01	NA	NA	NA	NA	NA	NA
9*	4.50E+01	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA
Mean:							6.92E+00
Standard Deviation:							3.79E-01

Table F-4. CHELATING AGENT LEACHING DATA FOR MILLSTONE-1 F33 SAMPLE

## Oxalic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.85E+03	5.00E-03	2.89E-09	2.23E-03	2.23E-03	4.42E-10	9.35E+00
2*	2.92E-01	3.02E+03	1.57E-03	9.08E-10	1.75E-03	3.99E-03	3.58E-10	9.45E+00
3*	1.00E+00	1.24E+03	1.89E-04	4.20E-10	2.76E-03	6.34E-03	2.05E-10	9.69E+00
4*	2.00E+00	1.41E+03	1.52E-04	3.37E-10	3.13E-03	9.47E-03	3.25E-10	9.49E+00
5*	3.00E+00	1.13E+03	1.22E-04	2.71E-10	2.51E-03	1.20E-02	3.54E-10	9.45E+00
6*	4.00E+00	9.56E+02	1.03E-04	2.29E-10	2.12E-03	1.41E-02	3.58E-10	NA
7*	5.00E+00	7.88E+02	8.52E-05	1.89E-10	1.75E-03	1.59E-02	3.14E-10	9.50E+00
8*	2.40E+01	1.26E+03	7.16E-06	1.59E-11	2.79E-03	1.86E-02	6.28E-12	1.12E+01
9*	5.30E+01	1.80E+03	6.70E-06	1.49E-11	3.99E-03	2.26E-02	1.60E-11	1.08E+01
10*	9.20E+01	6.43E+02	1.78E-06	3.95E-12	1.43E-03	2.41E-02	2.18E-12	1.17E+01
Mean:								1.01E+01
Standard Deviation:								8.19E-01

## Citric Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.40E+02	1.82E-04	4.70E-10	3.63E-04	3.63E-04	9.00E-12	1.10E+01
2*	2.92E-01	1.69E+02	8.76E-05	2.26E-10	4.36E-04	7.99E-04	1.72E-11	1.08E+01
3*	1.00E+00	2.92E+02	4.46E-05	1.15E-10	7.55E-04	1.55E-03	1.53E-11	1.08E+01
4*	2.00E+00	4.74E+02	5.13E-05	1.32E-10	1.23E-03	2.78E-03	4.99E-11	1.03E+01
5*	3.00E+00	1.46E+02	1.58E-05	4.08E-11	3.78E-04	3.16E-03	8.06E-12	1.11E+01
6*	4.00E+00	1.64E+02	1.77E-05	4.57E-11	4.23E-04	3.58E-03	1.42E-11	1.08E+01
7*	5.00E+00	9.83E+02	1.06E-04	2.74E-10	2.54E-03	6.12E-03	6.61E-10	9.18E+00
8*	2.40E+01	9.95E+02	5.67E-06	1.46E-11	2.57E-03	8.69E-03	5.32E-12	1.13E+01
9*	5.30E+01	1.05E+03	3.91E-06	1.01E-11	2.71E-03	1.14E-02	7.38E-12	1.11E+01
10*	9.20E+01	1.08E+03	2.99E-06	7.72E-12	2.79E-03	1.42E-02	8.29E-12	1.11E+01
Mean:								1.08E+01
Standard Deviation:								5.84E-01

## EDTA

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.63E+03	2.12E-03	5.64E-09	4.35E-03	4.35E-03	1.30E-09	8.89E+00
2*	2.92E-01	3.57E+03	1.85E-03	4.93E-09	9.51E-03	1.39E-02	8.15E-09	8.09E+00
3*	1.00E+00	7.10E+03	1.08E-03	2.88E-09	1.89E-02	3.28E-02	9.65E-09	8.02E+00
4*	2.00E+00	1.02E+04	1.10E-03	2.92E-09	2.71E-02	5.99E-02	2.44E-08	7.61E+00
5*	3.00E+00	5.61E+03	6.07E-04	1.61E-09	1.49E-02	7.48E-02	1.26E-08	7.90E+00
6*	4.00E+00	3.98E+03	4.30E-04	1.14E-09	1.06E-02	8.54E-02	8.92E-09	8.05E+00
7*	5.00E+00	2.55E+03	2.75E-04	7.32E-10	6.78E-03	9.22E-02	4.71E-09	8.33E+00
8*	2.40E+01	1.72E+04	9.79E-05	2.60E-10	4.58E-02	1.38E-01	1.69E-09	8.77E+00
9*	5.30E+01	1.93E+04	7.19E-05	1.91E-10	5.13E-02	1.89E-01	2.65E-09	8.58E+00
10*	9.20E+01	1.39E+04	3.86E-05	1.03E-10	3.71E-02	2.26E-01	3.85E-09	8.41E+00
Mean:								8.26E+00
Standard Deviation:								3.83E-01

Table F-5. CHELATING AGENT LEACHING DATA FOR MILLSTONE-1 F201 SAMPLE

## Oxalic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	1.03E+03	4.98E-04	1.00E-09	2.08E-03	NA	4.10E-10	9.39E+00
3*	1.00E+00	NA	NA	NA	NA	NA	NA	NA
4*	2.00E+00	1.57E+03	1.58E-04	3.19E-10	3.17E-03	NA	3.51E-10	9.45E+00
5*	3.00E+00	NA	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	7.69E+02	7.74E-05	1.56E-10	1.55E-03	NA	2.58E-10	9.59E+00
8*	2.40E+01	NA	NA	NA	NA	NA	NA	NA
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								9.48E+00
Standard Deviation:								8.34E-02

## Citric Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	1.38E+02	6.69E-05	1.57E-10	3.25E-04	NA	1.00E-11	1.10E+01
3*	1.00E+00	NA	NA	NA	NA	NA	NA	NA
4*	2.00E+00	2.43E+02	2.45E-05	5.74E-11	5.71E-04	NA	1.14E-11	1.09E+01
5*	3.00E+00	NA	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	9.39E+02	9.45E-05	2.21E-10	2.20E-03	NA	5.22E-10	9.28E+00
8*	2.40E+01	NA	NA	NA	NA	NA	NA	NA
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								1.04E+01
Standard Deviation:								7.97E-01

## EDTA

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	NA	NA	NA	NA	NA	NA	NA
2*	2.92E-01	2.14E+03	1.04E-03	2.50E-09	5.19E-03	NA	2.55E-09	8.59E+00
3*	1.00E+00	NA	NA	NA	NA	NA	NA	NA
4*	2.00E+00	8.00E+03	8.05E-04	1.94E-09	1.94E-02	NA	1.31E-08	7.88E+00
5*	3.00E+00	NA	NA	NA	NA	NA	NA	NA
6*	4.00E+00	NA	NA	NA	NA	NA	NA	NA
7*	5.00E+00	2.47E+03	2.49E-04	6.01E-10	5.98E-03	NA	3.84E-09	8.42E+00
8*	2.40E+01	NA	NA	NA	NA	NA	NA	NA
9*	5.30E+01	NA	NA	NA	NA	NA	NA	NA
10*	9.20E+01	NA	NA	NA	NA	NA	NA	NA
Mean:								8.30E+00
Standard Deviation:								3.02E-01

Table F-6. CHELATING AGENT LEACHING DATA FOR PILGRIM SAMPLE

## Oxalic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.06E+03	8.49E-04	2.49E-08	3.10E-02	3.10E-02	9.41E-08	NA
2*	2.92E-01	1.63E+03	5.23E-04	1.53E-08	4.78E-02	7.88E-02	2.94E-07	6.53E+00
3*	1.04E+00	1.56E+03	1.40E-04	4.09E-09	4.58E-02	1.25E-01	7.42E-08	7.13E+00
4*	2.04E+00	1.11E+03	7.42E-05	2.17E-09	3.26E-02	1.57E-01	5.17E-08	7.29E+00
5*	3.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.57E-01	0.00E+00	NA
6*	4.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.57E-01	0.00E+00	NA
7*	5.08E+00	2.19E+03	1.36E-04	3.97E-09	6.43E-02	2.21E-01	8.26E-08	7.08E+00
8*	2.52E+01	2.63E+03	8.74E-06	2.56E-10	7.72E-02	2.99E-01	3.28E-08	7.48E+00
9*	4.84E+01	1.44E+03	4.16E-06	1.22E-10	4.22E-02	3.41E-01	2.27E-08	7.64E+00
Mean:								7.19E+00
Standard Deviation:								3.53E-01

## Citric Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.61E+04	1.29E-02	3.65E-08	4.57E-02	4.57E-02	2.04E-07	6.69E+00
2*	2.92E-01	4.82E+04	1.55E-02	4.37E-08	1.36E-01	1.82E-02	2.40E-06	5.62E+00
3*	1.04E+00	4.15E+04	3.71E-03	1.05E-08	1.18E-01	3.00E-01	7.69E-07	6.11E+00
4*	2.04E+00	1.71E+04	1.15E-03	3.24E-09	4.85E-02	3.48E-01	5.55E-07	6.26E+00
5*	3.00E+00	8.60E+03	6.00E-04	1.69E-09	2.44E-02	3.73E-01	4.29E-07	6.37E+00
6*	4.00E+00	1.93E+03	1.29E-04	3.66E-10	5.48E-03	3.78E-01	3.41E-07	6.47E+00
7*	5.08E+00	1.88E+03	1.16E-04	3.29E-10	5.32E-03	3.83E-01	2.69E-07	6.57E+00
8*	2.52E+01	3.69E+03	1.22E-05	3.46E-11	1.04E-02	3.94E-01	5.76E-08	7.24E+00
9*	4.84E+01	4.05E+03	1.17E-05	3.31E-11	1.15E-02	4.05E-01	3.19E-08	7.50E+00
Mean:								6.54E+00
Standard Deviation:								5.34E-01



Table F-6. CHELATING AGENT LEACHING DATA FOR PILGRIM SAMPLE (Continued)

## EDTA

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	1.90E+04	1.53E-02	3.56E-08	4.46E-02	4.46E-02	1.94E-07	6.71E+00
2*	2.92E-01	9.24E+04	2.97E-02	6.93E-08	2.17E-01	2.61E-01	2.00E-06	5.70E+00
3*	1.04E+00	1.36E+05	1.22E-02	2.85E-08	3.20E-01	5.81E-01	3.73E-06	5.43E+00
4*	2.04E+00	8.50E+04	5.68E-03	1.33E-08	1.99E-01	7.80E-01	4.56E-06	5.34E+00
5*	3.00E+00	3.80E+04	2.65E-04	6.20E-09	8.92E-02	8.70E-01	4.71E-06	5.33E+00
6*	4.00E+00	9.37E+03	6.27E-04	1.47E-09	2.20E-02	8.92E-01	3.91E-06	5.41E+00
7*	5.08E+00	1.43E+03	8.87E-05	2.08E-10	3.36E-03	8.95E-01	3.08E-06	5.51E+00
8*	2.52E+01	6.01E+03	2.00E-05	4.67E-11	1.41E-02	9.09E-01	6.95E-07	6.16E+00
9*	4.84E+01	5.75E+04	1.66E-04	3.89E-10	1.35E-01	1.04E+00	7.91E-07	6.10E+00
Mean:								5.74E+00
Standard Deviation:								4.52E-01

## DTPA

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	8.33E-02	3.72E+02	2.99E-04	7.05E-09	8.84E-03	8.84E-03	7.64E-09	8.12E+00
2*	2.92E-01	1.30E+03	4.17E-04	9.86E-09	3.09E-02	3.97E-02	1.23E-07	6.91E+00
3*	1.04E+00	1.13E+03	1.01E-04	2.39E-09	2.68E-02	6.66E-02	2.55E-08	7.59E+00
4*	2.04E+00	6.94E+02	4.64E-05	1.10E-09	1.65E-02	8.31E-02	1.32E-08	7.88E+00
5*	3.00E+00	9.56E+02	6.66E-05	1.58E-09	2.27E-02	1.06E-01	4.55E-08	7.34E+00
6*	4.00E+00	1.23E+03	8.26E-05	1.96E-09	2.93E-02	1.35E-01	9.75E-08	NA
7*	5.08E+00	1.51E+03	9.36E-05	2.22E-09	3.59E-02	1.71E-01	1.63E-07	6.79E+00
8*	2.52E+01	5.02E+03	1.67E-05	3.95E-10	1.19E-01	2.90E-01	2.31E-08	7.64E+00
9*	4.84E+01	1.90E+03	5.49E-06	1.30E-10	4.51E-02	3.35E-01	1.78E-08	7.75E+00
Mean:								7.50E+00
Standard Deviation:								4.32E-01

Table F-7. CHELATING AGENT LEACHING DATA FOR PEACH BOTTOM-2 SAMPLE

## Oxalic Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	2.46E+03	5.77E-04	8.58E-10	3.65E-03	3.65E-03	3.65E-10	9.44E+00
2*	9.88E-01	1.15E+03	1.13E-04	1.69E-10	1.71E-03	5.36E-03	1.14E-10	9.94E+00
3*	2.23E+00	1.82E+03	1.01E-04	1.50E-10	2.70E-03	8.06E-03	2.35E-10	9.63E+00
4*	3.06E+00	1.02E+03	8.38E-05	1.25E-10	1.51E-03	9.57E-03	2.78E-10	9.56E+00
5*	4.04E+00	1.01E+03	7.08E-05	1.05E-10	1.50E-03	1.11E-02	2.67E-10	9.57E+00
6*	7.02E+00	9.10E+02	2.09E-05	3.11E-11	1.35E-03	1.24E-02	3.57E-11	1.04E+01
7*	2.23E+01	1.98E+03	8.91E-06	1.32E-11	2.94E-03	1.54E-02	1.62E-11	1.08E+01
8*	5.61E+01	1.35E+03	2.73E-06	4.07E-12	2.01E-03	1.74E-02	4.20E-12	NA
9*	9.51E+01	7.21E+02	1.27E-06	1.88E-12	1.07E-03	1.84E-02	1.80E-12	1.17E+01
Mean:								1.01E+01
Standard Deviation:								7.54E-01

## Citric Acid

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	4.82E+03	1.13E-03	1.96E-09	8.32E-03	8.32E-03	1.90E-09	8.72E+00
2*	9.88E-01	1.19E+03	1.17E-04	2.03E-10	2.06E-03	1.04E-02	1.65E-10	9.78E+00
3*	2.23E+00	9.87E+02	5.46E-05	9.44E-11	1.70E-03	1.21E-02	9.37E-11	1.00E+01
4*	3.06E+00	1.57E+03	1.29E-04	2.22E-10	2.70E-03	1.48E-02	8.85E-10	9.05E+00
5*	4.04E+00	7.35E+02	5.14E-05	8.89E-11	1.27E-03	1.61E-02	1.90E-10	9.72E+00
6*	7.02E+00	7.47E+02	1.72E-05	2.97E-11	1.29E-03	1.73E-02	3.26E-11	1.05E+01
7*	2.23E+01	1.82E+03	8.16E-06	1.41E-11	3.13E-03	2.05E-02	1.84E-11	1.07E+01
8*	5.61E+01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.05E-02	0.00E+00	NA
9*	9.51E+01	6.76E+02	1.19E-06	2.05E-12	1.17E-03	2.16E-02	2.14E-12	1.17E+01
Mean:								1.00E+01
Standard Deviation:								8.82E-01

## EDTA

SAMPLE NUMBER	TOTAL TIME (days)	QUANTITY LEACHED (ug)	RELEASE RATE (ug/cm <sup>2</sup> /s)	RELEASE RATE (F/cm <sup>2</sup> /s)	INCREMENTAL RELEASE	CUMULATIVE RELEASE	EFFECTIVE DIFFUSIVITY (cm <sup>2</sup> /s)	LEACH INDEX
1*	2.92E-01	3.55E+03	8.33E-04	1.48E-09	6.31E-03	6.31E-03	1.09E-09	8.96E+00
2*	9.88E-01	5.06E+03	4.98E-04	8.88E-10	9.00E-03	1.53E-02	3.16E-09	8.50E+00
3*	2.23E+00	8.49E+03	4.69E-04	8.37E-10	1.51E-02	3.04E-02	7.37E-09	8.13E+00
4*	3.06E+00	3.21E+03	2.63E-04	4.69E-10	5.71E-03	3.61E-02	3.94E-09	8.40E+00
5*	4.04E+00	1.89E+03	1.32E-04	2.35E-10	3.36E-03	3.95E-02	1.33E-09	8.88E+00
6*	7.02E+00	7.06E+03	1.62E-04	2.89E-10	1.26E-02	5.20E-02	3.09E-09	8.51E+00
7*	2.23E+01	9.73E+03	4.37E-05	7.79E-11	1.73E-02	6.94E-02	5.61E-10	9.25E+00
8*	5.61E+01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.94E-02	0.00E+00	NA
9*	9.51E+01	7.03E+02	1.23E-06	2.20E-12	1.25E-03	7.06E-02	2.45E-12	1.16E+01
Mean:								9.03E+00
Standard Deviation:								1.03E+00



**APPENDIX G**

**RADIONUCLIDE AND STABLE METAL  
CONCENTRATIONS IN LEACHATES**



**APPENDIX G**

**RADIONUCLIDE AND STABLE METAL  
CONCENTRATIONS IN LEACHATES**

Radionuclide and stable metal concentrations in leachates are shown in Tables G-1 through G-7.

TABLE G-1. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: INDIAN POINT-3  
(uCi/ml decay corrected to 06/24/85)

UNFILTERED (540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54
1*	2.30E-05 +/- 2.01E-06	4.68E-06 +/- 6.79E-08	1.04E-04 +/- 6.66E-07	1.97E-04 +/- 9.06E-07	4.37E-07 +/- 2.92E-07
2*	4.51E-05 +/- 2.79E-06	8.60E-06 +/- 1.01E-07	2.07E-04 +/- 7.87E-07	4.10E-04 +/- 2.05E-06	NA
3*	1.09E-04 +/- 4.03E-06	1.96E-05 +/- 1.92E-07	3.74E-04 +/- 1.08E-06	7.37E-04 +/- 2.95E-06	3.89E-07 +/- 4.86E-07
4*	1.20E-04 +/- 4.75E-06	2.18E-05 +/- 2.88E-07	3.74E-04 +/- 2.21E-06	7.01E-04 +/- 5.47E-06	NA
5*	8.81E-05 +/- 4.57E-06	1.71E-05 +/- 1.81E-07	2.59E-04 +/- 1.63E-06	5.09E-04 +/- 3.61E-06	< 1.46E-07
6*	8.20E-05 +/- 3.48E-06	1.46E-05 +/- 1.85E-07	2.27E-04 +/- 5.67E-07	4.29E-04 +/- 1.20E-06	NA
7*	6.11E-05 +/- 3.05E-06	1.22E-05 +/- 8.78E-08	1.82E-04 +/- 5.46E-07	3.42E-04 +/- 8.21E-07	8.75E-08 +/- 2.43E-07
8*	4.18E-04 +/- 1.26E-05	8.35E-05 +/- 1.27E-06	1.14E-03 +/- 6.73E-06	2.24E-03 +/- 2.67E-05	NA
9*	6.49E-04 +/- 3.97E-05	1.22E-04 +/- 1.76E-06	1.40E-03 +/- 7.28E-06	2.71E-03 +/- 2.11E-05	1.65E-07 +/- 2.92E-08
10*	2.26E-04 +/- 2.70E-05	4.57E-05 +/- 6.26E-07	4.95E-04 +/- 4.36E-06	9.49E-04 +/- 1.40E-05	1.51E-06 +/- 9.72E-08

UNFILTERED (540 ml)

SAMPLE	Sb-125	Sr-90	Ni-63	Fe-55	Pu-241
1*	< 3.26E-08	2.10E-08 +/- 1.05E-09	1.32E-05 +/- 2.03E-06	6.71E-06 +/- 1.03E-06	< 9.94E-07
2*	NA	NA	NA	NA	NA
3*	< 1.47E-07	1.13E-07 +/- 5.24E-09	1.12E-05 +/- 2.03E-06	2.24E-06 +/- 3.44E-07	< 4.42E-07
4*	NA	NA	NA	NA	NA
5*	< 9.77E-08	9.64E-08 +/- 4.19E-09	8.11E-06 +/- 1.01E-06	3.10E-07 +/- 5.16E-08	< 5.52E-07
6*	NA	NA	NA	NA	NA
7*	< 9.77E-08	7.02E-08 +/- 3.14E-09	5.88E-06 +/- 9.13E-07	1.72E-07 +/- 1.72E-08	< 5.52E-07
8*	NA	NA	NA	NA	NA
9*	5.86E-07 +/- 3.26E-08	6.60E-07 +/- 3.14E-08	4.66E-05 +/- 7.10E-06	7.92E-07 +/- 1.20E-07	< 4.42E-07
10*	< 4.88E-08	2.62E-07 +/- 1.05E-08	1.72E-05 +/- 2.03E-06	8.78E-06 +/- 1.38E-06	< 3.31E-07

UNFILTERED (540 ml)

SAMPLE	Iron	Nickel	Cobalt	Chromium
1*	2.00E-02 +/- 1.00E-03	7.00E-02 +/- 3.50E-03	< 4.00E-02	< 8.00E-03
2*	NA	NA	NA	NA
3*	< 8.00E-03	1.00E-01 +/- 5.00E-03	< 4.00E-02	< 8.00E-03
4*	NA	NA	NA	NA
5*	< 8.00E-03	8.00E-02 +/- 4.00E-03	< 4.00E-02	< 8.00E-03
6*	NA	NA	NA	NA
7*	< 8.00E-03	7.00E-02 +/- 3.50E-03	< 4.00E-02	< 8.00E-03
8*	NA	NA	NA	NA
9*	< 8.00E-03	2.40E-01 +/- 1.20E-02	< 4.00E-02	< 8.00E-03
10*	< 8.00E-03	1.20E-01 +/- 6.00E-03	< 4.00E-02	< 8.00E-03

TABLE 6-1. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: INDIAN POINT-3  
(uCi/ml decay corrected to 06/24/85)

(Continued)

CATION MEMBRANE FILTRATE (60 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	3.92E-06 +/- 9.88E-08	7.06E-05 +/- 7.84E-07	1.41E-04 +/- 3.10E-06	ND	ND
2*	ND	7.27E-06 +/- 1.77E-07	1.59E-04 +/- 1.54E-06	3.32E-04 +/- 6.24E-06	ND	ND
3*	ND	1.81E-05 +/- 3.49E-07	3.01E-04 +/- 4.39E-07	5.94E-04 +/- 2.08E-05	ND	ND
4*	8.56E-05 +/- 7.55E-06	2.20E-05 +/- 4.71E-07	2.40E-04 +/- 3.77E-06	4.72E-04 +/- 7.84E-06	ND	ND
5*	8.91E-05 +/- 2.45E-05	1.60E-05 +/- 2.61E-07	2.18E-04 +/- 1.81E-06	4.42E-04 +/- 7.20E-06	ND	ND
6*	7.14E-05 +/- 9.45E-06	1.24E-05 +/- 3.05E-07	1.57E-04 +/- 1.15E-06	3.42E-04 +/- 2.36E-06	ND	ND
7*	6.10E-05 +/- 8.97E-06	1.08E-05 +/- 2.03E-07	1.36E-04 +/- 1.26E-06	2.81E-04 +/- 5.73E-06	ND	ND
8*	4.21E-04 +/- 2.19E-05	7.44E-05 +/- 1.19E-06	9.36E-04 +/- 6.65E-06	1.86E-03 +/- 1.41E-05	ND	ND
9*	4.42E-04 +/- 6.76E-05	1.23E-04 +/- 2.20E-06	7.59E-04 +/- 6.00E-06	2.30E-03 +/- 3.08E-05	ND	2.75E-04 +/- 2.39E-06
10*	ND	2.50E-05 +/- 4.50E-07	2.31E-04 +/- 2.89E-06	6.35E-04 +/- 1.14E-05	ND	4.79E-06 +/- 6.46E-07

CATION MEMBRANE FILTERS (60 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	2.04E-07 +/- 2.37E-08	5.99E-07 +/- 2.16E-08	1.22E-06 +/- 4.23E-08	ND	ND
2*	NA	NA	NA	NA	NA	NA
3*	ND	1.55E-07 +/- 3.56E-08	1.61E-06 +/- 1.66E-07	3.95E-06 +/- 6.58E-07	ND	ND
4*	ND	2.06E-06 +/- 1.10E-07	3.07E-05 +/- 6.82E-07	7.16E-05 +/- 2.42E-06	ND	ND
5*	ND	7.12E-08 +/- 2.48E-08	1.02E-06 +/- 1.52E-07	2.65E-06 +/- 3.95E-07	ND	ND
6*	NA	NA	NA	NA	NA	NA
7*	ND	1.55E-07 +/- 4.34E-08	8.83E-07 +/- 1.38E-07	1.97E-06 +/- 2.96E-07	ND	ND
8*	NA	NA	NA	NA	NA	NA
9*	ND	1.08E-05 +/- 1.70E-06	2.40E-04 +/- 3.59E-05	6.09E-04 +/- 9.87E-05	ND	ND
10*	ND	1.27E-05 +/- 2.83E-07	1.26E-04 +/- 1.93E-06	2.64E-04 +/- 3.65E-06	ND	ND

FRACTION OF NUCLIDE THAT IS COMPLEXED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	9.50E-01 +/- 5.58E-03	9.92E-01 +/- 3.15E-04	9.91E-01 +/- 3.49E-04	ND	ND
2*	NA	NA	NA	NA	NA	NA
3*	ND	9.92E-01 +/- 1.94E-03	9.95E-01 +/- 5.44E-04	9.93E-01 +/- 1.12E-03	ND	ND
4*	1.00E+00	9.14E-01 +/- 4.50E-03	8.86E-01 +/- 2.74E-03	8.68E-01 +/- 4.31E-03	ND	ND
5*	1.00E+00	9.96E-01 +/- 1.54E-03	9.95E-01 +/- 6.91E-04	9.94E-01 +/- 8.88E-04	ND	ND
6*	NA	NA	NA	NA	NA	NA
7*	1.00E+00	9.86E-01 +/- 3.91E-03	9.94E-01 +/- 1.00E-03	9.93E-01 +/- 1.05E-03	ND	ND
8*	NA	NA	NA	NA	NA	NA
9*	1.00E+00	9.19E-01 +/- 1.18E-02	7.60E-01 +/- 2.73E-02	7.91E-01 +/- 2.69E-02	ND	1.00E+00
10*	ND	6.63E-01 +/- 6.40E-03	6.47E-01 +/- 4.52E-03	7.06E-01 +/- 4.71E-03	ND	1.00E+00
Mean:	1.00E+00	9.17E-01	8.95E-01	9.05E-01	ND	1.00E+00



TABLE G-2. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: COOPER MIXED BED RESIN  
(uCi/ml decay corrected to 11/30/84)

FILTRATE (490 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	1.76E-05 +/- 1.57E-07	2.97E-07 +/- 4.90E-08	7.51E-07 +/- 3.13E-08	ND	1.74E-06 +/- 6.98E-08
2*	ND	4.32E-06 +/- 6.74E-08	5.41E-07 +/- 3.05E-08	3.92E-06 +/- 5.64E-08	ND	2.13E-06 +/- 8.01E-08
3*	ND	4.25E-06 +/- 6.12E-08	3.24E-07 +/- 2.61E-08	2.02E-06 +/- 4.24E-08	3.86E-07 +/- 7.37E-08	1.45E-06 +/- 6.02E-08
4*	ND	2.45E-06 +/- 4.80E-08	2.81E-07 +/- 2.95E-08	4.18E-07 +/- 2.45E-08	ND	1.59E-06 +/- 6.06E-08
5*	ND	2.82E-06 +/- 5.41E-08	2.06E-07 +/- 2.53E-08	1.37E-06 +/- 3.63E-08	ND	1.21E-06 +/- 5.45E-08
6*	ND	1.61E-06 +/- 4.02E-08	1.46E-07 +/- 1.78E-08	1.01E-06 +/- 3.21E-08	ND	9.66E-07 +/- 4.64E-08
7*	ND	1.30E-06 +/- 3.51E-08	1.30E-07 +/- 2.43E-08	2.62E-06 +/- 4.72E-08	ND	1.36E-06 +/- 6.57E-08
8*	ND	1.30E-06 +/- 3.94E-08	1.66E-07 +/- 1.88E-08	3.21E-07 +/- 2.85E-08	ND	7.64E-07 +/- 5.26E-08
9*	ND	6.48E-07 +/- 5.82E-08	ND	6.60E-07 +/- 4.89E-08	ND	9.04E-07 +/- 9.85E-08
10*	ND	5.72E-07 +/- 2.20E-08	1.56E-07 +/- 1.47E-08	2.57E-07 +/- 1.55E-08	ND	4.87E-07 +/- 3.74E-08

FILTERS (540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	8.44E-05 +/- 3.36E-06	ND	ND	1.41E-05 +/- 9.38E-07	ND
2*	ND	1.82E-04 +/- 4.13E-06	ND	ND	3.38E-05 +/- 1.70E-06	1.20E-06 +/- 1.98E-07
3*	ND	5.16E-05 +/- 1.09E-06	ND	ND	9.18E-06 +/- 4.88E-07	4.26E-07 +/- 1.67E-07
4*	ND	9.98E-05 +/- 2.98E-06	ND	ND	1.73E-05 +/- 4.78E-07	6.28E-07 +/- 1.02E-07
5*	ND	1.13E-04 +/- 1.67E-05	ND	5.19E-08 +/- 1.67E-08	1.93E-05 +/- 2.96E-06	8.70E-07 +/- 1.30E-07
6*	NA	NA	NA	NA	NA	NA
7*	ND	7.48E-05 +/- 2.49E-06	ND	9.63E-09 +/- 2.78E-09	1.29E-05 +/- 9.06E-07	5.66E-07 +/- 1.32E-07
8*	ND	2.99E-05 +/- 8.98E-07	ND	ND	4.66E-06 +/- 3.70E-07	ND
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

FILTER + FILTRATE (490/540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	1.02E-04 +/- 3.36E-06	2.97E-07 +/- 4.90E-08	7.51E-07 +/- 3.13E-08	1.41E-05 +/- 9.38E-07	1.74E-06 +/- 6.98E-08
2*	ND	1.86E-04 +/- 4.13E-06	5.41E-07 +/- 3.05E-08	3.92E-06 +/- 5.64E-08	3.38E-05 +/- 1.70E-06	3.33E-06 +/- 2.14E-07
3*	ND	5.58E-05 +/- 1.09E-06	3.24E-07 +/- 2.61E-08	2.02E-06 +/- 4.24E-08	9.57E-06 +/- 4.93E-07	1.88E-06 +/- 1.78E-07
4*	ND	1.02E-04 +/- 2.98E-06	2.81E-07 +/- 2.95E-08	4.18E-07 +/- 2.45E-08	1.73E-05 +/- 4.78E-07	2.22E-06 +/- 1.19E-07
5*	ND	1.16E-04 +/- 1.67E-05	2.06E-07 +/- 2.53E-08	1.37E-06 +/- 3.63E-08	1.93E-05 +/- 2.96E-06	2.08E-06 +/- 1.41E-07
6*	NA	NA	1.46E-07 +/- 1.78E-08	1.01E-06 +/- 3.21E-08	NA	NA
7*	ND	7.61E-05 +/- 2.49E-06	1.30E-07 +/- 2.43E-08	2.62E-06 +/- 4.72E-08	1.29E-05 +/- 9.06E-07	1.93E-06 +/- 1.47E-07
8*	ND	3.12E-05 +/- 8.99E-07	1.66E-07 +/- 1.88E-08	3.21E-07 +/- 2.85E-08	4.66E-06 +/- 3.70E-07	7.64E-07 +/- 5.26E-08
9*	NA	NA	ND	6.60E-07 +/- 4.89E-08	NA	NA
10*	NA	NA	1.56E-07 +/- 1.47E-08	2.57E-07 +/- 1.55E-08	NA	NA

TABLE 6-2. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: COOPER MIXED BED RESIN (Continued)  
(uCi/ml decay corrected to 11/30/84)

UNFILTERED

SAMPLE	Sr-90	Ni-63	Fe-55	Pu-241
1*	1.06E-09 +/- 8.50E-10	2.25E-07 +/- 3.05E-08	1.10E-05 +/- 1.59E-06	< 5.67E-07
2*	3.40E-08 +/- 2.12E-09	6.41E-07 +/- 9.16E-08	3.19E-05 +/- 3.98E-06	< 4.54E-07
3*	2.44E-09 +/- 8.50E-10	1.63E-07 +/- 2.04E-08	1.20E-05 +/- 1.99E-06	< 5.67E-07
4*	2.23E-08 +/- 2.12E-09	1.63E-07 +/- 2.04E-08	1.99E-05 +/- 3.98E-06	< 2.27E-07
5*	NA	NA	NA	NA
6*	NA	NA	NA	NA
7*	NA	NA	NA	NA
8*	NA	NA	NA	NA
9*	6.80E-09 +/- 8.50E-10	2.14E-07 +/- 3.05E-08	1.79E-05 +/- 1.99E-06	< 2.27E-07
10*	NA	NA	NA	NA

UNFILTERED

SAMPLE	Iron	Nickel	Cobalt	Chromium
1*	< 8.00E-03	4.30E-02 +/- 2.00E-03	< 4.00E-02	9.00E-03 +/- 5.00E-04
2*	< 8.00E-03	< 4.00E-02	< 4.00E-02	< 8.00E-03
3*	< 8.00E-03	< 4.00E-02	< 4.00E-02	< 8.00E-03
4*	< 8.00E-03	4.50E-02 +/- 2.00E-03	< 4.00E-02	< 8.00E-03
5*	NA	NA	NA	NA
6*	NA	NA	NA	NA
7*	NA	NA	NA	NA
8*	NA	NA	NA	NA
9*	< 8.00E-03	< 4.00E-02	< 4.00E-02	< 8.00E-03
10*	NA	NA	NA	NA

TABLE 6-2. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: COOPER MIXED BED RESIN (Continued)  
(uCi/ml decay corrected to 11/30/84)

CATION MEMBRANE FILTRATE (50 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	3.36E-05 +/- 5.11E-07	3.92E-07 +/- 9.09E-08	6.94E-07 +/- 8.33E-08	ND	< 2.20E-06
2*	ND	3.77E-06 +/- 1.84E-07	3.34E-07 +/- 4.64E-08	2.80E-06 +/- 1.44E-07	ND	1.57E-06 +/- 1.79E-07
3*	ND	2.09E-06 +/- 1.15E-07	3.18E-07 +/- 6.87E-08	4.30E-07 +/- 4.64E-08	ND	1.48E-06 +/- 1.42E-07
4*	ND	1.38E-06 +/- 7.99E-08	2.68E-07 +/- 6.97E-08	1.47E-06 +/- 1.12E-07	ND	1.05E-06 +/- 1.18E-07
5*	ND	1.05E-06 +/- 6.76E-08	< 3.32E-07	7.80E-07 +/- 5.50E-08	ND	9.38E-07 +/- 2.00E-07
6*	ND	9.67E-07 +/- 6.49E-08	ND	6.34E-07 +/- 5.10E-08	ND	7.52E-07 +/- 1.43E-07
7*	ND	1.08E-06 +/- 7.00E-08	ND	1.62E-07 +/- 8.73E-09	ND	9.11E-07 +/- 1.22E-07
8*	ND	1.50E-06 +/- 8.76E-08	ND	3.56E-07 +/- 6.55E-08	ND	1.65E-06 +/- 2.85E-07
9*	ND	1.33E-06 +/- 6.97E-08	1.99E-06 +/- 1.04E-07	4.67E-06 +/- 1.49E-07	ND	ND
10*	ND	1.02E-06 +/- 7.28E-08	1.03E-06 +/- 6.93E-08	2.41E-06 +/- 8.87E-08	ND	1.02E-06 +/- 1.48E-07

CATION MEMBRANE FILTERS (50 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	1.14E-05 +/- 4.80E-07	ND	2.86E-07 +/- 5.45E-08	ND	ND
2*	ND	8.01E-07 +/- 2.60E-08	ND	4.32E-07 +/- 2.30E-08	ND	ND
3*	ND	7.34E-07 +/- 4.97E-08	ND	3.29E-07 +/- 4.24E-08	ND	ND
4*	ND	6.33E-07 +/- 4.10E-08	ND	3.65E-07 +/- 2.84E-08	ND	ND
5*	ND	5.60E-07 +/- 4.69E-08	ND	3.41E-07 +/- 4.43E-08	ND	ND
6*	ND	5.03E-07 +/- 2.38E-08	ND	3.44E-07 +/- 1.53E-08	ND	ND
7*	ND	5.68E-07 +/- 4.88E-08	ND	3.48E-07 +/- 3.55E-08	ND	ND
8*	ND	5.10E-07 +/- 3.19E-08	ND	1.44E-07 +/- 5.35E-08	ND	ND
9*	ND	1.22E-07 +/- 3.50E-08	1.77E-07 +/- 3.50E-08	3.33E-07 +/- 5.00E-08	ND	ND
10*	NA	NA	NA	NA	NA	NA

FRACTION OF NUCLIDE THAT IS COMPLEXED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	7.47E-01 +/- 8.49E-03	ND	7.08E-01 +/- 4.66E-02	ND	ND
2*	ND	8.25E-01 +/- 8.47E-03	ND	8.66E-01 +/- 8.57E-03	ND	1.00E+00
3*	ND	7.40E-01 +/- 1.68E-02	ND	5.67E-01 +/- 4.13E-02	ND	1.00E+00
4*	ND	6.85E-01 +/- 1.87E-02	ND	8.01E-01 +/- 1.74E-02	ND	1.00E+00
5*	ND	6.52E-01 +/- 2.40E-02	ND	6.96E-01 +/- 3.13E-02	ND	1.00E+00
6*	ND	6.58E-01 +/- 1.85E-02	ND	6.48E-01 +/- 2.09E-02	ND	1.00E+00
7*	ND	6.55E-01 +/- 2.43E-02	ND	3.17E-01 +/- 2.50E-02	ND	1.00E+00
8*	ND	7.46E-01 +/- 1.62E-02	ND	7.12E-01 +/- 8.50E-02	ND	1.00E+00
9*	ND	9.16E-01 +/- 2.25E-02	9.18E-01 +/- 1.53E-02	9.33E-01 +/- 9.53E-03	ND	ND
10*	NA	NA	NA	NA	NA	NA
Mean:	ND	7.36E-01	9.18E-01	6.94E-01	ND	1.00E+00

TABLE G-3. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: COOPER CATION RESIN  
(uCi/ml decay corrected to 11/30/84)

FILTRATE (540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	ND	1.03E-06 +/- 4.27E-08	ND	1.33E-07 +/- 2.03E-08	ND	ND
2*	ND	1.12E-04 +/- 1.76E-06	5.82E-05 +/- 2.09E-05	2.68E-07 +/- 4.98E-08	1.60E-05 +/- 6.90E-07	ND
3*	ND	4.45E-06 +/- 6.63E-08	9.63E-08 +/- 2.72E-08	4.86E-07 +/- 3.61E-08	ND	ND
4*	ND	7.67E-07 +/- 4.22E-08	ND	6.75E-08 +/- 1.43E-08	ND	ND
5*	ND	2.08E-06 +/- 4.85E-08	6.04E-08 +/- 8.21E-09	1.27E-07 +/- 1.25E-08	ND	1.03E-05 +/- 3.40E-06
6*	ND	2.52E-06 +/- 4.08E-08	8.48E-08 +/- 1.37E-08	8.02E-08 +/- 1.20E-08	ND	ND
7*	ND	1.01E-06 +/- 2.09E-08	ND	3.26E-08 +/- 1.28E-08	1.93E-07 +/- 4.46E-08	3.86E-07 +/- 1.41E-07
8*	ND	4.80E-06 +/- 1.41E-07	5.25E-07 +/- 5.41E-08	7.79E-07 +/- 5.37E-08	ND	ND
9*	ND	4.17E-06 +/- 5.05E-08	2.87E-06 +/- 5.14E-08	4.44E-06 +/- 5.15E-08	ND	ND
10*	ND	6.49E-06 +/- 1.34E-07	1.03E-06 +/- 7.87E-08	1.75E-06 +/- 7.56E-08	ND	ND

FILTERS (540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	NA	NA	NA	NA	NA	NA
2*	ND	2.51E-05 +/- 9.85E-07	ND	ND	5.00E-06 +/- 3.45E-07	ND
3*	ND	1.65E-05 +/- 5.13E-07	ND	ND	3.63E-06 +/- 2.82E-07	ND
4*	NA	NA	NA	NA	NA	NA
5*	NA	NA	NA	NA	NA	NA
6*	ND	1.13E-04 +/- 1.59E-06	ND	ND	1.25E-05 +/- 7.48E-07	ND
7*	ND	3.20E-05 +/- 4.67E-06	ND	ND	5.47E-06 +/- 8.00E-07	ND
8*	ND	9.88E-05 +/- 1.76E-06	ND	ND	1.01E-05 +/- 4.13E-07	ND
9*	NA	NA	NA	NA	NA	NA
10*	ND	9.51E-05 +/- 1.60E-06	ND	ND	ND	ND

FILTRATE + FILTER (540/1500 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	NA	NA	ND	1.33E-07 +/- 2.03E-08	NA	NA
2*	ND	1.37E-04 +/- 2.02E-06	5.82E-05 +/- 2.09E-05	2.68E-07 +/- 4.98E-08	2.10E-05 +/- 7.71E-07	ND
3*	ND	2.10E-05 +/- 5.17E-07	9.63E-08 +/- 2.72E-08	4.86E-07 +/- 3.61E-08	3.63E-06 +/- 2.82E-07	ND
4*	NA	NA	ND	6.75E-08 +/- 1.43E-08	NA	NA
5*	NA	NA	6.04E-08 +/- 8.21E-09	1.27E-07 +/- 1.25E-08	NA	NA
6*	ND	1.16E-04 +/- 1.59E-06	8.48E-08 +/- 1.37E-08	8.02E-08 +/- 1.20E-08	1.25E-05 +/- 7.48E-07	ND
7*	ND	3.30E-05 +/- 4.67E-06	ND	3.26E-08 +/- 1.28E-08	5.66E-06 +/- 8.01E-07	3.86E-07 +/- 1.41E-07
8*	ND	1.04E-04 +/- 1.76E-06	5.25E-07 +/- 5.41E-08	7.79E-07 +/- 5.37E-08	1.01E-05 +/- 4.13E-07	ND
9*	NA	NA	2.87E-06 +/- 5.14E-08	4.44E-06 +/- 5.15E-08	NA	NA
10*	ND	1.02E-04 +/- 1.60E-06	1.03E-06 +/- 7.87E-08	1.75E-06 +/- 7.56E-08	ND	ND

TABLE 6-3. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: COOPER CATION RESIN (Continued)  
(uCi/ml decay corrected to 11/30/84)

FILTRATE + FILTER

SAMPLE	Sr-90	Ni-63	Fe-55	Pu-241
1*	3.82E-06 +/- 9.56E-07	7.13E-04 +/- 1.02E-04	1.99E-02 +/- 3.98E-03	1.13E-05 +/- 2.27E-06
2*	7.75E-09 +/- 8.50E-10	1.22E-06 +/- 2.04E-07	2.79E-05 +/- 3.98E-06	< 2.27E-07
3*	1.06E-08 +/- 1.06E-09	2.24E-06 +/- 3.05E-07	1.10E-04 +/- 1.59E-05	< 1.13E-07
4*	NA	NA	NA	NA
5*	NA	NA	NA	NA
6*	3.08E-08 +/- 2.12E-09	2.55E-06 +/- 4.07E-07	1.59E-04 +/- 1.99E-05	< 1.13E-07
7*	NA	NA	NA	NA
8*	NA	NA	NA	NA
9*	9.56E-09 +/- 1.06E-09	1.02E-06 +/- 2.04E-07	5.38E-05 +/- 7.97E-06	< 1.13E-07
10*				

FILTRATE + FILTER

SAMPLE	Sr-90	Ni-63	Fe-55	Pu-241
1*	3.82E-06 +/- 9.56E-07	7.13E-04 +/- 1.02E-04	1.99E-02 +/- 3.98E-03	1.13E-05 +/- 2.27E-06
2*	7.75E-09 +/- 8.50E-10	1.22E-06 +/- 2.04E-07	2.79E-05 +/- 3.98E-06	< 2.27E-07
3*	1.06E-08 +/- 1.06E-09	2.24E-06 +/- 3.05E-07	1.10E-04 +/- 1.59E-05	< 1.13E-07
4*	NA	NA	NA	NA
5*	NA	NA	NA	NA
6*	3.08E-08 +/- 2.12E-09	2.55E-06 +/- 4.07E-07	1.59E-04 +/- 1.99E-05	< 1.13E-07
7*	NA	NA	NA	NA
8*	NA	NA	NA	NA
9*	9.56E-09 +/- 1.06E-09	1.02E-06 +/- 2.04E-07	5.38E-05 +/- 7.97E-06	< 1.13E-07
10*				

FILTRATE + FILTER

SAMPLE	Iron	Nickel	Cobalt	Chromium
1*	9.13E+01 +/- 4.57E+00	5.40E+00 +/- 2.70E-01	1.60E-01 +/- 8.00E-03	1.05E+00 +/- 5.25E-02
2*	< 8.00E-03	6.00E-02 +/- 3.00E-03	< 4.00E-02	< 8.00E-03
3*	3.70E-01 +/- 1.85E-02	6.00E-02 +/- 3.00E-03	< 4.00E-02	< 8.00E-03
4*	NA	NA	NA	NA
5*	NA	NA	NA	NA
6*	8.40E-01 +/- 4.20E-02	6.00E-02 +/- 3.00E-03	< 4.00E-02	< 8.00E-03
7*	NA	NA	NA	NA
8*	NA	NA	NA	NA
9*	2.90E-01 +/- 1.45E-02	5.00E-02 +/- 2.50E-03	< 4.00E-02	3.00E-02 +/- 1.50E-03
10*				

TABLE 6-3. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: COOPER CATION RESIN (Continued)  
(uCi/ml decay corrected to 11/30/84)

CATION MEMBRANE FILTRATE

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	1.15E-06 +/- 7.79E-08	1.31E-06 +/- 7.51E-08	3.34E-06 +/- 1.04E-07	ND	ND
2*	ND	7.40E-06 +/- 1.60E-07	1.18E-06 +/- 7.46E-08	3.22E-06 +/- 1.04E-07	ND	ND
3*	ND	3.32E-06 +/- 1.69E-07	9.81E-07 +/- 1.00E-07	3.10E-06 +/- 1.00E-07	ND	ND
4*	ND	1.77E-06 +/- 8.11E-08	5.94E-07 +/- 7.13E-08	1.62E-06 +/- 7.45E-08	ND	ND
5*	ND	8.81E-07 +/- 5.93E-08	7.91E-07 +/- 6.21E-08	2.11E-06 +/- 1.03E-07	ND	ND
6*	ND	1.86E-06 +/- 8.35E-08	ND	1.05E-06 +/- 6.37E-08	ND	ND
7*	ND	8.15E-07 +/- 6.07E-08	4.74E-07 +/- 5.59E-08	1.03E-06 +/- 8.85E-08	ND	ND
8*	ND	3.55E-06 +/- 1.47E-07	7.31E-07 +/- 6.70E-08	1.40E-06 +/- 9.46E-08	ND	ND
9*	ND	3.64E-06 +/- 1.12E-07	2.80E-06 +/- 1.20E-07	5.17E-06 +/- 1.28E-07	ND	ND
10*	NA	NA	NA	NA	NA	NA

CATION MEMBRANE FILTER

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	4.67E-07 +/- 1.00E-07	ND	2.67E-07 +/- 5.00E-08	ND	ND
2*	ND	4.19E-05 +/- 7.80E-07	ND	1.57E-07 +/- 1.97E-08	7.10E-06 +/- 3.10E-07	ND
3*	ND	6.83E-07 +/- 6.67E-08	1.77E-07 +/- 4.33E-08	3.67E-06 +/- 4.67E-06	ND	ND
4*	NA	NA	NA	NA	NA	NA
5*	ND	3.17E-07 +/- 6.67E-08	ND	1.25E-07 +/- 4.67E-08	ND	ND
6*	NA	NA	NA	NA	NA	NA
7*	ND	ND	ND	1.18E-07 +/- 3.33E-08	ND	ND
8*	NA	1.86E-07 +/- 1.76E-08	ND	1.35E-07 +/- 1.33E-08	ND	ND
9*	ND	3.67E-07 +/- 6.67E-08	2.27E-07 +/- 4.17E-08	4.33E-07 +/- 6.67E-08	ND	ND
10*	NA	1.48E-07 +/- 1.72E-08	ND	1.13E-07 +/- 1.55E-08	ND	ND

FRACTION OF NUCLIDE THAT IS COMPLEXED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	7.11E-01 +/- 4.61E-02	1.00E+00	9.26E-01 +/- 1.30E-02	ND	ND
2*	ND	1.50E-01 +/- 3.64E-03	1.00E+00	9.54E-01 +/- 5.74E-03	0.00E+00	ND
3*	ND	8.29E-01 +/- 1.56E-02	8.47E-01 +/- 3.44E-02	4.58E-01 +/- 3.16E-01	ND	ND
4*	NA	NA	NA	NA	NA	NA
5*	ND	7.36E-01 +/- 4.30E-02	1.00E+00	9.44E-01 +/- 1.99E-02	ND	ND
6*	NA	NA	NA	NA	NA	NA
7*	ND	1.00E+00	1.00E+00	8.97E-01 +/- 2.72E-02	ND	ND
8*	ND	9.50E-01 +/- 4.89E-03	1.00E+00	9.12E-01 +/- 9.58E-03	ND	ND
9*	ND	9.08E-01 +/- 1.53E-02	9.25E-01 +/- 1.31E-02	9.23E-01 +/- 1.11E-02	ND	ND
10*	NA	NA	NA	NA	NA	NA
Mean:	ND	7.55E-01	9.68E-01	8.59E-01	0.00E+00	ND

TABLE 6-4. RADIONUCLIDE CONCENTRATIONS IN LEACHATES; MILLSTONE-1 F33 SAMPLE  
(uCi/ml decay corrected to 06/20/84)

UNFILTERED (450/540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	< 7.00E-05	8.77E-06 +/- 3.58E-07	2.23E-06 +/- 1.38E-07	1.10E-05 +/- 2.85E-07	1.10E-06 +/- 2.96E-07	1.23E-06 +/- 3.17E-07
2*	ND	1.67E-05 +/- 2.99E-07	2.60E-06 +/- 1.91E-07	1.32E-05 +/- 2.81E-07	2.93E-06 +/- 1.22E-06	ND
3*	ND	1.62E-05 +/- 3.10E-07	4.44E-06 +/- 2.57E-07	2.25E-05 +/- 3.78E-07	ND	2.81E-06 +/- 4.33E-07
4*	ND	2.49E-05 +/- 9.71E-08	4.01E-06 +/- 3.61E-08	1.99E-05 +/- 1.28E-07	2.89E-06 +/- 2.03E-07	3.11E-06 +/- 8.12E-08
5*	ND	2.55E-05 +/- 1.15E-07	2.56E-06 +/- 3.79E-08	1.31E-05 +/- 5.00E-08	3.38E-06 +/- 2.12E-07	2.92E-06 +/- 5.37E-08
6*	< 1.01E-04	1.10E-05 +/- 2.80E-07	1.67E-06 +/- 9.85E-08	8.09E-06 +/- 2.60E-07	< 5.67E-07	2.81E-06 +/- 2.43E-07
7*	1.91E-04 +/- 8.44E-05	1.20E-05 +/- 9.97E-07	1.25E-06 +/- 2.31E-07	6.13E-06 +/- 3.93E-07	< 1.41E-06	2.25E-06 +/- 5.80E-07
8*	ND	3.13E-05 +/- 3.51E-07	3.66E-06 +/- 5.61E-08	1.84E-05 +/- 1.49E-07	2.97E-06 +/- 4.20E-07	1.68E-06 +/- 7.45E-08
9*	ND	2.20E-05 +/- 2.02E-07	1.19E-06 +/- 3.48E-08	5.76E-06 +/- 6.34E-08	ND	1.49E-06 +/- 4.36E-08
10*	ND	2.75E-05 +/- 4.01E-07	ND	1.69E-06 +/- 1.01E-07	ND	ND

TABLE 6-5. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: MILLSTONE-1 F201 SAMPLE  
(uCi/ml decay corrected to 06/20/84)

UNFILTERED (490/540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54
1*	< 1.08E-04	8.66E-06 +/- 2.81E-07	1.85E-06 +/- 1.32E-07	9.70E-06 +/- 2.68E-07	1.25E-06 +/- 3.26E-07
2*	< 6.19E-05	9.86E-06 +/- 2.64E-07	2.55E-06 +/- 1.04E-07	1.32E-05 +/- 2.59E-07	1.07E-07 +/- 1.94E-08
3*	< 7.68E-05	1.43E-05 +/- 2.70E-07	4.12E-06 +/- 1.36E-07	2.10E-05 +/- 3.89E-07	< 5.06E-07
4*	< 1.39E-04	1.74E-05 +/- 5.36E-07	3.17E-06 +/- 2.27E-07	1.69E-05 +/- 4.17E-07	1.17E-06 +/- 4.22E-07
5*	< 2.11E-04	1.64E-05 +/- 6.81E-07	2.01E-06 +/- 2.65E-07	1.08E-05 +/- 5.15E-07	1.37E-06 +/- 6.52E-07
6*	< 2.68E-04	1.06E-05 +/- 7.67E-07	1.64E-06 +/- 2.56E-07	6.85E-06 +/- 5.14E-07	< 1.89E-06
7*	< 1.21E-04	1.16E-05 +/- 2.39E-07	1.53E-06 +/- 1.06E-07	6.62E-06 +/- 1.82E-07	< 7.81E-07
8*	ND	3.74E-04 +/- 8.76E-06	9.41E-06 +/- 1.62E-07	1.79E-05 +/- 4.27E-07	ND
9*	ND	4.02E-05 +/- 7.65E-07	1.60E-06 +/- 7.45E-08	8.02E-06 +/- 1.50E-07	1.53E-06 +/- 2.98E-07
10*	ND	1.21E-04 +/- 2.69E-06	ND	2.52E-06 +/- 1.11E-07	2.99E-05 +/- 1.99E-06

UNFILTERED (490/540 ml)

SAMPLE	Sb-125	Sr-90	Ni-63	Fe-55	Pu-241
1*	1.11E-06 +/- 3.14E-07	2.15E-07 +/- 1.07E-08	9.19E-06 +/- 1.02E-06	1.27E-05 +/- 1.79E-06	< 4.64E-07
2*	1.48E-06 +/- 1.79E-07	NA	NA	NA	NA
3*	3.20E-06 +/- 3.15E-07	7.19E-07 +/- 3.22E-08	3.57E-05 +/- 5.11E-06	1.79E-05 +/- 2.23E-06	< 2.32E-07
4*	5.03E-06 +/- 5.73E-07	NA	NA	NA	NA
5*	4.11E-06 +/- 7.60E-07	6.65E-07 +/- 3.22E-08	3.68E-05 +/- 5.11E-06	2.24E-05 +/- 2.23E-06	< 9.27E-07
6*	4.02E-06 +/- 7.40E-07	6.12E-07 +/- 3.22E-08	2.55E-05 +/- 4.08E-06	1.56E-05 +/- 2.23E-06	< 5.80E-07
7*	3.04E-06 +/- 2.49E-07	NA	NA	NA	NA
8*	2.06E-06 +/- 2.54E-07	2.79E-06 +/- 1.07E-07	1.12E-04 +/- 2.04E-06	4.02E-06 +/- 6.70E-07	< 3.48E-07
9*	1.62E-06 +/- 9.90E-08	NA	NA	NA	NA
10*	3.75E-06 +/- 3.03E-07	2.68E-06 +/- 1.07E-07	5.72E-05 +/- 8.17E-06	3.80E-06 +/- 6.70E-07	< 1.16E-07

UNFILTERED 490/540 ML

SAMPLE	Iron	Nickel	Cobalt	Chromium
1*	< 8.00E-03	6.00E-02 +/- 3.00E-03	< 4.00E-02	< 8.00E-03
2*	NA	NA	NA	NA
3*	< 8.00E-03	1.30E-01 +/- 6.50E-03	< 4.00E-02	< 8.00E-03
4*	NA	NA	NA	NA
5*	< 8.00E-03	1.20E-01 +/- 6.00E-03	< 4.00E-02	< 8.00E-03
6*	< 8.00E-03	1.10E-01 +/- 5.50E-03	< 4.00E-02	< 8.00E-03
7*	NA	NA	NA	NA
8*	< 8.00E-03	3.30E-01 +/- 1.65E-02	< 4.00E-02	< 8.00E-03
9*	NA	NA	NA	NA
10*	< 8.00E-03	1.80E-01 +/- 9.00E-03	< 4.00E-02	< 8.00E-03



TABLE 6-5. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: MILLSTONE-1 F201 SAMPLE (Continued)  
(uCi/ml decay corrected to 06/20/84)

FILTRATE (490/540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 1.66E-04	8.47E-06 +/- 5.79E-07	1.21E-06 +/- 4.39E-07	8.97E-06 +/- 4.91E-07	1.70E-06 +/- 7.45E-07	< 1.61E-06
2*	< 4.40E-04	9.73E-06 +/- 7.25E-07	2.03E-06 +/- 2.72E-07	1.37E-05 +/- 6.23E-07	< 1.20E-06	1.42E-06 +/- 6.39E-07
3*	< 3.02E-04	1.32E-05 +/- 6.15E-07	4.64E-06 +/- 3.75E-07	2.17E-05 +/- 7.44E-07	< 1.68E-06	3.17E-06 +/- 8.24E-07
4*	< 8.16E-05	1.67E-05 +/- 2.54E-07	3.50E-06 +/- 1.28E-07	1.78E-05 +/- 4.68E-07	1.15E-06 +/- 3.35E-07	4.02E-06 +/- 2.47E-07
5*	< 1.70E-04	1.49E-05 +/- 3.68E-07	2.28E-06 +/- 1.47E-07	1.18E-05 +/- 3.74E-07	1.36E-06 +/- 3.20E-07	3.42E-06 +/- 3.45E-07
6*	NA	NA	NA	NA	NA	NA
7*	NA	NA	NA	NA	NA	NA
8*	ND	2.20E-05 +/- 5.28E-07	3.56E-06 +/- 2.67E-07	2.05E-05 +/- 6.03E-07	< 1.51E-06	1.63E-06 +/- 5.04E-07
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

FILTERS (490/540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 7.51E-05	3.28E-06 +/- 1.70E-07	< 6.39E-08	< 6.44E-08	1.59E-06 +/- 2.19E-07	< 1.71E-07
2*	< 1.55E-06	1.24E-06 +/- 7.89E-08	< 5.86E-08	< 6.29E-08	1.85E-07 +/- 4.96E-08	< 9.58E-08
3*	< 4.50E-05	3.79E-07 +/- 5.08E-08	< 3.62E-08	< 6.44E-08	2.76E-07 +/- 1.28E-07	< 7.44E-08
4*	NA	NA	NA	NA	NA	NA
5*	NA	NA	NA	NA	NA	NA
6*	< 4.89E-05	1.51E-05 +/- 8.85E-07	< 4.46E-08	< 6.90E-08	5.36E-07 +/- 1.61E-07	< 1.20E-07
7*	< 3.45E-05	1.91E-06 +/- 9.99E-08	< 7.81E-08	< 4.72E-08	8.21E-07 +/- 2.27E-07	< 9.30E-08
8*	NA	NA	NA	NA	NA	NA
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

FILTRATE + FILTER (490/540 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 2.41E-04	1.18E-05 +/- 6.03E-07	< 1.27E-06	< 9.03E-06	3.29E-06 +/- 7.76E-07	< 1.78E-06
2*	< 4.42E-04	1.10E-05 +/- 7.29E-07	< 2.09E-06	< 1.38E-05	1.38E-06 +/- 4.96E-08	< 1.52E-06
3*	< 3.47E-04	1.36E-05 +/- 6.17E-07	< 4.68E-06	< 2.18E-05	1.96E-06 +/- 1.28E-07	< 3.24E-06
4*	NA	NA	NA	NA	NA	NA
5*	NA	NA	NA	NA	NA	NA
6*	NA	NA	NA	NA	NA	NA
7*	NA	NA	NA	NA	NA	NA
8*	NA	NA	NA	NA	NA	NA
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

TABLE 6-5. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: MILLSTONE-1 F201 SAMPLE (Continued)  
(uCi/ml decay corrected to 06/20/84)

CATION MEMBRANE FILTRATE (50 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 3.00E-04	1.05E-05 +/- 6.12E-07	1.60E-06 +/- 3.68E-07	5.70E-06 +/- 4.53E-07	< 1.48E-06	2.58E-06 +/- 8.90E-07
2*	ND	6.37E-06 +/- 4.96E-07	1.88E-06 +/- 4.08E-07	1.08E-05 +/- 6.56E-07	< 2.87E-06	< 3.18E-06
3*	< 3.07E-04	1.19E-05 +/- 1.02E-06	2.37E-06 +/- 4.22E-07	1.85E-05 +/- 7.58E-07	< 2.43E-06	2.73E-06 +/- 9.23E-07
4*	NA	NA	NA	NA	NA	NA
5*	< 2.78E-04	1.24E-05 +/- 6.56E-07	2.45E-06 +/- 3.48E-07	1.09E-05 +/- 6.09E-07	< 1.98E-06	2.86E-06 +/- 9.32E-07
6*	< 4.49E-04	9.77E-06 +/- 5.74E-07	1.18E-06 +/- 3.06E-07	7.41E-06 +/- 4.81E-07	1.86E-06 +/- 8.59E-07	2.97E-06 +/- 7.87E-07
7*	ND	1.46E-05 +/- 4.18E-07	1.26E-06 +/- 1.02E-07	5.99E-06 +/- 1.59E-07	ND	2.11E-06 +/- 1.69E-07
8*	NA	NA	NA	NA	NA	NA
9*	ND	2.21E-05 +/- 3.66E-07	1.34E-06 +/- 1.16E-07	6.39E-06 +/- 1.92E-07	ND	1.53E-06 +/- 1.76E-07
10*	NA	NA	NA	NA	NA	NA

CATION MEMBRANE FILTERS (50 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 5.81E-05	4.91E-07 +/- 6.92E-08	6.04E-07 +/- 1.33E-07	1.62E-06 +/- 9.17E-08	< 2.92E-07	< 2.53E-07
2*	ND	9.36E-07 +/- 1.41E-07	< 4.28E-07	1.69E-06 +/- 1.76E-07	< 1.34E-06	< 5.64E-07
3*	ND	6.78E-07 +/- 1.06E-07	ND	1.13E-06 +/- 1.82E-07	ND	ND
4*	ND	5.94E-07 +/- 3.04E-08	< 2.17E-07	6.97E-07 +/- 2.62E-08	ND	ND
5*	ND	5.94E-07 +/- 1.06E-07	ND	7.88E-07 +/- 1.21E-07	ND	ND
6*	< 1.19E-04	5.48E-07 +/- 9.21E-08	< 2.98E-07	7.88E-07 +/- 1.22E-07	< 7.70E-07	< 3.49E-07
7*	ND	6.78E-07 +/- 1.27E-07	1.88E-07 +/- 5.58E-08	4.85E-07 +/- 8.08E-08	ND	ND
8*	NA	NA	NA	NA	NA	NA
9*	ND	7.32E-07 +/- 3.30E-08	ND	< 3.08E-08	ND	ND
10*	NA	NA	NA	NA	NA	NA

FRACTION OF NUCLIDE THAT IS COMPLEXED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	9.55E-01 +/- 6.51E-03	7.26E-01 +/- 6.33E-02	7.79E-01 +/- 1.68E-02	ND	> 9.11E-01
2*	ND	8.72E-01 +/- 1.90E-02	> 8.15E-01	8.65E-01 +/- 1.41E-02	ND	ND
3*	ND	9.46E-01 +/- 9.10E-03	1.00E+00	9.42E-01 +/- 9.01E-03	ND	1.00E+00
4*	NA	NA	NA	NA	NA	NA
5*	ND	9.54E-01 +/- 8.12E-03	1.00E+00	9.33E-01 +/- 1.03E-02	ND	1.00E+00
6*	ND	9.47E-01 +/- 8.95E-03	> 7.98E-01	9.04E-01 +/- 1.46E-02	> 7.07E-01	> 8.95E-01
7*	ND	9.56E-01 +/- 8.04E-03	8.70E-01 +/- 3.48E-02	9.25E-01 +/- 1.17E-02	ND	1.00E+00
8*	NA	NA	NA	NA	NA	NA
9*	ND	9.68E-01 +/- 1.49E-03	1.00E+00	> 9.95E-01	ND	1.00E+00
10*	NA	NA	NA	NA	NA	NA
Mean:	ND	9.43E-01	8.87E-01	9.06E-01	> 7.07E-01	9.68E-01

TABLE 6-6. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: PILGRIM SAMPLE  
(uCi/ml decay corrected to 08/17/84)

UNFILTERED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54
1*	< 8.28E-04	1.01E-03 +/- 1.11E-05	< 1.58E-06	< 1.47E-06	< 3.69E-06
2*	< 3.29E-03	1.64E-03 +/- 1.93E-05	< 7.85E-06	< 5.18E-06	< 1.99E-05
3*	< 2.08E-03	3.14E-03 +/- 4.43E-05	< 2.83E-06	< 3.91E-06	< 1.94E-05
4*	< 1.16E-03	1.92E-03 +/- 2.36E-05	< 1.27E-06	< 1.36E-06	< 4.98E-06
5*	< 3.87E-03	1.17E-03 +/- 1.63E-05	< 5.23E-06	< 3.25E-06	< 2.75E-05
6*	ND	9.87E-04 +/- 9.87E-06	ND	8.26E-07 +/- 1.17E-07	ND
7*	ND	9.13E-04 +/- 6.12E-06	ND	ND	ND
8*	ND	6.58E-03 +/- 1.65E-05	ND	4.61E-07 +/- 1.01E-07	ND
9*	ND	2.65E-03 +/- 2.33E-05	ND	9.92E-07 +/- 1.16E-07	ND
10*	ND	1.64E-03 +/- 2.39E-05	ND	1.99E-06 +/- 1.18E-07	ND

UNFILTERED

SAMPLE	Sb-125	Sr-90	Ni-63	Fe-55	Pu-241
1*	< 2.53E-06	2.99E-07 +/- 1.07E-08	5.61E-04 +/- 8.16E-05	7.29E-07 +/- 1.07E-07	< 4.60E-07
2*	< 1.69E-05	NA	NA	NA	NA
3*	6.42E-06 +/- 2.84E-06	4.70E-07 +/- 2.14E-08	2.14E-03 +/- 3.06E-04	< 1.50E-07	< 5.75E-07
4*	< 5.54E-06	NA	NA	NA	NA
5*	< 1.39E-05	1.03E-07 +/- 4.28E-09	7.14E-04 +/- 1.02E-04	2.14E-07 +/- 2.15E-08	< 4.60E-07
6*	ND	1.17E-07 +/- 5.35E-09	5.10E-04 +/- 7.14E-05	< 4.29E-07	< 3.45E-07
7*	ND	NA	NA	NA	NA
8*	ND	2.01E-07 +/- 8.55E-09	3.06E-03 +/- 4.08E-04	3.65E-07 +/- 4.29E-08	< 4.60E-07
9*	ND	NA	NA	NA	NA
10*	ND	NA	NA	NA	NA

UNFILTERED

SAMPLE	Iron	Nickel	Cobalt	Chromium
1*	NA	NA	NA	NA
2*	NA	NA	NA	NA
3*	< 8.00E-03	1.21E+00 +/- 6.05E-02	< 4.00E-02	< 8.00E-03
4*	NA	NA	NA	NA
5*	< 8.00E-03	4.20E-01 +/- 2.10E-02	< 4.00E-02	< 8.00E-03
6*	< 8.00E-03	3.90E-01 +/- 1.95E-02	< 4.00E-02	< 8.00E-03
7*	NA	NA	NA	NA
8*	< 8.00E-03	1.70E+00 +/- 8.50E-02	< 4.00E-02	< 8.00E-03
9*	NA	NA	NA	NA
10*	NA	NA	NA	NA

TABLE B-6. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: PILGRIM SAMPLE  
(uCi/ml decay corrected to 08/17/84)

(Continued)

CATION MEMBRANE FILTRATE

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	8.76E-04 +/- 1.09E-05	ND	ND	ND	ND
2*	ND	1.38E-03 +/- 2.00E-05	ND	ND	ND	ND
3*	ND	2.17E-03 +/- 2.91E-05	ND	ND	ND	ND
4*	ND	1.62E-03 +/- 2.22E-05	ND	ND	ND	ND
5*	ND	9.77E-04 +/- 1.36E-05	ND	ND	ND	ND
6*	ND	8.28E-04 +/- 1.20E-05	ND	ND	ND	ND
7*	ND	5.85E-04 +/- 6.66E-06	ND	ND	ND	ND
8*	ND	5.68E-03 +/- 5.74E-05	ND	ND	ND	ND
9*	ND	2.26E-03 +/- 1.38E-05	ND	2.08E-06 +/- 3.42E-07	ND	ND
10*	NA	NA	NA	NA	NA	NA

CATION MEMBRANE FILTER

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	1.02E-04 +/- 1.43E-06	ND	ND	ND	ND
2*	ND	1.42E-04 +/- 1.52E-06	ND	ND	ND	ND
3*	ND	2.08E-04 +/- 1.89E-06	ND	ND	ND	ND
4*	ND	1.07E-04 +/- 1.07E-06	ND	ND	ND	ND
5*	ND	8.14E-05 +/- 1.62E-06	ND	ND	ND	ND
6*	ND	8.52E-05 +/- 2.02E-06	ND	< 8.64E-08	ND	ND
7*	NA	NA	NA	NA	NA	NA
8*	ND	3.09E-04 +/- 3.49E-06	ND	ND	ND	ND
9*	ND	1.92E-04 +/- 1.98E-06	ND	ND	ND	ND
10*	NA	NA	NA	NA	NA	NA

FRACTION OF NUCLIDE THAT IS COMPLEXED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	ND	8.95E-01 +/- 1.75E-03	ND	ND	ND	ND
2*	ND	9.07E-01 +/- 1.52E-03	ND	ND	ND	ND
3*	ND	9.13E-01 +/- 1.29E-03	ND	ND	ND	ND
4*	ND	9.38E-01 +/- 9.85E-04	ND	ND	ND	ND
5*	ND	9.23E-01 +/- 1.72E-03	ND	ND	ND	ND
6*	ND	9.07E-01 +/- 2.35E-03	ND	ND	ND	ND
7*	NA	1.00E+00	NA	NA	NA	NA
8*	ND	9.48E-01 +/- 7.41E-04	ND	ND	ND	ND
9*	ND	9.22E-01 +/- 8.66E-04	ND	1.00E+00	ND	ND
10*	NA	NA	NA	NA	NA	NA
Mean:	ND	9.28E-01	ND	1.00E+00	ND	ND

TABLE 6-7. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: PEACH BOTTOM-2 SAMPLE  
(uCi/ml decay corrected to 08/09/84)

UNFILTERED (450 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54
1*	< 3.93E-05	5.47E-05 +/- 7.39E-07	3.12E-06 +/- 1.33E-07	6.17E-06 +/- 1.74E-07	< 5.80E-07
2*	< 1.11E-04	6.81E-05 +/- 1.25E-06	3.46E-06 +/- 3.66E-07	6.32E-06 +/- 5.67E-07	< 1.49E-06
3*	< 1.48E-04	8.06E-05 +/- 2.35E-06	3.19E-06 +/- 4.65E-07	6.96E-06 +/- 4.37E-07	< 1.64E-06
4*	9.61E-05 +/- 2.34E-05	3.87E-05 +/- 1.16E-06	< 2.34E-07	3.33E-06 +/- 3.71E-07	1.51E-06 +/- 2.98E-07
5*	< 2.52E-05	4.87E-05 +/- 1.29E-06	1.60E-06 +/- 1.59E-07	3.11E-06 +/- 1.79E-07	< 4.39E-07
6*	< 3.49E-06	8.31E-05 +/- 1.12E-06	2.40E-06 +/- 1.30E-07	5.92E-06 +/- 2.02E-07	< 6.64E-07
7*	< 5.36E-04	2.38E-04 +/- 4.24E-06	8.89E-06 +/- 1.05E-06	1.92E-05 +/- 1.30E-06	< 5.02E-06
8*	< 5.23E-04	4.46E-04 +/- 4.55E-06	1.14E-05 +/- 6.38E-07	2.22E-05 +/- 6.80E-07	3.82E-06 +/- 1.85E-06
9*	ND	3.21E-04 +/- 6.29E-06	8.28E-06 +/- 8.35E-07	1.62E-05 +/- 8.48E-07	< 3.26E-06
10*	ND	9.03E-04 +/- 1.10E-05	1.06E-05 +/- 3.16E-07	1.98E-05 +/- 2.04E-07	ND

UNFILTERED (450 ml)

SAMPLE	Sb-125	Sr-90	Ni-63	Fe-55	Pu-241
1*	< 6.32E-07	1.07E-08 +/- 2.14E-09	3.06E-06 +/- 4.08E-07	2.16E-06 +/- 2.16E-07	< 8.06E-07
2*	< 1.34E-06	NA	NA	NA	NA
3*	< 2.71E-06	6.42E-09 +/- 2.14E-09	3.06E-06 +/- 4.08E-07	< 4.32E-07	< 1.15E-06
4*	< 5.37E-07	NA	NA	NA	NA
5*	< 3.22E-07	< 6.42E-09	1.84E-06 +/- 3.06E-07	< 2.16E-07	< 6.91E-07
6*	< 7.79E-07	< 6.42E-09	3.37E-06 +/- 5.10E-07	< 4.32E-07	< 6.91E-07
7*	< 6.85E-06	1.28E-08 +/- 2.14E-09	7.14E-06 +/- 1.02E-06	< 4.32E-07	< 9.21E-07
8*	< 3.03E-06	NA	NA	NA	NA
9*	< 4.08E-06	NA	NA	NA	NA
10*	ND	NA	NA	NA	NA

UNFILTERED (450 ml)

SAMPLE	Iron	Nickel	Cobalt	Chromium
1*	< 8.00E-03	7.00E-02 +/- 3.50E-03	< 4.00E-02	< 8.00E-03
2*	NA	NA	NA	NA
3*	< 8.00E-03	7.00E-02 +/- 3.50E-03	< 4.00E-02	< 8.00E-03
4*	NA	NA	NA	NA
5*	< 8.00E-03	7.00E-02 +/- 3.50E-03	< 4.00E-02	< 8.00E-03
6*	< 8.00E-03	7.00E-02 +/- 3.50E-03	< 4.00E-02	< 8.00E-03
7*	< 8.00E-03	1.10E-01 +/- 5.50E-03	< 4.00E-02	< 8.00E-03
8*	NA	NA	NA	NA
9*	NA	NA	NA	NA
10*	NA	NA	NA	NA

TABLE 6-7. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: PEACH BOTTOM-2 SAMPLE (Continued)  
(uCi/ml decay corrected to 08/09/84)

FILTRATE (450 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 2.26E-04	6.59E-05 +/- 2.66E-06	< 3.26E-06	5.82E-06 +/- 7.69E-07	< 2.86E-06	< 3.08E-06
2*	< 8.02E-05	6.25E-05 +/- 8.12E-07	2.97E-06 +/- 1.74E-07	5.82E-06 +/- 3.66E-07	< 1.22E-06	1.31E-06 +/- 3.73E-07
3*	1.27E-04 +/- 2.81E-05	7.66E-05 +/- 1.34E-06	4.01E-06 +/- 3.95E-07	6.82E-06 +/- 6.59E-07	< 1.32E-06	8.92E-07 +/- 3.39E-07
4*	< 1.48E-04	3.62E-05 +/- 1.13E-06	< 2.23E-06	2.54E-06 +/- 3.07E-07	< 1.34E-06	< 1.15E-06
5*	< 1.68E-04	4.32E-05 +/- 5.32E-07	< 1.86E-06	3.00E-06 +/- 3.35E-07	< 2.42E-06	< 1.58E-06
6*	< 2.56E-04	9.29E-05 +/- 2.30E-06	2.40E-06 +/- 6.37E-07	6.15E-06 +/- 5.83E-07	< 5.55E-06	2.31E-06 +/- 1.08E-06
7*	< 6.95E-04	2.12E-04 +/- 5.99E-06	< 9.11E-06	1.70E-05 +/- 1.58E-06	< 8.35E-06	< 4.57E-06
8*	NA	NA	NA	NA	NA	NA
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

FILTER (450 ml)

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 1.26E-05	6.14E-06 +/- 1.01E-07	< 3.78E-08	< 3.22E-08	< 1.37E-07	< 1.15E-07
2*	< 2.02E-06	4.14E-06 +/- 4.80E-08	< 1.96E-08	3.05E-08 +/- 6.89E-09	< 5.06E-08	< 2.60E-08
3*	NA	NA	NA	NA	NA	NA
4*	NA	NA	NA	NA	NA	NA
5*	NA	NA	NA	NA	NA	NA
6*	NA	NA	NA	NA	NA	NA
7*	NA	NA	NA	NA	NA	NA
8*	NA	NA	NA	NA	NA	NA
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

FILTRATE + FILTER

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
1*	< 2.39E-04	7.20E-05 +/- 2.66E-06	< 3.29E-06	< 5.86E-06	< 2.99E-06	< 3.19E-06
2*	< 8.22E-05	6.66E-05 +/- 8.13E-07	< 2.99E-06	5.86E-06 +/- 3.66E-07	< 1.27E-06	< 1.34E-06
3*	NA	NA	NA	NA	NA	NA
4*	NA	NA	NA	NA	NA	NA
5*	NA	NA	NA	NA	NA	NA
6*	NA	NA	NA	NA	NA	NA
7*	NA	NA	NA	NA	NA	NA
8*	NA	NA	NA	NA	NA	NA
9*	NA	NA	NA	NA	NA	NA
10*	NA	NA	NA	NA	NA	NA

TABLE 6-7. RADIONUCLIDE AND STABLE METAL CONCENTRATIONS IN LEACHATES: PEACH BOTTOM-2 SAMPLE (Continued)  
(uCi/ml decay corrected to 08/09/84)

CATION MEMBRANE FILTRATE

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	ND	4.28E-05 +/- 6.80E-07	2.27E-06 +/- 1.16E-07	4.58E-06 +/- 1.56E-07	ND	ND
2*	ND	6.39E-05 +/- 7.98E-07	2.56E-06 +/- 2.30E-07	5.00E-06 +/- 1.37E-07	ND	ND
3*	ND	6.04E-05 +/- 7.85E-07	2.36E-06 +/- 1.38E-07	4.64E-06 +/- 1.32E-07	ND	ND
4*	ND	3.80E-05 +/- 4.45E-07	1.74E-06 +/- 1.18E-07	3.00E-06 +/- 1.33E-07	ND	ND
5*	ND	3.91E-05 +/- 6.37E-07	1.87E-06 +/- 3.18E-07	2.43E-06 +/- 1.87E-07	ND	ND
6*	ND	8.36E-05 +/- 7.52E-07	5.73E-06 +/- 2.95E-07	1.06E-05 +/- 2.74E-07	ND	ND
7*	ND	1.82E-04 +/- 2.52E-06	6.75E-06 +/- 1.97E-07	1.39E-05 +/- 2.86E-07	ND	ND
8*	NA	NA	NA	NA	NA	NA
9*	ND	2.74E-04 +/- 3.26E-06	6.79E-06 +/- 2.24E-07	1.45E-05 +/- 4.14E-07	ND	ND
10*	NA	NA	NA	NA	NA	NA

CATION MEMBRANE FILTER

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	ND	3.08E-06 +/- 6.96E-08	ND	ND	ND	ND
2*	ND	2.05E-06 +/- 3.03E-08	ND	1.67E-08 +/- 6.20E-09	ND	ND
3*	ND	2.27E-06 +/- 4.80E-08	ND	< 3.22E-08	ND	ND
4*	ND	2.36E-06 +/- 8.99E-08	ND	< 5.50E-08	ND	ND
5*	ND	2.07E-06 +/- 6.15E-08	ND	ND	ND	ND
6*	NA	NA	NA	NA	NA	NA
7*	ND	4.37E-06 +/- 8.92E-08	ND	ND	ND	ND
8*	NA	NA	NA	NA	NA	NA
9*	ND	1.31E-05 +/- 2.62E-07	ND	ND	ND	ND
10*	NA	NA	NA	NA	NA	NA

FRACTION OF NUCLIDE THAT IS COMPLEXED

SAMPLE	Co-58	Co-60	Cs-134	Cs-137	Mn-54	Sb-125
=====	=====	=====	=====	=====	=====	=====
1*	ND	9.33E-01 +/- 1.73E-03	1.00E+00	1.00E+00	ND	ND
2*	ND	9.69E-01 +/- 5.83E-04	1.00E+00	9.97E-01 +/- 1.23E-03	NM	NM
3*	ND	9.64E-01 +/- 8.66E-04	1.00E+00	> 9.93E-01 +/- 1.95E-04	ND	ND
4*	ND	9.42E-01 +/- 2.19E-03	1.00E+00	> 9.82E-01 +/- 7.84E-04	ND	ND
5*	ND	9.50E-01 +/- 1.62E-03	1.00E+00	1.00E+00	ND	ND
6*	NA	NA	NA	NA	NA	NA
7*	ND	9.77E-01 +/- 5.63E-04	1.00E+00	1.00E+00	ND	ND
8*	NA	NA	NA	NA	NA	NA
9*	ND	9.54E-01 +/- 1.01E-03	1.00E+00	1.00E+00	ND	ND
10*	NA	NA	NA	NA	NA	NA
Mean:	ND	9.55E-01	1.00E+00	9.96E-01	ND	ND

**APPENDIX H**  
**CONCENTRATIONS OF ORGANIC ACIDS IN LEACHATES**





## **APPENDIX H**

### **CONCENTRATIONS OF ORGANIC ACIDS IN LEACHATES**

The concentrations of organic acids in leachates are given in Table H-1.

TABLE H-1. CONCENTRATIONS OF ORGANIC ACIDS IN LEACHATES (ug/ml)

INDIAN POINT-3 LOMI WASTE FORM

SAMPLE =====	FORMIC ACID =====	PICOLINIC ACID =====
1*	7.20E+00 +/- 9.00E-01	4.61E+00 +/- 6.77E-01
2*	1.35E+01 +/- 9.00E-01	8.67E+00 +/- 6.77E-01
3*	2.03E+01 +/- 9.00E-01	1.99E+01 +/- 6.77E-01
4*	2.03E+01 +/- 1.00E-01	1.81E+01 +/- 6.78E-01
5*	1.87E+01 +/- 2.00E-01	1.36E+01 +/- 6.77E-01
6*	1.48E+01 +/- 8.00E-01	1.09E+01 +/- 6.81E-01
7*	1.30E+01 +/- 6.00E-01	7.28E+00 +/- 6.77E-01
8*	6.02E+01 +/- 4.10E+00	4.75E+01 +/- 6.60E+00
9*	5.94E+01 +/- 4.10E+00	3.99E+01 +/- 2.00E+00
10*	2.00E+01 +/- 5.00E+00	NA

COOPER MIXED BED RESIN AP/CITROX WASTE FORM

SAMPLE =====	OXALIC ACID =====	CITRIC ACID =====
1*	2.33E+00 +/- 4.16E-01	3.77E+00 +/- 2.68E-01
2*	1.83E+00 +/- 4.15E-01	3.03E+00 +/- 3.87E-02
3*	1.21E+00 +/- 4.13E-01	1.20E+00 +/- 1.32E-01
4*	8.61E-01 +/- 4.12E-01	6.73E-01 +/- 4.09E-02
5*	8.70E-01 +/- 4.13E-01	6.72E-01 +/- 4.40E-02
6*	8.85E-01 +/- 4.12E-01	6.72E-01 +/- 6.01E-02
7*	8.36E-01 +/- 4.14E-01	1.17E+00 +/- 4.67E-02
8*	6.17E-01 +/- 4.12E-01	7.97E-01 +/- 4.44E-02
9*	6.64E-01 +/- 4.12E-01	9.08E-01 +/- 3.53E-02
10*	8.65E-01 +/- 4.20E-01	6.50E-01 +/- 4.60E-02

COOPER CATION RESIN AP/CITROX WASTE FORM

SAMPLE =====	OXALIC ACID =====	CITRIC ACID =====
1*	ND	1.65E-01 +/- 3.53E-02
2*	NA	NA
3*	NA	NA
4*	ND	9.91E-02 +/- 3.49E-02
5*	NA	NA
6*	NA	NA
7*	NA	NA
8*	NA	NA
9*	NA	NA
10*	NA	NA

TABLE H-1. CONCENTRATIONS OF ORGANIC ACIDS IN LEACHATES (ug/ml)  
(Continued)

PILGRIM DOW NS-1 WASTE FORM

SAMPLE *****	OXALIC ACID *****	CITRIC ACID *****	EDTA *****	DTPA *****
1*	6.91E-01 +/- 4.13E-01	1.06E+01 +/- 2.82E-01	1.24E+01 +/- 3.44E-01	2.43E-01 +/- 1.29E-01
2*	1.06E+00 +/- 4.15E-01	3.15E+01 +/- 6.16E-01	6.04E+01 +/- 1.98E-01	8.50E-01 +/- 1.39E-01
3*	1.02E+00 +/- 4.13E-01	2.71E+01 +/- 3.27E-01	8.92E+01 +/- 4.05E+00	7.39E-01 +/- 1.36E-01
4*	7.25E-01 +/- 4.13E-01	1.12E+01 +/- 3.83E-02	5.55E+01 +/- 4.00E-01	4.53E-01 +/- 1.31E-01
5*	ND	5.62E+00 +/- 1.79E-01	2.49E+01 +/- 8.58E-01	6.25E-01 +/- 1.34E-01
6*	ND	1.26E+00 +/- 3.48E-02	6.12E+00 +/- 5.75E-01	8.07E-01 +/- 1.38E-01
7*	1.43E+00 +/- 4.12E-01	1.23E+00 +/- 9.79E-02	9.36E-01 +/- 9.41E-02	9.88E-01 +/- 1.42E-01
8*	1.72E+00 +/- 4.35E-01	2.41E+00 +/- 3.53E-02	3.93E+00 +/- 1.06E-01	3.28E+00 +/- 3.70E-01
9*	9.41E-01 +/- 4.20E-01	2.65E+00 +/- 5.09E-02	3.76E+01 +/- 2.10E+00	1.24E+00 +/- 1.38E-01
10*	NA	NA	NA	NA

PEACH BOTTOM-2 CAN-DECON WASTE FORM

SAMPLE *****	OXALIC ACID *****	CITRIC ACID *****	EDTA *****
1*	1.89E+00 +/- 4.40E-01	3.71E+00 +/- 6.61E-02	2.73E+00 +/- 8.33E-02
2*	8.87E-01 +/- 4.12E-01	9.17E-01 +/- 3.63E-02	3.89E+00 +/- 2.03E-01
3*	1.40E+00 +/- 4.13E-01	7.59E-01 +/- 3.71E-02	6.53E+00 +/- 3.28E-01
4*	7.85E-01 +/- 4.12E-01	1.20E+00 +/- 3.47E-02	2.47E+00 +/- 7.68E-02
5*	7.80E-01 +/- 4.12E-01	5.65E-01 +/- 4.23E-02	1.45E+00 +/- 1.18E-01
6*	7.00E-01 +/- 4.12E-01	5.75E-01 +/- 3.61E-02	5.43E+00 +/- 2.76E-01
7*	1.53E+00 +/- 4.13E-01	1.40E+00 +/- 5.78E-02	7.48E+00 +/- 2.36E-01
8*	NA	NA	NA
9*	5.55E-01 +/- 4.12E-01	5.20E-01 +/- 3.47E-02	5.41E-01 +/- 1.05E-01
10*	NA	NA	NA

TABLE H-1. CONCENTRATIONS OF ORGANIC ACIDS IN LEACHATES (ug/ml)  
(Continued)

MILLSTONE-1 F33 CAN-DECON WASTE FORM

SAMPLE =====	OXALIC ACID =====	CITRIC ACID =====	EDTA =====
1*	6.01E-01 +/- 4.12E-01	1.31E-01 +/- 6.30E-02	1.53E+00 +/- 1.00E-01
2*	9.10E-01 +/- 4.12E-01	1.58E-01 +/- 3.47E-02	3.34E+00 +/- 2.43E-01
3*	1.16E+00 +/- 4.49E-01	2.73E-01 +/- 5.66E-02	6.65E+00 +/- 9.94E-02
4*	1.32E+00 +/- 4.13E-01	4.44E-01 +/- 4.97E-02	9.51E+00 +/- 4.89E-01
5*	1.05E+00 +/- 4.13E-01	1.37E-01 +/- 3.55E-02	5.25E+00 +/- 2.28E-01
6*	8.94E-01 +/- 4.13E-01	1.53E-01 +/- 3.57E-02	3.72E+00 +/- 1.70E-01
7*	7.37E-01 +/- 4.12E-01	9.20E-01 +/- 3.77E-02	2.38E+00 +/- 1.07E-01
8*	1.18E+00 +/- 4.12E-01	9.31E-01 +/- 3.55E-02	1.61E+01 +/- 6.62E-01
9*	1.68E+00 +/- 4.16E-01	9.80E-01 +/- 6.43E-02	1.80E+01 +/- 7.41E-01
10*	6.01E-01 +/- 4.12E-01	1.01E+00 +/- 6.56E-02	1.30E+01 +/- 5.38E-01

MILLSTONE-1 F201 CAN-DECON WASTE FORM

SAMPLE =====	OXALIC ACID =====	CITRIC ACID =====	EDTA =====
1*	NA	NA	NA
2*	8.43E-01 +/- 4.13E-01	1.13E-01 +/- 3.71E-02	1.75E+00 +/- 8.73E-02
3*	NA	NA	NA
4*	1.28E+00 +/- 4.14E-01	1.99E-01 +/- 3.65E-02	6.54E+00 +/- 6.23E-01
5*	NA	NA	NA
6*	NA	NA	NA
7*	6.28E-01 +/- 4.12E-01	7.67E-01 +/- 7.46E-02	2.02E+00 +/- 1.42E-01
8*	NA	NA	NA
9*	NA	NA	NA
10*	NA	NA	NA

**APPENDIX I**

**QUANTITIES OF RADIONUCLIDES, METALS, AND ORGANIC  
ACIDS SOLIDIFIED IN EACH LINER**



## **APPENDIX I**

### **QUANTITIES OF RADIONUCLIDES, METALS, AND ORGANIC ACIDS SOLIDIFIED IN EACH LINER**

The total activities solidified in each liner are given in Table I-1, and the total quantities of metals

and organic acids solidified in each liner are given in Table I-2.



Table 1-1. TOTAL ACTIVITY SOLIDIFIED IN EACH LINER (Ci/liner)  
(Activities are decay-corrected to sample collection dates)

Nuclide	Indian Point-3 (06/24/85)		Cooper (MBR) (11/30/84)		Cooper (CR) (11/30/84)		Millstone-1 (06/20/84)		Pilgrim (08/17/84)		Peach Bottom-2 (08/09/84)	
H-3	1.76E-03 +/- 1.73E-04		8.06E-04 +/- 2.12E-05		1.02E-05 +/- 5.22E-07		1.45E-03 +/- 5.08E-05		3.32E-05 +/- 1.40E-06		3.82E-02 +/- 1.47E-03	
C-14	1.99E-04 +/- 3.31E-05	< 2.54E-05			< 3.13E-05		5.49E-05 +/- 3.05E-06		9.36E-05 +/- 1.87E-05		NA	
Mn-54	1.66E+00 +/- 2.42E-02		3.07E-01 +/- 1.48E-02		2.71E+00 +/- 7.83E-02		3.10E+00 +/- 5.08E-02		5.93E-01 +/- 3.12E-02		2.47E-01 +/- 1.21E-03	
Fe-55	5.83E+00 +/- 2.76E-01		8.48E-01 +/- 4.24E-02		1.49E+01 +/- 7.83E-01		2.54E+01 +/- 1.27E+00		4.21E+00 +/- 1.56E-01		NA	
Co-57	1.52E-01 +/- 1.38E-02		ND		ND		ND		ND		ND	
Co-58	7.76E+01 +/- 3.45E-01		8.27E-02 +/- 1.27E-02		1.85E+00 +/- 1.57E-01		6.35E-01 +/- 2.54E-01		1.40E-01 +/- 1.40E-01		ND	
Co-60	1.36E+01 +/- 3.45E-02		1.55E+00 +/- 1.70E-02		1.75E+01 +/- 7.83E-02		9.92E+00 +/- 3.81E-02		8.10E+00 +/- 3.12E-02		1.73E+01 +/- 1.47E-01	
Ni-63	1.66E+01 +/- 6.90E-01		7.63E-01 +/- 4.24E-02		2.69E+01 +/- 1.30E+00		1.10E+00 +/- 5.08E-02		4.37E-01 +/- 1.56E-02		NA	
Sr-89	1.73E-02 +/- 3.45E-03	< 4.24E-05			< 2.35E-05		1.65E-02 +/- 5.08E-03		1.56E-03 +/- 9.36E-04		NA	
Sr-90	4.48E-04 +/- 6.90E-05		4.24E-04 +/- 6.36E-05		1.83E-04 +/- 2.61E-05		7.49E-04 +/- 5.08E-05		2.34E-04 +/- 3.12E-05		2.35E-05 +/- 4.41E-06	
Nb-95	ND		1.91E-02 +/- 6.36E-03		ND		ND		ND		ND	
Tc-99	NA		2.33E-04 +/- 6.36E-05		7.83E-05 +/- 7.83E-05		ND		3.43E-05 +/- 9.36E-06		NA	
I-129	< 3.45E-05	< 6.36E-05			< 1.04E-04		< 2.54E-06		< 3.12E-06		NA	
Sb-125	3.21E-03 +/- 2.41E-04		8.90E-02 +/- 1.06E-02		< 5.22E-04		2.03E-01 +/- 5.08E-02		1.31E-02 +/- 6.24E-04	< 8.82E-05		
Cs-137	4.62E-01 +/- 1.48E-02		4.39E-04 +/- 4.24E-05		< 5.22E-04		2.72E-03 +/- 1.27E-04		4.68E-02 +/- 1.56E-02		4.16E-03 +/- 2.25E-05	
Subtotal:	1.16E+02 +/- 8.21E-01		3.66E+00 +/- 6.65E-02		6.38E+01 +/- 1.53E+00		4.04E+01 +/- 1.30E+00		1.35E+01 +/- 2.16E-01		1.76E+01 +/- 1.47E-01	
Pu-238	4.00E-05 +/- 6.90E-07		6.57E-05 +/- 4.24E-06		2.01E-04 +/- 7.83E-06		6.22E-03 +/- 1.27E-04		3.43E-04 +/- 1.56E-05		NA	NA
Pu-239/40	4.48E-05 +/- 2.42E-06		3.18E-05 +/- 1.70E-06		7.83E-05 +/- 5.22E-05		3.30E-03 +/- 6.35E-05		4.84E-04 +/- 1.56E-05		NA	NA
Pu-241	1.69E-01 +/- 2.07E-02		4.66E-02 +/- 2.12E-03		2.64E-02 +/- 1.04E-03		4.45E-01 +/- 1.27E-02		5.46E-02 +/- 1.56E-03		7.50E-03 +/- 8.82E-04	
Am-241	3.24E-05 +/- 1.38E-06		4.98E-05 +/- 6.36E-07		2.74E-04 +/- 7.83E-06		5.07E-03 +/- 5.08E-05		8.27E-06 +/- 3.12E-07		NA	NA
Cm-242	6.56E-04 +/- 3.45E-05		1.35E-04 +/- 1.27E-06		1.77E-04 +/- 5.22E-06		5.21E-04 +/- 2.54E-05		2.71E-05 +/- 9.36E-07		NA	NA
Cm-244	3.69E-05 +/- 3.10E-06		1.41E-04 +/- 1.91E-06		2.11E-04 +/- 5.22E-06		4.32E-03 +/- 5.08E-05		2.15E-05 +/- 3.12E-07		NA	NA
Subtotal:	1.70E-01 +/- 2.07E-02		4.71E-02 +/- 2.12E-03		2.73E-02 +/- 1.05E-03		4.64E-01 +/- 1.27E-02		5.55E-02 +/- 1.56E-03		7.50E-03 +/- 8.82E-04	
	(m*3)	(ft*3)	(m*3)	(ft*3)	(m*3)	(ft*3)	(m*3)	(ft*3)	(m*3)	(ft*3)	(m*3)	(ft*3)
Waste volume:	3.45	122	2.12	75	2.61	92	1.27	45	1.56	55	1.47	52
Liner type:	NA	---	L8-120	---	L14-170	---	L6-80	---	NA	---	L6-80	---
Liner capacity:	NA	---	3.28	116	4.79	169	2.24	79	NA	---	2.24	79

Table I-2. TOTAL QUANTITIES OF METALS AND ORGANIC ACIDS SOLIDIFIED IN EACH LINER (kg/liner)

Metal/ Acid	Indian Point-3 (06/24/85)	Cooper (MBR) (11/30/84)	Cooper (CR) (11/30/84)	Millstone-1 (06/20/84)	Pilgrim (08/17/84)	Peach Bottom-2 (08/09/84)
=====	=====	=====	=====	=====	=====	=====
Cr	3.11E-01 +/- 1.73E-02	NA	7.83E-01 +/- 2.61E-01	4.45E-01 +/- 2.54E-02	NA	NA
Fe	2.28E+00 +/- 1.04E-01	1.10E+00 +/- 1.27E-01	2.87E+01 +/- 1.04E+01	7.37E+00 +/- 3.81E-01	1.72E+00 +/- 7.80E-02	NA
Co	< 4.00E-02	< 3.00E-02	1.57E-01 +/- 5.22E-02	2.03E-02 +/- 1.02E-03	1.50E-02 +/- 7.80E-04	NA
Ni	2.46E+00 +/- 1.39E-01	1.57E-01 +/- 1.27E-02	3.13E+00 +/- 1.31E+00	2.67E+00 +/- 1.27E-01	2.18E-01 +/- 1.09E-02	NA
=====	=====	=====	=====	=====	=====	=====
Subtotal:	5.09E+00 +/- 1.74E-01	1.29E+00 +/- 1.28E-01	3.28E+01 +/- 1.05E+01	1.05E+01 +/- 4.02E-01	1.95E+00 +/- 7.88E-02	NA
Citric	ND	5.30E+01 +/- 2.12E+00	2.51E-01 +/- 2.87E-02	8.00E+00 +/- 3.81E-01	4.68E+00 +/- 1.56E-01	7.47E+00 +/- 8.84E-02
Oxalic	ND	3.43E+01 +/- 6.36E-01	7.05E-01 +/- 2.61E-03	9.40E+00 +/- 2.54E-01	4.57E-01 +/- 1.25E-02	8.69E+00 +/- 1.03E-01
EDTA	ND	ND	ND	7.75E+00 +/- 5.08E-01	5.77E+00 +/- 4.68E-01	7.20E+00 +/- 4.41E-01
DTPA	ND	ND	ND	ND	5.65E-01 +/- 1.40E-02	ND
Picolinic	1.42E+01 +/- 6.93E-01	ND	ND	ND	ND	ND
=====	=====	=====	=====	=====	=====	=====
Subtotal:	1.42E+01 +/- 6.93E-01	8.73E+01 +/- 2.21E+00	9.56E-01 +/- 2.88E-02	2.52E+01 +/- 6.84E-01	1.15E+01 +/- 4.94E-01	2.34E+01 +/- 4.61E-01
=====	=====	=====	=====	=====	=====	=====



NRC FORM 338 (2-84) NRCM 1102, 3201, 3202 <b>BIBLIOGRAPHIC DATA SHEET</b> SEE INSTRUCTIONS ON THE REVERSE		U.S. NUCLEAR REGULATORY COMMISSION		1 REPORT NUMBER (Assigned by TIDC add Vol. No., if any) NUREG/CR-5224 EGG-2549	
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5 AUTHOR(S) C.V. McIsaac, J.W. Mandler				4 DATE REPORT COMPLETED MONTH: September YEAR: 1988	
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11a TYPE OF REPORT  b PERIOD COVERED (Include dates)				12 SUPPLEMENTARY NOTES	
13 ABSTRACT (200 words or less) <p>Data are presented on the release of radionuclides, stable metals, and organic reagents from decontamination ion-exchange resin wastes solidified in Portland cement. Both solidified and unsolidified decontamination resin waste samples were collected from five commercial light water reactors following chemical decontaminations of primary coolant systems. The decontaminations were performed using the Can-Decon, AP/Citrox, Dow NS-1, and LOMI processes. Samples of unsolidified decontamination resin waste were analyzed for concentrations of 10 CFR Part 61 radionuclides, selected transition metals, and organic chelating and complexing agents. The organic acids included oxalic acid, citric acid, formic acid, picolinic acid, ethylenediaminetetraacetic acid (EDTA), and diethylenetriaminepentaacetic acid (DPTA). Seven solidified resin waste samples were leached in deionized water following the ANS 16.1 leach-test procedure. Release rates and leachability indexes of radionuclides, transition metals, and organic acids were determined.</p> <p>Analytical methods are described which were used to measure organic acids in resin wastes and in leachate solutions generated during leach-testing of waste form samples. Gas-liquid and ion chromatography methods were developed which allowed detection of organic acids in the tenths-of-a-ppm range.</p>					
14 DOCUMENT ANALYSIS • KEYWORDS/DESCRIPTORS leach-testing decontamination ion-exchange resin wastes  15 IDENTIFIERS/OPEN ENDED TERMS				15 AVAILABILITY STATEMENT Unlimited  16 SECURITY CLASSIFICATION (This paper) Unclassified (This report) Unclassified 17 NUMBER OF PAGES  18 PRICE	

