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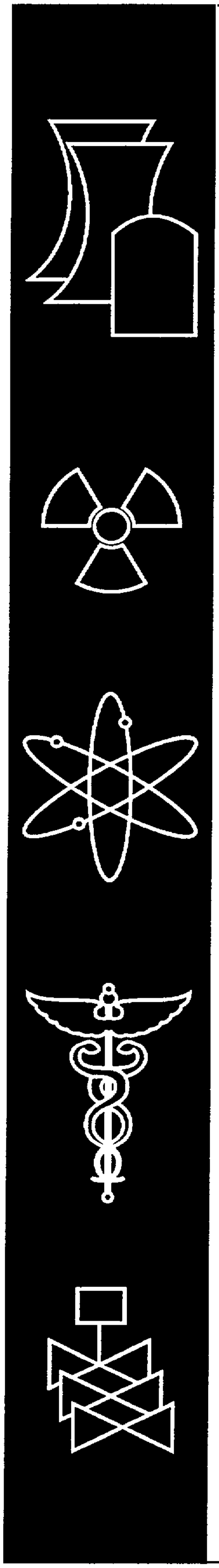

Release of Radionuclides and Chelating Agents From Full-System Decontamination Ion-Exchange Resins

Idaho National Engineering and Environmental Laboratory

U.S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
Washington, DC 20555-0001

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ABSTRACT

As part of a study being performed for the Nuclear Regulatory Commission (NRC), small-scale waste specimens were collected following an AP/CAN-DEREM full-system decontamination of the reactor vessel of the Indian Point 2 reactor vessel in March 1995. CAN-DEREM is a modified version of a reduction process developed by Atomic Energy of Canada Ltd. for that country's heavy-water CANDU reactors. The INEEL research was undertaken to support licensing needs for reviewing radiological and chemical data and information to address uncertainties associated with disposal of low-level radioactive decontamination waste in performance assessment modeling. This report presents information on radionuclide and chelate characteristics of decontamination radwaste on ion-exchange resins. The information is expected to reduce uncertainties associated with developing radionuclide inventories for the performance assessment.

Job Code A6359—Characteristics Level Radioactive Waste:
Decontamination Waste

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

The purpose of this research program was to characterize the ion-exchange resins for radionuclide and chelating content, perform leaching experiments on the ion-exchange resins, and determine release rates for radionuclides and chelating agents from a full-system decontamination when subjected to a simulated Hanford groundwater. In this study, plant personnel collected untreated ion-exchange resin wastes from resin storage tanks following decontamination of the reactor vessel at Indian Point 2 in March 1995. A five-step AP/CAN-DEREM process was performed on the reactor vessel following removal of the reactor fuel and the borated primary coolant water.

The INEEL research was undertaken to support licensing needs for reviewing radiological and chemical data and information to address uncertainties associated with disposal of low-level radioactive decontamination waste in performance assessment modeling. This report presents information on radionuclide and chelate characteristics of decontamination radwaste on ion-exchange resins. The information is expected to reduce uncertainties associated with developing radionuclide inventories for the performance assessment.

The five-step AP/CAN-DEREM decontamination process is a modified version of a reduction process developed by Atomic Energy of Canada Ltd. (AECL) for that country's heavy-water CANDU reactors. Con Edison had funded some of AECL's original development of the process and later worked with ESEERCO in developing the AP (alkaline permanganate) pretreatment step. Principal reagents used in this process include several proprietary chelating agents. Although based on prior formulations, it is expected that oxalic acid, citric acid, and EDTA (ethylene diaminetetraacetic) are present. These components can function as chelating agents. Chelating agents are used in reactor system decontamination formulas because they form strong complexes with actinides, lanthanides, heavy metals, and transition metals and help them to stay in solution. These chemical decontamination solutions, once used, are treated with ion-exchange resins to extract soluble metals and chemicals; these resins constitute the final waste to be processed and disposed of from the decontamination process.

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

A simplified leach test was performed to assess the release of chelating agents and radionuclides from the material. Measurements were performed for pH and conductivity of the leachate; for the concentrations of radionuclides, stable metals, and expected chelating agents in the waste; and for the releases of these constituents from the waste resin into the leachant. The releases to the leachant are quantified in terms of absolute and fractional release rates, and cumulative fractional release.

Several transuranic radionuclides, including ^{239}Pu, ^{241}Am, $^{242.244}$Cm , ^{241}Pu, and ^{238}Pu, were found in the resins. The highest radionuclide concentration was for ^{241}Pu at 1.00E+5 pCi/g in the cation resin. The radionuclide concentrations for the transuranics are 1–2 orders of magnitude higher than those found in previous decontamination ion-exchange resins analyzed as part of this project and summarized in NUREG/CR 6201.

Primary conclusions of this evaluation are that the fractional release rates for the decontamination radionuclides ^{60}Co, ^{59,63}Ni, ^{54}Mn, and ^{65}Zn range from 3E-3 to 1E-1 or 0.3–10% for leach periods up to 93 days. This is a fairly highly release for these radionuclides and is considerably higher than that expected from cement solidified waste forms. Typically, the release rates from the anion and deborating resins are higher than those from the cation resin, which may be considered more likely because the decontamination radionuclides are cations.

There appears to be little difference in the release over 37 and 93 days, which suggests that most of the radioactive material release may occur relatively early in the leach period, with little additional release over the longer term. These data would suggest that most of the inventory is retained in the resin and is not subject to significant release until the resin begins to age and deteriorate, which generally occurs relatively rapidly.

Despite containing much higher radionuclide activities, the leachate from the cation resin does not contain the highest concentrations of corrosion product derived transition metals; the anion resin leachate contains the most. And, in general, the leachates from the deborating resin contain the lowest concentrations of transition metals. This may be likely, as the cation resin may bind the cation decontamination elements more tightly than does the anion or deborating resin.

Only the cation resin leachate contained measurable concentrations of EDTA. The concentration. (70.1 mg/L) is equivalent to 2.4×10^{-4} M. EDTA, which is expected to be the primary chelating agent present and appears to be largely destroyed during the CAN-DEREM process, with only a low concentration remaining in the decontamination waste. This suggests that potential chelating agent mobility of radionuclides in wastes may be relatively limited due to the small amount of EDTA present.

FOREWORD

This contractor technical report was prepared by the Idaho National Engineering and Environmental Laboratory[1] under their DOE Interagency Work Order (JCN A6359) with the Radiation Protection, Environmental Risk and Waste Management Branch, Division of Systems Analysis and Regulatory Effectiveness, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission.

This report is the last in a series of contractor reports documenting INEEL's research results on decontamination of low-level radioactive waste collected from operating nuclear power stations.

The report documents the research results from studies performed on decontamination radwaste obtained from a full reactor core decontamination of the Indian Point 2 nuclear station using the newly developed CAN-DEREM decontamination process

The INEEL research was undertaken to support licensing needs for reviewing radiological and chemical data and information to address uncertainties associated with disposal of low-level radioactive decontamination waste in performance assessment modeling as outlined in *A Performance Assessment Methodology for Low-Level Radioactive Waste Disposal Facilities*, NUREG-1573. This INEEL report presents information on radionuclide and chelate characteristics of decontamination radwaste on ion-exchange resins. The information is expected to reduce uncertainties associated with developing radionuclide inventories for the performance assessment. The report also describes a leaching methodology for decontamination waste on ion-exchange resins and presents data on the leaching of radionuclides and chelating agents from decontamination waste. The leaching information can be used in source term models to estimate source term releases in performance assessments and as data for research studies to evaluate the formation and behavior of radionuclide-chelating complexes released to the groundwater from waste disposal facilities.

This report is not a substitute for NRC regulations, and compliance is not required. The approaches and methods described in this NUREG/CR report are provided for information only. Publication of the report does not necessarily constitute NRC approval or agreement with the information contained herein. Use of product or trade names is for identification only and does not constitute endorsement by the NRC or the Idaho National Engineering and Environmental Laboratory.

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ACRONYMS

AECL	Atomic Energy of Canada, Ltd.
ANSI/ANS	American National Standards Institute/American Nuclear Society
AP	alkaline permanganate
CAN-DEREM	Proprietary AECL process
CFR	cumulative fractional release
HPGe	hyperpure germanium
ICP-AES	inductively coupled plasma–atomic emission spectroscopy
INEEL	Idaho National Engineering and Environmental Laboratory
ISO	International Standards Organization

Release of Radionuclides and Chelating Agents from Full-System Decontamination Ion-Exchange Resins

INTRODUCTION

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

The AP/CAN-DEREM process is among the chemical processes most frequently used to decontaminate primary system components. Principal reagents used in this process are proprietary chelating agents, which may include citric acid, oxalic acid, or EDTA. Chelating agents are used in reactor system decontamination formulas because they form strong complexes with actinides, lanthanides, heavy metals, and transition metals, and help them to stay in solution. These chemical decontamination solutions, once used, are treated with ion-exchange resins to extract soluble metals and chemicals; these resins constitute the final waste to be processed and disposed of from the decontamination process. The resin wastes contain quantities of chelates or complexing agents in addition to inventories of radioactive corrosion products and lesser inventories of fission products. A potential problem with the chelated decontamination wastes is the potential for increased solubility of the organo-radionuclide complexes in groundwaters at low-level waste repositories. The resins from this process are not currently being solidified so the testing being performed as part of this program will be performed on the exhausted resin material.

The U.S. Nuclear Regulatory Commission (NRC) is concerned with safe disposal of these chemical decontamination wastes and has made provisions for their disposal in "Licensing Requirements for Land Disposal of Radioactive Waste" (U.S. Code of Federal Regulations Standard 10, Part 61). Section 61.56 states requirements for the stability of waste forms that must be met for the waste form to be acceptable for near-surface disposal. Additional requirements for the disposal of chelated wastes are also given in burial site regulations. These additional references to chelating agents in Part 61 include Part 61.2 (definitions), Part 12.12 (f)(the description of the construction and operation of the land disposal facility must also include a description of the methods be employed in the handling and disposal of wastes containing chelating agents), and the Statement of Considerations to 10 CFR Part 61 which, among other issues, indicates that chelating agents have been shown to increase the migration of certain sites and states it is the Commission's desire to evaluate the disposal of large quantities of chelating agents on a case-by-case basis.

In Section 61.54, the wastes are classified as Class A, B, or C, based on the concentrations of radionuclides in the wastes. Class A wastes have lower concentrations and may be disposed of without stabilization; however, Class A wastes buried with Class B and C wastes must be stabilized. Class B and C wastes must be structurally stabilized to ensure that the waste form does not degrade and does not promote slumping, collapse, or failure of the cap or cover of the near-surface disposal trench. In addition, the stability of the waste form limits exposure to inadvertent intruders. Class B and C low-level wastes from light water reactors (LWRs) may be solidified at LWR sites using cement to meet the stability requirements of the NRC's "Technical Position on Waste Form," Revision 1. Solidification of these wastes is intended to provide the structural stability needed to ensure that no collapse of the disposal trench occurs and that the release of radionuclides via leaching is minimized.

Test procedures to demonstrate waste form stability and to quantify leachability for the low-level wastes from LWRs have been specified by the NRC in the Low-Level Waste Management Branch's "Technical Position on Waste Form,"[1] published in 1983, and in the "Technical Position on Waste Form," Revision 1,[2] published January 1991. Both publications of the Technical Position specify that small-scale waste-form specimens must be prepared and tested to certify the stability of the full-scale waste form. However, in the case of the Indian Point-2 full-system decontamination ion-exchange resins, which were disposed of in high integrity containers (HIC) as unsolidified resin, leachability testing was performed directly on the resin wastes. Consequently, the full ANSI 16.1 procedure was not used.

The leachability of decontamination ion-exchange resin wastes that contain chelating agents was evaluated because the leachability of these wastes is a function of a number of factors, including the chemical characteristics of the radionuclide being leached, resin waste chemistry, and chelating agent. Burial site hydrology and groundwater chemistry also influence leaching rates, as do cyclic wet and dry conditions. The complexity of the interactions that occur among radionuclides, chelating agents, groundwater, and soil introduces uncertainties into the models used to predict the impact of the decontamination resin wastes on the performance of shallow-land burial sites. Therefore, it is important to establish a database on the stability and leachability of decontamination resin wastes representative of wastes commonly generated at operating commercial power stations in leachants that are expected to be representative or more aggressive than the actual disposal site groundwaters. In this case, the simulated Hanford groundwater was chosen as a representative aggressive leachant, and the testing used was a modified immersion test rather than the ANSI 16.1 standard.[3] This is primarily due to the fact that the resins being were not solidified but were samples of discrete resin particles being tested with no surrounding barrier material.

A number of studies have been performed that address the leachability of limited decontaminations followed by waste form solidification. Common to these studies is an emphasis on more realistic leaching situations involving local groundwater,[4,5,6] seawater, and actual nuclear power plant resin wastes.[5,6,7,8,9] In these studies, solidified ion-exchange resin specimens were subjected to leach tests following either the ISO 6961 leach-test procedure[10] or the ANSI/ANS 16.1 leach-test procedure. Both of these methods are nonequilibrium tests in the respect that the solidified waste-form specimen is completely immersed over an extended period of time in a large volume of leachate that is periodically replaced with new leachant. More recent studies[11-15] performed using various waste types have suggested that leachant effects, limited solubility, and breakdown of organo-radionuclide complexes may limit the effects of chelating agents on releases from waste forms. As noted previously, the current study does not address solidified waste forms; however, it is expected that some of the same release phenomena identified for solidified wastes will be present for the resin wastes with no barrier material.

Differences in leaching behavior have been observed in the studies that appear to be a result of changes in leachant composition. The slower release rates are thought to occur because of the much higher ionic strength of the seawater leachant and cement/waste-leachant reactions. However, studies at the Idaho National Engineering and Environmental Laboratory (INEEL) (Reference 9) indicate that the effect of seawater on radionuclide releases from solidified decontamination ion-exchange resins varies and may be similar to groundwaters.

The present study is a continuation of studies summarized in Reference 9 previously performed at the INEEL for the NRC that measured the compressive strength and leachability of cement-solidified evaporator concentrates and decontamination ion-exchange bead resin waste forms leached in deionized water, groundwaters, and seawater. The studies at the INEEL are the only continuing studies in which actual commercial nuclear power plant decontamination radioactive waste is being evaluated.

The following sections present a nonproprietary description of the five-step AP/CAN-DEREM process, followed by a description of the experimental procedures used and a summary of the results of the measurements and implications relative to the quantities of radionuclides expected in resin wastes disposed of at waste sites.

FIVE-STEP AP/CAN-DEREM PROCESS

This section describes the five-step AP/CAN-DEREM process used at Indian Point-2, including a summary description of the equipment and chemistry associated with the process. More detailed descriptions are presented in the proceedings of the 1995 EPRI Seminar on Decontamination.[10] This description of the chemistry involved in the process is brief, as many aspects of the AP/CAN-DEREM process are proprietary. The five-step CAN-DEREM process comprises the following elements: (1) CAN-DEREM, (2) alkaline permanganate (AP) – Destruction, (3) CAN-DEREM, (4) alkaline permanganate (AP) destruction, (5) CAN-DEREM. The chemical objectives and the relative amounts of the chemical constituents used and the amount of cleanup resin used for each step are summarized below:

- CAN-DEREM. The CAN-DEREM agent is a proprietary compound developed by Atomic Energy of Canada Limited (AECL). It is a combination of weak organic acids, and the process requires that about 1000 ppm of the CAN-DEREM agent be in the reactor coolant system for the process to be effective. Replenishment of a primary agent may be required during the process if the concentration of the agent falls below this level. It has been estimated that about 800 lb of CAN-DEREM agent would be required for a four-loop PWR, and about 200 lb of the agent replenished.

  As a combination of weak organic acids, the CAN-DEREM agent is removed on anion resin. The published rate of removal is 60 g of CAN-DEREM per liter of resin. In addition, a 1:6 ratio of cation resin is added as a final polishing agent for the solution.

- AP-Destruction. The alkaline potassium permanganate (AP) step is employed to remove the protective chromium from the corrosion film; it renders the corrosion film susceptible to being attached by the CAN-DEREM agents. The maximum expected concentration is potassium permanganate ($KMnO_4$). This reagent is reduced during the AP step and forms MnO_2, a brown precipitate. Additional injections of the AP may be required. It is estimated for full system decontamination that about 1600 lb of resin may be required, considering initial injection and replenishments. In addition, to achieve the required pH = 10.5 for the AP step, about 2000 lb of NaOH may be required for full system decontamination.

  The AP step is followed by a destruction step, which reduces all the $KMnO_4$ remaining to a water-soluble manganous ion (Mn^{+2}) for removal. This destruction also reduces all the manganese dioxide to manganous ion, dissolving all the precipitate. Oxalic acid dihydrate is typically employed for this reduction/destruction process, since the reaction product of the oxalic acid is carbon dioxide, a gas.

The above process is repeated as described for the five-step process. The cations from the reactor vessel walls (i.e., Fe^{+3}, Ni^{+2}, and Co^{+2}) are removed with a strong acid resin and with a strong base resin during the process. Initial estimates of the resin loading are shown in Table1.

Table 1. Estimates of resin loading during the five-step CAN-DEREM process (high boron content).

Resin	Chelates (%)[a]	Activity (%)
Cation	4	75
Mixed Bed	6	5
AP Cation Resin	0	7
AP Mixed Bed 3/11[b]	0.3	13

a. Does not equal 100%, due to the destruction of some chelate constituents.

b. AP mixed bed 3/11 = deborating.

EXPERIMENTAL PROCEDURES

This section summarizes the experimental procedures used in the study. Many are specific to the characterization of commercial reactor waste streams. Limited information is available on the full-system decontamination process due to its proprietary nature. Plant staff collected the resin wastes at the Indian Point-2 following the AP/CAN-DEREM full-system decontamination of the Indian Point 2 reactor vessel in March 1995. The samples of anion, cation, and mixed-bed resin wastes collected were expected to be representative of the wastes produced from this process. Summarized below are the analytical methods used and the leach test procedure.

Analytical Methods

Analysis of the Indian Point resins to assess leachability requires knowledge of the initial inventories of the resin waste being tested and a knowledge of their concentrations in the leachates generated during leach testing. In order to obtain a basis for estimating the initial inventories of species of interest, samples of resin waste and leachate were analyzed using several different analytical techniques. Because of the differences in their physical characteristics and because the concentrations of radionuclides and metals in the leachates were expected to be as much as several orders of magnitude lower in the leachates compared to the resins, the methods required to prepare the resin and leachate samples for analysis were generally different.

Prior to analysis, leachate solutions were partitioned into volumetric samples. 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, gas proportional counting, inductively coupled plasma-atomic emission spectroscopy, and ion chromatography. The detection of chelating agents in some leachate solutions required that the chelating agent be chemically separated from other components of the sample prior to ion chromatography analysis.

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

Radionuclide Analysis

Resin waste samples were analyzed for radionuclides specified in 10 CFR 61 (i.e., ^{14}C, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{241}Am, and ^{244}Cm) and for other radionuclides using standard environmental analysis procedures. Concentrations of gamma-emitting radionuclides (e.g., ^{60}Co, ^{137}Cs, and other measurable gamma emitters) in each resin sample were measured by diluting an aliquot of the dissolved resin with water and analyzing this volumetric sample using gamma-spectrometric techniques. When resin waste-form samples were analyzed, a small sample of the solidified waste was analyzed directly at a distance of 10 cm from the detector. The HPGe spectrometers used to analyze these samples were calibrated using reference sources traceable to the National Institute of Standards and Technology.

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

The procedure used to analyze the leachates for ^{238}Pu, ^{239}Pu, ^{241}Am, and ^{244}Cm was the same as that used for the resins in that a fusion and the following analysis program were the same for both the resin wastes samples and the leachates. The resin and waste-form samples analyzed for concentrations of transuranic isotopes were wet-ashed using nitric, sulfuric, and perchloric acids. This procedure was followed by a fusion to dissolve any remaining undissolved compounds. The fusion was then dissolved in HCl, followed by a precipitation and redissolution for clean up purposes. The final precipitate was dissolved in acid, and then the solution was oxidized to adjust the oxidation state of plutonium. Americium and curium were precipitated as fluorides and, following filtration, this precipitate was mounted for analysis using a high-resolution alpha spectrometer. The filtrate was reduced, and plutonium was precipitated as plutonium fluoride. The plutonium fraction was also analyzed using a high-resolution alpha spectrometer.

Samples of the resin were also forwarded to the Pacific Northwest National Laboratory for radiochemical analysis of a suite of long-lived radionuclides. These radionuclides included ^{10}Be, ^{36}Cl, ^{93m}Nb, ^{94}Nb, and ^{126}Sn. These radionuclides are not required for measurement by 10 CFR Part 61, but they may be present in significant quantities in some types of low-level radioactive waste. Because the environmental behavior and fate of these radionuclides are not well understood, and because several of them may be quite mobile in the environs, it was felt important to include these radionuclides in the group of 10 CFR Part 61 radionuclides previously measured.

The methods used for the analysis of the long-lived radionuclides present in the spent ion exchange resins are provided in more detail in NUREG/CR-6567.[11] The following summarizes the analysis methods used for these long-lived radionuclides.

Beryllium-10

Beryllium-10 is removed from the acid leach of resins and dissolution of metals by precipitating with NH_4OH, dissolving the precipitate in 6 M HCl and passing the solution through an anion column, which removes most of the transition metals. Beryllium-10 is reprecipitated with NH_4OH and the precipitate dissolved in 0.1 M HCl. Ultima Gold scintillator is added and the ^{10}Be is measured in a Quantulus beta spectrometer. Yields are determined by ^{7}Be tracer.

Chlorine-36

Chlorine-36 is separated from HCl leach of the resin and the metal dissolution by adjusting the solution to 1 M HCl and passing it through a cation resin column. Mercurous nitrate is added to the effluent of the column and Hg_2Cl_2 is precipitated. The precipitate is washed, centrifuged, slurried with water, and transferred to a liquid scintillation vial. The slurry is suspended in Insta-Gel scintillation cocktail and the ^{36}Cl beta measured in a Quantulus liquid scintillation beta spectrometer. Yields are determined using ^{38}Cl tracer.

Niobium-93m and Niobium-94

Niobium-95 tracer, Ni, Sr, Fe, Co, Sb, and Nb carriers are added to an aliquot of acid leachate or dissolution sample. The sample is then made basic with carbonate-free sodium hydroxide, and the resulting precipitate is centrifuged. The precipitate is dissolved in hydrofluoric acid, and Co and Sb carriers are again added. The sample is transferred to a beaker containing concentrated nitric acid and heated with stirring until the volume of acid is reduced to half. The sample is cooled and transferred to a centrifuge tube and the resulting precipitate centrifuged. The precipitate is washed three times with water and then redissolved in HF. The sample is again transferred to a beaker containing concentrated nitric acid and heated with stirring until the volume is reduced to half. The precipitate is then washed twice with water and transferred to a 2-inch diameter planchet and dried. The planchets are covered with two layers of thin Mylar plastic sheets and counted on the Low Energy Photon Spectrometer (LEPS) detector to determine the ^{93m}Nb. The samples are then counted on the gamma detector for 1000 minutes to determine the ^{94}Nb and the ^{95}Nb yield tracer.

Tin-126

An aliquot of each sample is placed in a beaker along with the following hold back carriers: Sn (II), Co, Ni, Nb and Cs. Then, ^{117m}Sn tracer and 2-3 mL of concentrated HCl are added, and the samples are evaporated to dryness. Sulfuric acid and hydrogen peroxide are added, and the samples are taken to fumes. The samples are cooled, water is added, and the samples are chilled in an ice bath. Potassium iodide is then added to each sample, and the tin is extracted twice into toluene. The tin is then back-extracted into a weak sulfuric acid solution, neutralized with sodium hydroxide, and precipitated as the sulfide using thioacetamide. The sulfide precipitate is destroyed with concentrated nitric acid, which results in tin oxide being formed. This oxide is washed with hot water, centrifuged, and slurried with a small amount of water. The samples are first counted on an intrinsic germanium gamma detector to determine the yield of the ^{117m}Sn, a liquid scintillation cocktail is then added, and the samples are counted in the Quantulus liquid scintillation counter to determine the ^{126}Sn content.

Gamma-ray-emitting radionuclides in the leachate samples (e.g., ^{54}Mn, ^{60}Co, and ^{137}Cs) were analyzed using gamma-ray spectrometry. Concentrations of ^{14}C, ^{55}Fe, ^{63}Ni, ^{99}Tc, ^{129}I, and ^{90}Sr in leachate samples were determined by first using radiochemical techniques to selectively extract these radionuclides, then to concentrate them. Carriers and tracers were added to volumetric samples of the leachates, and the samples were then evaporated to dryness. The samples were then dissolved in an HCl solution. The samples were then processed and analyzed using the same procedures used for the resin-waste samples.

Chelating Agent Analysis

The waste-form specimens and leachates were analyzed for EDTA, oxalic acid, and potassium permanganate. A Dionex ion chromatograph equipped with an ion-exchange column was employed. All the various EDTA-metal complexes in the leachates were converted to one species using either one of two transition metals: Ni or Cu. The addition of the strong metal (either Ni or Cu) to the leachate converted all the EDTA-metal complexes to one species that eluted at one time. By systematic study with known reagents, we determined that we could quantify the total EDTA in the leachates by forming either the strong complex with Ni or Cu. Thus, the process was to add an excess of Ni or Cu to the leachates to convert all the EDTA species into the Me-EDTA^{2-} species, where Me is either Ni^{2+} or Cu^{2+}.

Leach-Test Method

A batch leach test method was used to measure the release of radionuclides, transition metals, and chelating agents from the decontamination resin wastes. As part of this process, the Indian Point resins were initially characterized for radionuclide content, some metals, and expected chelating agents. Each resin waste was then leached in a simulated Hanford groundwater for periods of 37 and 93 days. The leach testing was performed at ambient temperatures, and the pH was measured at the beginning and end of the period. The weight of the resin leached was approximately 25 g; the volume of leachant was about 250 ml. The resins were leached in glass beakers with covers to minimize evaporation during the leach test, but the containers were not sealed.

Table 2 shows the composition of the leachant. Following the leach test, the leachates were analyzed for radionuclide content, chelating agents, and ions in solution that may contribute to mobility.

The method used to analyze the data is to determine the quantity of radionuclides, metals, and chelating agent in each resin waste sample and in each leachate to determine the quantitative release rates in terms of fraction per year and pCi/year.

Table 2. Simulated Hanford groundwater formulation (pH=8.9).

Constituent	Concentration (g/L)	Ions in solution
$CaCO_3$	2.624	Ca^+ CO_3^-
HNO_3	0.002	H^+ NO_3^-
KCl	0.269	K^+ Cl^-
$MgCl_2 \cdot 6H_2O$	0.813	Mg^+ Cl^-
$MgCO_3$	0.086	Mg^+ CO_3^-
$Na_2SiO_3 \cdot 9H_2O$	2.967	SiO_3+
H_2SO_4	1.647	H^+ SO_4^{-2}

EXPERIMENTAL RESULTS

Experimental results from the Indian Point-2 wastes can be divided into the radionuclide analysis of the resin and leachates and the chemical analysis of the resins and leachates, including the chelating agents. In this study, waste stream samples were obtained to determine the inventories of radionuclides and chelating agents that would be leached from the waste resins being tested. The radionuclide concentrations and release rates are summarized in the following sections, followed by a summary of the organic and inorganic chemical contents of the resins.

Resin Radionuclide Inventories

Table 3 lists the concentrations of radionuclides present in the Indian Point 2 wastes. In Table 3, the primary decontamination-produced radionuclides are ^{60}Co, ^{59,63}Ni, ^{54}Mn, and ^{65}Zn. Greater than 90% of the activity present is ^{60}Co, with most activity on the cation resin, which is expected to collect the metal wastes from the CAN-DEREM process. The anion and deborating resins have lesser concentrations of activity, with the deborating resin containing the least.

The results indicate the presence of a number of fission products: ^{137}Cs, ^{99}Tc, ^{152,154}Eu, and ^{90}Sr. Some of these radionuclides (i.e., ^{137}Cs, ^{99}Tc) are expected to be relatively mobile and would be expected to be found in the reactor coolant system; however, the ^{152,154}Eu is less mobile and is generally found with fuel material. No ^{235}U was found in the samples; however, relatively large amounts of ^{239}Pu (3.12 E+3 pCi/g) were found on the cation resin, with lesser amounts on the other resins. Some fission of this ^{239}Pu may have contributed to the presence of the relatively insoluble fission products (e.g., ^{152,154}Eu).

Several transuranic radionuclides, including ^{239}Pu, ^{241}Am, ^{242,244}Cm , ^{241}Pu, and ^{238}Pu, were found in the resins. The highest radionuclide concentration was for the ^{241}Pu, at 1.00E+5 pCi/g, in the cation resin. The radionuclide concentrations for the transuranics are 1–2 orders of magnitude higher than those found in previous decontamination ion-exchange resins analyzed as part of this project and summarized in NUREG/CR 6201.

The results of the long-lived radionuclide analyses of the Indian Point-Unit 2 spent ion exchange resins are listed in Table 3. The long-lived neutron activation products ^{10}Be, ^{36}Cl, ^{93m}Nb, ^{94}Nb, and ^{126}Sn were measured in the anion resin and a composite of the cation and deborating resins (the least radioactive samples). Since the enactment of 10 CFR Part 61, other long-lived radionuclides have been identified (e.g., ^{10}Be, ^{36}Cl, ^{93m}Nb, and ^{121m}Sn) that are not routinely measured, but which potentially may be of concern in certain types of LLW.[11] These radionuclides are primarily associated with neutron-activated metal wastes (e.g., spent control rod assemblies and reactor internal components) and other types of LLW (e.g., spent ion exchange cleanup resins) being generated by the nuclear power industry for disposal at shallow-land LLW burial grounds.

Beryllium-10 was measured in the "hot" resin samples, but was not detected in the "composite" resin sample. Beryllium-10 is of special interest because of its potential to be produced in relatively large quantities in nuclear power station LLW, originating in spent BWR and PWR control rods and in spent primary coolant demineralization resins. The ^{10}Be is produced by neutron activation of ^{10}B present as neutron absorbing materials, boron carbide and borosilicate glass in control rods, and as soluble borate added to PWR reactor coolant. The extremely long half-life of ^{10}Be (1.6E6 years), together with a relatively high ingestion dose conversion factor and a relatively low Kd value for adsorption onto sandy types of soil, create the potential for long-term migration and offsite radiation exposure of this radionuclide from LLW disposal facilities

Chlorine-36 was nondetectible in both resin samples. Chlorine-36 is of potential interest because its very long half-life (3.01E5 years), relatively high ingestion dose conversion factor, and high environmental mobility combine to

Table 3. Resin waste inventories.
(pCi/g on 1/3/2000)

Radionuclide	Cation Resin	Unc. (±)	Anion Resin	Unc. (±)	Deborating Resin	Unc. (±)
Alpha Spectrometry						
^{241}Am	6.75E+03	2.33E+02	8.73E+01	4.52E+00	1.08E+00	6.40E-01
^{242}Cm	3.61E+01	9.82E+00	< 8.29E+00		< 9.20E-01	
^{244}Cm	2.12E+03	9.66E+01	2.71E+01	2.61E+00	< 1.65E+00	
^{238}Pu	3.71E+03	1.16E+02	4.84E+01	2.10E+00	< 2.10E+00	
$^{239/240}$Pu	3.12E+03	9.87E+01	3.76E+01	1.79E+00	< 1.18E+00	
^{233,234}U	1.36E+02	1.44E+01	1.84E+01	1.43E+00	9.58E+00	1.01E+00
^{235}U	<[a] 1.43E+01		< 1.06E+00		< 9.64E-01	
^{238}U	< 1.43E+01		< 1.48E+00		< 9.60E-01	
Gamma Spectrometry						
^{108m}Ag	< 3.47E+04		< 5.86E+02		< 1.35E+02	
^{110m}Ag	< 2.61E+04		< 1.00E+03		< 1.06E+02	
^{214}Bi	< 4.87E+04		< 1.78E+03		5.66E+02	9.04E+01
^{144}Ce	9.63E+04	3.82E+04	< 1.55E+04		< 7.11E+02	
^{58}Co	< 3.98E+04		< 1.03E+03		< 1.44E+02	
^{60}Co	7.50E+07	1.16E+06	5.42E+05	6.40E+03	1.01E+03	7.83E+01
^{134}Cs	< 2.29E+04		< 1.05E+03		< 1.82E+02	
^{137}Cs	< 2.91E+04		< 8.82E+02		2.93E+02	4.51E+01
^{152}Eu	4.42E+04	1.51E+04	< 9.12E+02		< 2.89E+02	
^{154}Eu	3.27E+04	9.85E+03	< 5.91E+02		< 1.86E+02	
^{155}Eu	< 2.54E+04		< 6.94E+02		< 1.88E+02	
^{54}Mn	1.02E+05	1.54E+04	< 1.08E+03		< 1.47E+02	
^{94}Nb	< 2.84E+02		8.26E-03	3.22E-04	< 1.24E-04	
^{95}Nb	< 3.65E+04		< 9.50E+02		< 1.34E+02	
^{239}Np	< 3.44E+04		< 8.41E+02		< 2.38E+02	
^{214}Pb	< 8.60E+04		< 1.26E+03		< 5.77E+02	
^{226}Ra	< 3.76E+05		< 9.13E+03		< 2.94E+03	
^{103}Ru	< 1.52E+04		< 6.20E+03		< 7.35E+01	
^{106}Ru	< 2.35E+05		< 7.33E+03		< 9.91E+02	
^{125}Sb	< 2.61E+04		1.59E+05	1.32E+04	5.48E+03	2.91E+02
^{235}U	< 2.30E+04		< 5.58E+02		< 1.79E+02	
^{65}Zn	4.25E+05	2.97E+04	5.70E+03	1.10E+03	< 4.01E+02	
^{95}Zr	< 6.48E+04		< 1.69E+03		< 2.22E+02	
Other radionuclides						
^{129}I	< 1.25E+02		< 1.07E+01		< 1.34E+01	
^{59}Ni	1.12E+04	7.02E+02	1.36E+04	7.12E+02	< 9.41E+01	
^{14}C	1.73E+03	5.59E+01	1.32E+03	3.93E+01	< 2.51E+01	
^{63}Ni	2.19E+06	1.81E+05	3.03E+06	2.50E+05	3.35E+03	2.79E+02
^{241}Pu	1.00E+05	2.42E+03	1.63E+03	3.67E+01	< 1.36E+02	
^{90}Sr	4.35E+03	2.94E+02	3.78E+01	3.58E+00	< 2.47E+01	
^{99}Tc	5.22E+02	2.17E+01	1.22E+02	4.19E+00	<	
^{10}Be	4.31E+01	1.6E0	< 1.37E0			
^{36}Cl	< 2.83E+01		< 6.77E0			
^{93m}Nb	5.90E+02	5.8E+01	1.537E+04	8.0E+04		
^{126}Sn	< 6.49E+01		< 7.07E0			
Weight(g) 1	2.78E+01		2.09E+01		2.24E+01	
Leach Volume 1 (ml)	264.10		252.90		259.10	
Leach Volume 2 (ml)	256.5		248.1		250.2	
RadField(mRem/hr)	1.60E+02		1.60E+01		5.00E-01	

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a. Lower limit of detection is defined as a "<"

create the potential for long-term migration from a LLW disposal facility. Fortunately, the concentrations of ^{36}Cl in LLW from commercial nuclear power stations are quite low. Chlorine-36 is produced almost exclusively in nuclear power plants by neutron activation of soluble chloride impurities present in the reactor coolant water and is then concentrated on primary demineralization resins used for reactor coolant cleanup. Because chloride ions accelerate the corrosion of stainless steel in the reactor primary loop, great care is taken to minimize the chloride concentrations in reactor coolant water, and typical concentrations are in the low ppb range. This is sufficient to produce about 0.0001 to 0.002 uCi/g of ^{36}Cl in spent primary resins. The Indian Point-Unit 2 resins were below these concentrations and indicate good control of chloride concentrations at this nuclear power station.

Tin-126 was nondetectible in both resin samples. Tin-126 is a fission product and is added to reactor coolant water in trace amounts from leaking fuel pins. The nondetectible quantities of this radionuclide in the spent ion exchange resin indicates that the fuel integrity at Indian Point-Unit 2 was quite good.

Niobium-93m was detected in both resin samples. It was surprising to find higher concentrations of ^{93m}Nb in the "composite" resin sample compared to the "hot" resin sample. The reason for this observation is not known. Niobium-93m is produced by the reaction ^{93}Nb (n, n') ^{93m}Nb, by beta decay of ^{93}Zr, and by the electron capture decay of ^{93}Mo. It is, therefore, produced by neutron activation of Nb, Zr, and Mo. Niobium is present as an impurity in stainless steel and some Inconel types at concentrations of 5 to 400 ppm, and is present as an additive in some types of Inconel at 10,000 to 50,000 ppm. Niobium-93m has a half-life of 16.1 years and decays with the emission of 16.5 keV Nb x-rays and a 30 keV gamma-ray. Because two of the ^{93m}Nb parents, ^{93}Mo and ^{93}Zr, have extremely long half-lives, the ^{93m}Nb will remain in some types of neutron-activated metal wastes and spent ion exchange resins for thousands of years.

Leachate Radionuclide Inventories

Table 4 lists the leachate analysis results for the 37- and 93-day leach intervals. The highest concentration of radionuclides was, again, in the anion sample, with the same radionuclides generally detectable in all cases. However, the uranium isotopes, which were not detectable in the resins are detectable in the leachates. This suggests that due to the high activity associated with the resins, dilution and potential interferences may have made detecting these radionuclides difficult in the resins.

Fractional Release Rates

This section summarizes the fractional release rates, for the radionuclides measured in the resins. Table 5 lists the fractional release rates for the radionuclides measured. The fractional release rates for the decontamination radionuclides ^{60}Co, ^{59,63}Ni, ^{54}Mn, and ^{65}Zn range from 3E-3 to 1E-1, or 0.3–10%. This is a fairly high release for these radionuclides and is considerably higher than those expected from cement-solidified waste forms. Typically, the release rates from the anion and deborating resins are higher than those from the cation resin, which may be considered more likely, as the decontamination radionuclides are cations. There appears to be little difference in the release over 37 and 93 days, which suggests that most of the radioactive materials release may occur relatively early in the leach period, with little additional release over the longer term. These data suggest that most of the inventory is retained in the resin and is not subject to significant release until the resin begins to age and deteriorate, which generally occurs relatively rapidly.

The fractional release rates for the fission product radionuclides are generally similar to those for the cations, except for the ^{137}Cs, where essentially all of this radionuclide was released from the deborating resin. Also, about 20% of the ^{125}Sb was released from the deborating resin. The release of most fission product

Table 4. Resin leach results for 37 and 93 days.
(pCi/L on 3/3/2000)

	Cation resin pH not measured Interval = 37 days		Cation resin pH 1.93 Interval = 93 days		Anion resin pH 7.49 Interval = 37 days		Anion resin pH–Not measured Interval = 93 days		Deborating resin pH–9.39 Interval = 37 days		Deborating resin pH–9.61 Interval = 93 days	
Nuclide	pCi/L	Unc. (±)	pCi/L	Unc. (±)	pCi/L	Unc. (±)	pCi/L	Unc. (±)	pCi/L	Unc. (±)	PCi/L	Unc. (±)
^{241}Am	1.92E+01	7.13E+00	1.57E+01	7.21E+00	3.29E+01	9.87E+00	1.59E+01	6.89E+00	< 2.42E+00		< 1.76E+00	
^{242}Cm	< 1.85E+00		< 1.97E+00		< 1.87E+00		< 3.36E+00		< 1.76E+00		< 1.94E+00	
^{244}Cm	4.52E+00	3.64E+00	4.42E+00	3.92E+00	8.82E+00	5.07E+00	< 1.68E+00		< 2.51E+00		< 1.76E+00	
^{238}Pu	9.30E+00	3.06E+00	9.71E+00	3.26E+00	< 4.18E+00		1.01E+01	3.30E+00	< 4.52E+00		< 4.35E+00	
$^{239/240}$Pu	8.18E+00	2.66E+00	9.43E+00	2.89E+00	< 2.56E+00		9.70E+00	2.60E+00	3.31E+00	1.86E+00	< 2.50E+00	
^{233,234}U	6.73E+01	9.60E+00	4.67E+01	6.78E+00	2.75E+01	6.45E+00	5.68E+00	2.59E+00	1.02E+01	2.66E+00	2.02E+01	4.04E+00
^{235}U	3.92E+00	2.23E+00	3.89E+00	1.85E+00	3.47E+00	2.35E+00	< 1.86E+00		< 1.57E+00		1.42E+00	1.31E+00
^{238}U	2.07E+00	1.79E+00	< 1.82E+00		2.41E+00	2.21E+00	< 1.86E+00		< 1.75E+00		1.42E+00	1.31E+00
Gamma Spectrometry												
^{108m}Ag	< 1.97E+04		< 2.24E+04		< 1.34E+03		< 2.34E+03		< 1.67E+02		< 3.04E+02	
^{110m}Ag	< 1.79E+04		< 1.93E+04		< 1.21E+03		< 2.02E+03		< 1.36E+03		< 2.22E+03	
^{214}Bi												
^{144}Ce	< 6.08E+04		< 5.31E+04		< 3.58E+03		< 8.63E+03		< 2.03E+03		< 3.20E+03	
^{58}Co	< 2.42E+04		< 2.75E+04		< 1.78E+03		< 2.89E+03		< 2.23E+02		< 3.42E+02	
^{60}Co	3.14E+07	9.80E+05	2.02E+07	4.93E+05	3.01E+06	7.46E+04	7.40E+05	1.78E+04	3.79E+03	3.45E+02	3.06E+03	3.81E+02
^{134}Cs	< 1.48E+04		< 1.66E+04		2.89E+04	1.92E+03	1.76E+04	2.05E+03	1.98E+03	2.13E+02	2.39E+04	2.10E+03
^{137}Cs	1.51E+04	1.08E+04	2.04E+04	1.62E+04	3.31E+03	3.30E+02	< 2.18E+03		3.71E+04	1.19E+03	4.38E+04	1.54E+03
^{152}Eu	< 4.18E+04		< 3.36E+04		< 3.35E+03		< 4.56E+03		< 7.61E+02		< 1.15E+03	
^{154}Eu	< 3.77E+04		< 3.76E+04		< 3.05E+03		< 4.05E+03		< 4.98E+02		< 7.85E+02	
^{155}Eu	< 2.03E+04		< 2.28E+04		1.14E+03	8.26E+02	< 3.10E+03		< 7.49E+02		< 1.23E+03	
^{54}Mn	1.60E+04	1.07E+04	< 2.70E+04		< 1.66E+03		< 2.69E+03		< 1.95E+02		< 3.07E+02	
^{94}Nb												
^{95}Nb												
^{239}Np												
^{214}Pb												
^{226}Ra	< 2.68E+04		< 2.71E+04		< 1.76E+03		< 3.45E+03		< 9.15E+02		< 1.37E+03	

Table 4. (continued).

	Cation resin pH not measured Interval = 37 days		Cation resin pH 1.93 Interval = 93 days		Anion resin pH 7.49 Interval = 37 days		Anion resin pH–Not measured Interval = 93 days		Deborating resin pH–9.39 Interval = 37 days		Deborating resin pH–9.61 Interval = 93 days	
Nuclide	pCi/L	Unc. (±)	pCi/L	Unc. (±)	pCi/L	Unc. (±)	pCi/L	Unc. (±)	pCi/L	Unc. (±)	PCi/L	Unc. (±)
^{103}Ru	< 1.75E+04		< 1.92E+4		1.14E+03		< 2.08E+03		< 4.24E+02		< 7.32E+02	
^{106}Ru	1.83E+05	1.31E+05	< 1.72E+05		1.08E+04		< 1.82E+04		< 2.79E+03		< 4.17E+03	
^{125}Sb	< 3.75E+04		< 4.00E+04		3.15E+05	4.02E+04	6.91E+04	5.94E+03	9.17E+04	1.19E+04	8.93E+04	1.67E+04
^{235}U	< 6.19E+04		< 5.32E+04		< 3.60E+03		< 8.75E+03		< 2.07E+03		< 3.13E+03	
^{65}Zn	3.05E+05	5.46E+04	2.49E+05	6.38E+04	2.74E+04	3.48E+03	1.68E+04	6.88E+03	< 5.10E+02		< 6.53E+02	
^{95}Zr	< 4.01E+04		< 4.56E+04		< 2.94E+03		< 4.77E+03				< 5.22E+02	
Other radionuclides									< 3.96E+02			
^{129}I												
^{59}Ni	1.42E+04	2.04E+03	5.93E+03	7.22E+02	1.62E+05	2.21E+04	1.67E+04	2.22E+03	< 2.48E+02		< 3.57E+02	
^{14}C	4.21E+03	2.08E+02	5.45E+03	2.28E+02	6.47E+03	2.43E+02	< 2.14E+02		2.59E+02	1.26E+02	2.00E+02	1.25E+02
^{63}Ni	7.68E+05	1.25E+05	5.33E+05	8.73E+04	1.41E+07	2.31E+06	1.51E+06	2.47E+05	2.01E+03	3.62E+02	2.22E+03	3.96E+02
^{241}Pu	5.44E+02	4.52E+01	B	3.91E+01	7.10E+02	5.21E+01	5.13E+02	4.18E+01	3.47E+01	3.71E+00	2.53E+02	3.00E+01
^{90}Sr	5.72E+02	4.92E+01	2.65E+02	2.33E+01	< 1.24E+02		< 9.38E+01		1.58E+01	1.63E+00	5.04E+02	4.24E+01
^{99}Tc	6.05E+01	7.16E+00	1.21E+02	1.41E+01	2.53E+02	2.81E+01	< 1.21E+02		2.49E+01	2.94E+00	< 1.17E+02	

Table 5. Fractional release rates.

Radionuclide	Cation Resin 37 days Fraction Release	Cation Resin 93 days Fraction Release	Anion Resin 37 days Fraction Release	Anion Resin 93 days Fraction Release	Deborating Resin 37 days Fraction Release	Deborating Resin 93 days Fraction Release
Alpha Spectrometry						
^{241}Am	2.67E-05	2.60E-05	4.57E-03	1.74E-03	< 2.59E-02	< 1.76E-02
^{242}Cm	<[a] 4.82E-04	< 6.11E-04	< 2.73E-03	< 3.87E-03	< 2.21E-02	< 2.28E-02
^{244}Cm	2.00E-05	2.33E-05	3.94E-03	< 5.91E-04	< 1.76E-02	< 1.15E-02
^{238}Pu	2.36E-05	2.93E-05	< 1.05E-03	1.99E-03	< 2.49E-02	< 2.24E-02
$^{239/240}$Pu	2.47E-05	3.38E-05	< 8.25E-04	2.46E-03	3.25E-02	< 2.29E-02
^{233,234}U	4.65E-03	3.84E-03	1.81E-02	2.94E-03	1.23E-02	2.28E-02
^{235}U	>[b] 2.58E-03	> 3.04E-03	> 3.97E-02	< 1.67E-02	< 1.89E-02	1.59E-02
^{238}U	> 1.36E-03	< 1.42E-03	> 1.97E-02	< 1.20E-02	< 2.11E-02	1.60E-02
Gamma Spectrometry						
^{108m}Ag	< 5.34E-03	< 7.22E-03	< 2.77E-02	< 3.81E-02	< 1.43E-02	< 2.43E-02
^{110m}Ag	< 6.45E-03	< 8.28E-03	< 1.47E-02	< 1.93E-02	< 1.49E-01	< 2.26E-01
^{144}Ce	< 5.94E-03	< 6.17E-03	< 2.80E-03	< 5.31E-03	< 3.31E-02	< 4.86E-02
^{58}Co	< 5.72E-03	< 7.73E-03	< 2.09E-02	< 2.68E-02	< 1.79E-02	< 2.56E-02
^{60}Co	3.94E-03	3.01E-03	6.73E-02	1.30E-02	4.34E-02	3.27E-02
^{134}Cs	< 6.08E-03	< 8.11E-03	> 3.34E-01	> 1.60E-01	1.26E-01	1.42E+00[b]
^{137}Cs	4.88E-03	> 7.85E-03	> 4.55E-02	< 2.36E-02	1.47E+00[b]	1.61E+00[b]
^{152}Eu	< 8.89E-03	< 8.51E-03	< 4.45E-02	< 4.77E-02	< 3.05E-02	< 4.30E-02
^{154}Eu	< 1.08E-02	< 1.29E-02	< 6.25E-02	< 6.54E-02	< 3.10E-02	< 4.56E-02
^{155}Eu	< 7.51E-03	< 1.00E-02	> 1.99E-02	< 4.26E-02	< 4.61E-02	< 7.06E-02
^{54}Mn	1.47E-03	< 2.96E-03	< 1.86E-02	< 2.38E-02	< 1.54E-02	< 2.26E-02
^{226}Ra	< 6.70E-04	< 8.07E-04	< 2.34E-03	< 3.60E-03	< 3.60E-03	< 5.03E-03
^{103}Ru	< 1.08E-02	< 1.41E-02	> 2.23E-03	< 3.20E-03	< 6.68E-02	< 1.08E-01
^{106}Ru	< 7.32E-03	< 8.19E-03	> 1.79E-02	< 2.37E-02	< 3.26E-02	< 4.54E-02
^{125}Sb	< 1.35E-02	< 1.72E-02	2.40E-02	4.15E-03	1.94E-01	1.76E-01
^{235}U	< 2.53E-02	< 2.59E-02	< 7.82E-02	< 1.50E-01	< 1.34E-01	< 1.89E-01
^{65}Zn	6.75E-03	6.56E-03	5.83E-02	2.81E-02	< 1.47E-02	< 1.76E-02
^{95}Zr	< 5.82E-03	< 7.88E-03	< 2.11E-02	< 2.69E-02		< 2.54E-02
Other radionuclides						
^{129}I						
^{59}Ni	1.19E-02	5.93E-03	1.44E-01	1.17E-02	< 3.05E-02	< 4.10E-02
^{14}C	2.29E-02	3.53E-02	5.94E-02	< 1.55E-03	1.19E-01	8.60E-02
^{63}Ni	3.30E-03	2.72E-03	5.64E-02	4.75E-03	6.95E-03	7.16E-03
^{241}Pu	5.11E-05		5.28E-03	3.00E-03	2.95E-03	2.01E-02
^{90}Sr	1.24E-03	6.82E-04	< 3.98E-02	< 2.37E-02	7.41E-03	2.20E-01
^{99}Tc	1.09E-03	2.59E-03	2.51E-02	< 9.46E-03		

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a. Lower limit of detection is defined as a "<"

b. Resin concentration is below the Minimum Detectable Activity. Consequently if calculating a release rate with a measurable leach concentration the fractional release becomes greater than the listed value

c. Possible contamination of sample

radionuclides is typically on the order of 1% over the period.

The transuranic radionuclides have fractional release rates that are typically similar to those for the decontamination radionuclides, with releases on the order of 1%, and there appears to be little difference in the release over 37 and 93 days, which suggests that most of the release occurs relatively early in the leach period. As noted previously, the radionuclide concentrations for the transuranics are 1–2 orders of magnitude higher than those found in previous decontamination ion-exchange resins analyzed as part of this project and summarized in NUREG/CR 6201. The release rates for these radionuclides are generally much lower than the fractional release for fission products and neutron activation products.

Comparisons with the results of other INEEL studies summarized in NUREG/CR 6201, would suggest that the fractional release rates of uranium and the transuranics are considerably higher than those that would be expected. No external references have been identified that would indicate that these release rates are expected for resin wastes.

Annual Release Rates

Tables 6 and 7 list the annual release rates from the resin wastes in pCi/yr/g and pCi/yr of resin. In both cases, the release rates are highest for the decontamination radionuclides (i.e., ^{60}Co, ^{59,63}Ni, ^{54}Mn, and ^{65}Zn) with ^{60}Co the highest at about 2.87 E+6 uCi/yr/g or 1.03E+5 pCi/yr for the cation resin. This is a relatively significant release rate, although the fractional release is only about 0.4%. The release of the transuranics and fission products are generally similar, with releases on the order of 10E-3 to 10E-2 pCi/yr/g. These releases seem higher than expected.

Organic and Inorganic Leachate Composition

This section presents the results of the chelating agent analysis of the leachates, along with the results of analyses of the leachate elemental analysis results. Table 8 shows the ICP data for the water leachates from contacting the spent resins at a solid-resin-to-simulated-groundwater ratio of about 1:10 [g:mL]. The common cations come from both the simulated groundwater and from reagents used to make up the various lixiviants pumped into the reactor piping system, such as K from the alkaline permanganate, sodium from pH adjustment, and Mn from the permanganate. The transition metals (Cr, Ni, Fe) are corrosion products that dissolve out of the cooling system during the decontamination process. As shown in Table 8, cation resin leachate contains significantly higher concentrations of most of the transition metals associated with corrosion products. Despite containing much higher radionuclide activities, the leachate from the deborating resin does not contain the highest concentrations of corrosion product-derived transition metals. In general, the leachates from anion resin contain the lowest concentrations of transition metals.

Table 9 shows the leachate inorganic anion compositions, as determined by ion chromatography. The leachates for anion resin are dominated by bicarbonate, likely from dissolution of carbon dioxide into the AP fluid during the destruction of oxalic acid used to reduce the MnO_2 formed during the permanganate oxidation step. The highly acidic leachate from contact of simulated groundwater with deborating resin is dominated by sulfate and chloride, but the source is not known.

Table 10 lists the data for organic ligands in the leachates, as measured by ion chromatography. We were surprised that none of the leachates showed evidence of citric acid, which is supposed to be present in most CAN-DEREM recipes along with EDTA. We also did not find any evidence of the residual oxalic acid used to dissolve the MnO_2 solids that form when the permanganate is used up to oxidize the Cr(III) oxide films in the pipes. According to the Westinghouse proprietary report, most of the oxalic acid is decomposed to carbon dioxide during the dissolution of the MnO_2. The small concentrations of formic acid in the deborating spent resin is likely degradation products of the oxalic acid that have not been converted to carbon dioxide. Only the cation resin leachate contained measurable concentrations of EDTA. The concentration [70.1 mg/L] is equivalent to 2.4×10^{-4} M. This value was confirmed within ±20% by independent spiking of aliquots of this leachate with Ni^{2+} or Cu^{2+}. The EDTA is expected to be the primary chelating agent present and appears to be relatively well destroyed during the CAN-DEREM process.

Table 6. pCi/yr/g release rates.

Radionuclide	Cation Resin 37 days	Cation Resin 93 days	Anion Resin 37 days	Anion Resin 93 days	Deborating Resin 37 days	Deborating Resin 93 days
Alpha Spectrometry						
^{241}Am	1.76E+00	6.80E-01	3.88E+00	5.92E-01	< 9.59E-02	< 7.35E-02
^{242}Cm	< 1.69E-01	< 8.53E-02	< 2.20E-01	< 1.25E-01	< 4.52E-01	< 8.11E-02
^{244}Cm	4.13E-01	1.91E-01	1.04E+00	< 6.25E-02	< 2.14E-01	< 7.36E-02
^{238}Pu	8.51E-01	4.21E-01	< 4.93E-01	3.76E-01	< 1.31E-01	< 1.82E-01
$^{239/240}$Pu	7.48E-01	4.09E-01	< 3.02E-01	3.61E-01	1.41E+00	< 1.04E-01
^{233,234}U	6.16E+00	2.02E+00	3.24E+00	2.11E-01	1.78E-01	8.44E-01
^{235}U	3.59E-01	1.69E-01	4.09E-01	< 6.92E-02	< 1.24E-01	5.94E-02
^{238}U	1.89E-01	< 7.88E-02	2.84E-01	< 6.92E-02		5.94E-02
Gamma Spectrometry						
^{108m}Ag	< 1.80E+03	< 9.70E+02	< 1.58E+02	< 8.71E+01	< 6.20E+01	< 1.27E+01
^{110m}Ag	< 1.64E+03	< 8.36E+02	< 1.43E+02	< 7.52E+01		< 9.28E+02
^{144}Ce	< 5.56E+03	< 2.30E+03	< 4.22E+02	< 3.21E+02	< 9.12E+01	< 1.34E+02
^{58}Co	< 2.21E+03	< 1.19E+03	< 2.10E+02	< 1.08E+02	< 1.54E+05	< 1.43E+02
^{60}Co	2.87E+06	8.75E+05	3.55E+05	2.75E+04	1.48E+03	1.28E+02
^{134}Cs	< 1.35E+03	< 7.19E+02	3.41E+03	> 6.55E+02	1.70E+02	9.99E+02
^{137}Cs	1.38E+03	8.84E+02	3.90E+02	< 8.11E+01	1.72E+02	1.83E+03
^{152}Eu	< 3.82E+03	< 1.46E+03	< 3.95E+02	< 1.70E+02	< 1.56E+02	< 4..81E+02
^{154}Eu	< 3.45E+03	< 1.63E+03	< 3.60E+02	< 1.51E+02	< 5.84E+01	< 3.28E+01
^{155}Eu	< 1.86E+03	< 9.88E+02	1.34E+02	< 1.15E+02	< 8.51E+01	< 5.14E+01
^{54}Mn	1.46E+03	< 1.17E+03	< 1.96E+02	< 1.00E+02		< 1.28E+01
^{226}Ra	< 2.45E+03	< 1.17E+03	< 2.08E+02	< 1.28E+02	< 5.84E+01	< 5.72E+01
^{103}Ru	< 1.60E+03	< 8.32E+02	1.34E+02	< 7.74E+01	< 5.54E+02	< 3.06E+01
^{106}Ru	1.67E+04	< 7.45E+03	1.27E+03	< 6.77E+02	< 1.61E+04	< 1.74E+02
^{125}Sb	< 3.43E+03	< 1.73E+03	3.71E+04	2.57E+03	1.85E+02	3.73E+03
^{235}U	< 5.66E+03	< 2.30E+03	< 4.24E+02	< 3.26E+02	< 1.40E+03	< 1.30E+02
^{65}Zn	2.79E+04	1.08E+04	3.23E+03	6.25E+02	< 1.51E+02	< 2.73E+01
^{95}Zr	< 3.67E+03	< 1.98E+03	< 3.47E+02	< 1.78E+02		< 2.18E+01
Other radionuclides						
^{59}Ni	1.30E+03	2.57E+02	1.91E+04	6.22E+02	< 3.32E+02	< 1.49E+01
^{14}C	3.85E+02	2.36E+02	7.63E+02	< 7.97E+00	7.23E+05	8.36E+00
^{63}Ni	7.03E+04	2.31E+04	1.66E+06	5.62E+04	3.64E+01	9.28E+01
^{241}Pu	4.98E+01		8.37E+01	1.91E+01	6.36E+00	1.06E+01
^{90}Sr	5.23E+01	1.15E+01	< 1.46E+01	< 3.49E+00	1.30E+01	2.11E+01
^{99}Tc	5.53E+00	5.24E+00	2.98E+01	< 4.50E+00		

Table 7. pCi/yr release rates.

Radionuclide	Cation Resin 37 days	Cation Resin 93 days	Anion Resin 37 days	Anion Resin 93 days	Deborating Resin 37 days	Deborating Resin 93 days
Alpha Spectrometry						
^{241}Am	6.34E-02	2.97E-02	1.86E-01	2.29E-02	< 4.29E-03	< 3.17E-03
^{242}Cm	< 6.09E-03	< 3.72E-03	< 1.05E-02	< 4.84E-03	< 2.02E-02	< 3.50E-03
^{244}Cm	1.49E-02	8.33E-03	4.98E-02	< 2.42E-03	< 9.56E-03	< 3.18E-03
^{238}Pu	3.07E-02	1.84E-02	< 2.36E-02	1.46E-02	< 5.85E-03	< 7.85E-03
$^{239/240}$Pu	2.69E-02	1.78E-02	< 1.45E-02	1.40E-02	6.30E-02	< 4.49E-03
^{233,234}U	2.22E-01	8.81E-02	1.55E-01	8.18E-03	7.95E-03	3.64E-02
^{235}U	1.29E-02	7.37E-03	1.96E-02	< 2.68E-03	< 5.54E-03	2.56E-03
^{238}U	6.81E-03	< 3.44E-03	1.36E-02	< 2.68E-03		2.56E-03
^{108m}Ag	< 6.48E+01	< 4.23E+01	< 7.57E+00	< 3.38E+00	< 2.77E+00	< 5.48E-01
^{110m}Ag	< 5.91E+01	< 3.65E+01	< 6.85E+00	< 2.91E+00		< 4.01E+01
^{144}Ce	< 2.00E+02	< 1.00E+02	< 2.02E+01	< 1.24E+01	< 4.08E+00	< 5.78E+00
^{58}Co	< 7.96E+01	< 5.19E+01	< 1.01E+01	< 4.19E+00	< 6.88E+03	< 6.17E+00
^{60}Co	1.03E+05	3.82E+04	1.70E+04	1.07E+03	6.61E+01	5.52E+00
^{134}Cs	< 4.86E+01	< 3.14E+01	1.63E+02	> 2.54E+01	7.60E+00	4.31E+01
^{137}Cs	4.97E+01	3.86E+01	1.87E+01	< 3.14E+00	7.69E+00	7.90E+01
^{152}Eu	< 1.38E+02	< 6.37E+01	< 1.89E+01	< 6.59E+00	< 6.97E+00	<
^{154}Eu	< 1.24E+02	< 7.11E+01	< 1.72E+01	< 5.85E+00	< 2.61E+00	< 1.42E+00
^{155}Eu	< 6.70E+01	< 4.31E+01	6.42E+00	< 4.46E+00	< 3.80E+00	< 2.22E+00
^{54}Mn	5.26E+01	< 5.10E+01	< 9.39E+00	< 3.88E+00		< 5.52E-01
^{226}Ra	< 8.83E+01	< 5.10E+01	< 9.97E+00	< 4.96E+00	< 2.61E+00	< 2.47E+00
^{103}Ru	< 5.76E+01	< 3.63E+01	6.42E+00	< 3.00E+00	< 2.48E+01	< 1.32E+00
^{106}Ru	6.02E+02	< 3.25E+02	6.09E+01	< 2.62E+01	< 7.19E+02	< 7.51E+00
^{125}Sb	< 1.24E+02	< 7.55E+01	1.78E+03	9.96E+01	8.27E+00	1.61E+02
^{235}U	< 2.04E+02	< 1.00E+02	< 2.03E+01	< 1.26E+01	< 6.26E+01	< 5.61E+00
^{65}Zn	1.01E+03	4.71E+02	1.55E+02	2.42E+01	< 6.75E+00	< 1.18E+00
^{95}Zr	< 1.32E+02	< 8.64E+01	< 1.66E+01	< 6.90E+00		< 9.41E-01
^{59}Ni	4.68E+01	1.12E+01	9.15E+02	2.41E+01	< 1.48E+01	< 6.43E-01
^{14}C	1.39E+01	1.03E+01	3.66E+01	< 3.09E-01	3.23E+04	3.61E-01
^{63}Ni	2.53E+03	1.01E+03	7.95E+04	2.18E+03	1.63E+00	4.01E+00
^{241}Pu	1.79E+00		4.01E+00	7.40E-01	2.84E-01	4.57E-01
^{90}Sr	1.88E+00	5.02E-01	< 7.00E-01	< 1.35E-01	5.81E-01	9.11E-01
^{99}Tc	1.99E-01	2.29E-01	1.43E+00	< 1.74E-01		

Table 8. Leachate concentrations (ug/L) for cations and transition metals.

Sample	Anion Resin	Deborating Resin	Cation Resin	Anion Resin	Deborating resin	Cation Resin
Time (d)	37	37	37	93	93	93
Element						
Al	<250	239	—[a]	—[a]	<250	212
Ba	120	120	—	—	79	41
Ca	11965	115860	—	—	181547	17413
Cd	<250	<250	—	—	<250	<250
Co	23	12	—	—	15	27
Cr	5796	12	—	—	0	23
Cu	76	0	—	—	16	17
Fe	549	53	—	—	346	335
K	27200	1413	—	—	1354	1407
Mg	4554	1390	—	—	1115	541
Mn	13507	<1125	—	—	<1125	3015
Ni	7516	10	—	—	16	510
Pb	76	211	—	—	<250	109
Sr	12	68	—	—	90	28
Zn	382	221	—	—	198	359
Na	119249	430998	—	—	448721	92951
Si	61394	2065	—	—	5465	74379
Zr	1	2	—	—	69	62

a. Column not analyzed because of insufficient sample.

Table 9. Leachate concentrations (mg/L) for inorganic anions.

Sample	Anion Resin	Deborating Resin	Cation Resin	Anion Resin	Deborating resin	Cation Resin
Leach period (d)	37	37	37	93	93	93
Species						
Cl	167.5	5.0	—[a]	—[a]	4.4	646
SO_4	55.5	1.72	—	—	<1	1760
NO_3	<1	<1	—	—	<1	3.57
PO_4	<1	<1	—	—	<1	0.3
HCO_3	<25	1500	—	—	1750	<1

a. Column not analyzed because of insufficient sample.

Table 10. Leachate concentrations (mg/L) for organic ligands.

Sample	Anion Resin	Deborating Resin	Cation Resin	Anion Resin	Deborating resin	Cation Resin
Leach period (d)	37	37	37	93	93	93
Species						
Formate	<1	9.51	—[a]	—[a]	17.3	<1
Oxalate	<3	<3	—	—	<3	<3
Citrate	<1	<1	—	—	<1	<1
EDTA	70.1	<1	—	—	<1	<1

a. Column not analyzed because of insufficient sample.

SUMMARY

This research evaluated the radionuclide inventories, leaching results, release fractions, and annual releases of the chelating agents and radionuclides from full-system decontamination when subjected to a simulated Hanford groundwater. Plant personnel collected untreated ion-exchange resin wastes from resin storage tanks following decontamination of the reactor vessel at Indian Point 2, March 1995. This was the first decontamination of an actual full reactor vessel from which the fuel had been removed. The resins analyzed were cation, anion, and what appears to be a deborating resin.

Several transuranic radionuclides, including ^{239}Pu, ^{241}Am, ^{242,244}Cm, ^{241}Pu, and ^{238}Pu, were found in the resins. The highest radionuclide concentration was for the ^{241}Pu, at 1.00E+5 pCi/g, in the cation resin. The radionuclide concentrations for the transuranics are 1–2 orders of magnitude higher than those found in previous decontamination ion-exchange resins analyzed as part of this project and summarized in NUREG/CR 6201.

The long-lived neutron activation products ^{10}Be, ^{36}Cl, ^{93m}Nb, ^{94}Nb, and ^{126}Sn were measured in the anion resin and a composite of the cation and deborating resins (the least radioactive samples). Since the enactment of 10 CFR Part 61, other long-lived radionuclides have been identified (e.g., ^{10}Be, ^{36}Cl, ^{93m}Nb, and ^{121m}Sn) which are not routinely measured, but which potentially may be of concern in certain types of LLW.[11] These radionuclides are primarily associated with neutron-activated metal wastes (e.g., spent control rod assemblies and reactor internal components) and other types of LLW (e.g., spent ion exchange cleanup resins) being generated by the nuclear power industry for disposal at shallow-land LLW burial grounds. ^{10}Be is produced by neutron activation of ^{10}B present as neutron absorbing materials, boron carbide and borosilicate glass in control rods, and as soluble borate added to PWR reactor coolant. It was detected in both samples whereas ^{36}Cl and ^{126}Sn were not detected in either sample which suggests that the reactor coolant system at Indian Point has little chloride contamination or breakdown of tin containing components.

Niobium-93m was detected in both resin samples at relatively high concentrations. Niobium is present as an impurity in stainless steel and some Inconel types at concentrations of 5 to 400 ppm, and is present as an additive in some types of Inconel at 10,000 to 50,000 ppm. Although Niobium-93m has a half-life of 16.1 years, ^{94}Nb has a half-life of 20,000 years. The high concentration of the ^{93m}Nb, would suggest relatively high concentrations of the ^{94}Nb. Because two of the ^{93m}Nb parents, ^{93}Mo and ^{93}Zr, also have extremely long half-lives, the ^{93m}Nb will remain in some types of neutron-activated metal wastes and spent ion exchange resins for thousands of years.

Primary conclusions of this evaluation are that the fractional release rates for the decontamination radionuclides ^{60}Co, ^{59,63}Ni, ^{54}Mn, and ^{65}Zn range from 3E-3 to 1E-1, or 0.3–10%. This is a fairly highly release for these radionuclides and is considerably higher than those expected from cement solidified waste forms. Typically, the release rates from the anion and deborating resins are higher than those from the cation resin, which may be considered more likely because the decontamination radionuclides are cations. The higher than expected fractional releases for the uranium radionuclides and the transuranics are significant as they would suggest relatively high releases.

There appears to be little difference in the release over 37 and 93 days, which suggests that most of the radioactive materials release may occur relatively early in the leach period, with little additional release over the longer term. These data suggest that most of the inventory is retained in the resin and is not subject to significant release until the resin begins to age and deteriorate, which generally occurs relatively rapidly.

Despite containing much higher radionuclide activities, the leachate from the cation resin does not contain the highest concentrations of corrosion product-derived transition metals; the anion resin leachate contains the most. And, in general, the leachates from the deborating resin contain the lowest concentrations of transition metals. This may be likely, as the cation resin may bind the cation decontamination elements more tightly than does the anion or deborating resin.

Only the cation resin leachate contained measurable concentrations of EDTA. The concentration [70.1 mg/L] is equivalent to 2.4×10^{-4} M. EDTA is expected to be the primary chelating agent present and appears to be relatively well destroyed during the CAN-DEREM process with only a low concentration remaining in the decontamination waste. This suggests that potential chelating agent mobility of radionuclides may be relatively limited due to the small amount of EDTA present. The leach rate for EDTA was determined to be $1.4\ E^{-5}$ mole/year.

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Appendix A

Resin Leach Rate Data

Table A-1. Cation resin leach results (37 days).

Radionuclide	pCi inventory 27.76 g	IP3-37 Cation Resin 264.1 mL	Fraction Release	pCi/yr/g resin
Alpha Spectrometry				
^{241}Am	1.87E+05	5.01E+00	2.67E-05	1.76E+00
^{242}Cm	1.00E+03	< 4.83E-01	< 4.82E-04	< 1.69E-01
^{244}Cm	5.89E+04	1.18E+00	2.00E-05	4.13E-01
^{238}Pu	1.03E+05	2.43E+00	2.36E-05	8.51E-01
$^{239/240}$Pu	8.66E+04	2.13E+00	2.47E-05	7.48E-01
^{233,234}U	3.78E+03	1.76E+01	4.65E-03	6.16E+00
^{235}U	< 3.97E+02	1.02E+00	> 2.58E-03	3.59E-01
^{238}U	< 3.97E+02	5.40E-01	> 1.36E-03	1.89E-01
Gamma Spectrometry				
^{108m}Ag	< 9.63E+05	< 5.14E+03	< 5.34E-03	< 1.80E+03
^{110m}Ag	< 7.25E+05	< 4.67E+03	< 6.45E-03	< 1.64E+03
^{214}Bi	< 1.35E+06			
^{144}Ce	2.67E+06	< 1.59E+04	< 5.94E-03	< 5.56E+03
^{58}Co	< 1.10E+06	< 6.32E+03	< 5.72E-03	< 2.21E+03
^{60}Co	2.08E+09	8.20E+06	3.94E-03	2.87E+06
^{134}Cs	< 6.36E+05	< 3.86E+03	< 6.08E-03	< 1.35E+03
^{137}Cs	< 8.08E+05	3.94E+03	4.88E-03	1.38E+03
^{152}Eu	1.23E+06	< 1.09E+04	< 8.89E-03	< 3.82E+03
^{154}Eu	9.08E+05	< 9.84E+03	< 1.08E-02	< 3.45E+03
^{155}Eu	< 7.05E+05	< 5.30E+03	< 7.51E-03	< 1.86E+03
^{54}Mn	2.83E+06	4.18E+03	1.47E-03	1.46E+03
^{94}Nb	< 7.88E+03			
^{95}Nb	< 1.01E+06			
^{239}Np	< 9.55E+05			
^{214}Pb	< 2.39E+06			
^{226}Ra	< 1.04E+07	< 6.99E+03	< 6.70E-04	< 2.45E+03
^{103}Ru	< 4.22E+05	< 4.57E+03	< 1.08E-02	< 1.60E+03
^{106}Ru	< 6.52E+06	4.78E+04	< 7.32E-03	1.67E+04
^{125}Sb	< 7.25E+05	< 9.79E+03	< 1.35E-02	< 3.43E+03
^{235}U	< 6.38E+05	< 1.62E+04	< 2.53E-02	< 5.66E+03
^{65}Zn	1.18E+07	7.96E+04	6.75E-03	2.79E+04
^{95}Zr	< 1.80E+06	< 1.05E+04	< 5.82E-03	< 3.67E+03

Table A-1. (continued).

Radionuclide		pCi inventory 27.76 g	IP3-37 Cation Resin 264.1 mL	Fraction Release	pCi/yr/g resin
Other radionuclides					
^{129}I	<	3.47E+03			
^{59}Ni		3.11E+05	3.71E+03	1.19E-02	1.30E+03
^{14}C		4.80E+04	1.10E+03	2.29E-02	3.85E+02
^{63}Ni		6.08E+07	2.00E+05	3.30E-03	7.03E+04
^{241}Pu		2.78E+06	1.42E+02	5.11E-05	4.98E+01
^{90}Sr		1.21E+05	1.49E+02	1.24E-03	5.23E+01
^{99}Tc		1.45E+04	1.58E+01	1.09E-03	5.53E+00

Table A-2. Cation resin leach results (93 days).

Radionuclide	Cation pCi inventory 22.92 g	IP3-93 Cation Resin 256.5 mL	Fraction Release	pCi/yr/g resin
Alpha Spectrometry				
^{241}Am	1.55E+05	4.03E+00	2.60E-05	6.80E-01
^{242}Cm	8.27E+02	< 5.05E-01	< 6.11E-04	< 8.53E-02
^{244}Cm	4.86E+04	1.13E+00	2.33E-05	1.91E-01
^{238}Pu	8.50E+04	2.49E+00	2.93E-05	4.21E-01
$^{239/240}$Pu	7.15E+04	2.42E+00	3.38E-05	4.09E-01
^{233,234}U	3.12E+03	1.20E+01	3.84E-03	2.02E+00
^{235}U	< 3.28E+02	9.98E-01	> 3.04E-03	1.69E-01
^{238}U	< 3.28E+02	< 4.67E-01	< 1.42E-03	< 7.88E-02
Gamma Spectrometry				
^{108m}Ag	< 7.95E+05	< 5.75E+03	< 7.22E-03	< 9.70E+02
^{110m}Ag	< 5.98E+05	< 4.95E+03	< 8.28E-03	< 8.36E+02
^{214}Bi	< 1.12E+06			
^{144}Ce	2.21E+06	< 1.36E+04	< 6.17E-03	< 2.30E+03
^{58}Co	< 9.12E+05	< 7.05E+03	< 7.73E-03	< 1.19E+03
^{60}Co	1.72E+09	5.18E+06	3.01E-03	8.75E+05
^{134}Cs	< 5.25E+05	< 4.26E+03	< 8.11E-03	< 7.19E+02
^{137}Cs	< 6.67E+05	5.23E+03	> 7.85E-03	8.84E+02
^{152}Eu	1.01E+06	< 8.62E+03	< 8.51E-03	< 1.46E+03
^{154}Eu	7.49E+05	< 9.64E+03	< 1.29E-02	< 1.63E+03
^{155}Eu	< 5.82E+05	< 5.85E+03	< 1.00E-02	< 9.88E+02
^{54}Mn	2.34E+06	< 6.93E+03	< 2.96E-03	< 1.17E+03
^{94}Nb	< 6.51E+03			
^{95}Nb	< 8.37E+05			
^{239}Np	< 7.88E+05			
^{214}Pb	< 1.97E+06			
^{226}Ra	< 8.62E+06	< 6.95E+03	< 8.07E-04	< 1.17E+03
^{103}Ru	< 3.48E+05	< 4.92E+03	< 1.41E-02	< 8.32E+02
^{106}Ru	< 5.39E+06	< 4.41E+04	< 8.19E-03	< 7.45E+03
^{125}Sb	< 5.98E+05	< 1.03E+04	< 1.72E-02	< 1.73E+03
^{235}U	< 5.27E+05	< 1.36E+04	< 2.59E-02	< 2.30E+03
^{65}Zn	9.74E+06	6.39E+04	6.56E-03	1.08E+04
^{95}Zr	< 1.49E+06	< 1.17E+04	< 7.88E-03	< 1.98E+03

Table A-2. (continued).

Radionuclide		Cation pCi inventory 22.92 g	IP3-93 Cation Resin 256.5 mL	Fraction Release	pCi/yr/g resin
Other radionuclides					
^{129}I	<	2.87E+03			
^{59}Ni		2.57E+05	1.52E+03	5.93E-03	2.57E+02
^{14}C		3.97E+04	1.40E+03	3.53E-02	2.36E+02
^{63}Ni		5.02E+07	1.37E+05	2.72E-03	2.31E+04
^{241}Pu		2.29E+06	0.00E+00	0.00E+00	
^{90}Sr		9.97E+04	6.80E+01	6.82E-04	1.15E+01
^{99}Tc		1.20E+04	3.10E+01	2.59E-03	5.24E+00

Table A-3. Anion resin leach results (37 days).

Nuclide		#487022-46 anion 20.87 g		IP1 37 #487022-46 anion 252.9mL		Fraction Release		pCi/yr/g resin
Alpha Spectrometry								
^{241}Am		1.82E+03		8.32E+00		4.57E-03		3.88E+00
^{242}Cm	<	1.73E+02	<	4.73E-01	<	2.73E-03	<	2.20E-01
^{244}Cm		5.66E+02		2.23E+00		3.94E-03		1.04E+00
^{238}Pu		1.01E+03	<	1.06E+00	<	1.05E-03	<	4.93E-01
$^{239/240}$Pu		7.85E+02	<	6.47E-01	<	8.25E-04	<	3.02E-01
$^{233.234}$U		3.84E+02		6.95E+00		1.81E-02		3.24E+00
^{235}U	<	2.21E+01		8.78E-01	>	3.97E-02		4.09E-01
^{238}U	<	3.09E+01		6.09E-01	>	1.97E-02		2.84E-01
Gamma Spectrometry								0.00E+00
^{108m}Ag	<	1.22E+04	<	3.39E+02	<	2.77E-02	<	1.58E+02
^{110m}Ag	<	2.09E+04	<	3.06E+02	<	1.47E-02	<	1.43E+02
^{214}Bi	<	3.71E+04						0.00E+00
^{144}Ce	<	3.23E+05	<	9.05E+02	<	2.80E-03	<	4.22E+02
^{58}Co	<	2.15E+04	<	4.50E+02	<	2.09E-02	<	2.10E+02
^{60}Co		1.13E+07		7.61E+05		6.73E-02		3.55E+05
^{134}Cs	<	2.19E+04		7.31E+03	>	3.34E-01		3.41E+03
^{137}Cs	<	1.84E+04		8.37E+02	>	4.55E-02		3.90E+02
^{152}Eu	<	1.90E+04	<	8.47E+02	<	4.45E-02	<	3.95E+02
^{154}Eu	<	1.23E+04	<	7.71E+02	<	6.25E-02	<	3.60E+02
^{155}Eu	<	1.45E+04		2.88E+02	>	1.99E-02		1.34E+02
^{54}Mn	<	2.25E+04	<	4.20E+02	<	1.86E-02	<	1.96E+02
^{94}Nb	<	1.72E-01						
^{95}Nb	<	1.98E+04						
^{239}Np	<	1.76E+04						
^{214}Pb	<	2.63E+04						
^{226}Ra	<	1.91E+05	<	4.45E+02	<	2.34E-03	<	2.08E+02
^{103}Ru	<	1.29E+05		2.88E+02	>	2.23E-03		1.34E+02
^{106}Ru	<	1.53E+05		2.73E+03	>	1.79E-02		1.27E+03
^{125}Sb		3.32E+06		7.97E+04		2.40E-02		3.71E+04
^{235}U	<	1.16E+04	<	9.10E+02	<	7.82E-02	<	4.24E+02
^{65}Zn		1.19E+05		6.93E+03		5.83E-02		3.23E+03
^{95}Zr	<	3.53E+04	<	7.44E+02	<	2.11E-02	<	3.47E+02

Table A-3. (continued).

Nuclide	#487022-46 anion 20.87 g	IP1 37 #487022-46 anion 252.9mL	Fraction Release	pCi/yr/g resin
Other radionuclides				
^{129}I	< 2.23E+02			
^{59}Ni	2.84E+05	4.10E+04	1.44E-01	1.91E+04
^{14}C	2.75E+04	1.64E+03	5.94E-02	7.63E+02
^{63}Ni	6.32E+07	3.57E+06	5.64E-02	1.66E+06
^{241}Pu	3.40E+04	1.80E+02	5.28E-03	8.37E+01
^{90}Sr	7.89E+02	< 3.14E+01	< 3.98E-02	< 1.46E+01
^{99}Tc	2.55E+03	6.40E+01	2.51E-02	2.98E+01

Table A-4. Anion resin leach results (93 days).

Nuclide	#487022-46 anion 25.8 g	IP1-93 #487022-46 anion 248.1 mL	Fraction Release	pCi/yr/g resin
Alpha Spectrometry				
^{241}Am	2.27E+03	3.94E+00	1.74E-03	5.92E-01
^{242}Cm	< 2.16E+02	< 8.34E-01	< 3.87E-03	< 1.25E-01
^{244}Cm	7.05E+02	< 4.17E-01	< 5.91E-04	< 6.25E-02
^{238}Pu	1.26E+03	2.51E+00	1.99E-03	3.76E-01
$^{239/240}$Pu	9.78E+02	2.41E+00	2.46E-03	3.61E-01
^{233,234}U	4.79E+02	1.41E+00	2.94E-03	2.11E-01
^{235}U	< 2.76E+01	< 4.61E-01	< 1.67E-02	< 6.92E-02
^{238}U	< 3.85E+01	< 4.61E-01	< 1.20E-02	< 6.92E-02
Gamma Spectrometry				0.00E+00
^{108m}Ag	< 1.52E+04	< 5.81E+02	< 3.81E-02	< 8.71E+01
^{110m}Ag	< 2.60E+04	< 5.01E+02	< 1.93E-02	< 7.52E+01
^{214}Bi	< 4.63E+04	0.00E+00	0.00E+00	0.00E+00
^{144}Ce	< 4.03E+05	< 2.14E+03	< 5.31E-03	< 3.21E+02
^{58}Co	< 2.68E+04	< 7.17E+02	< 2.68E-02	< 1.08E+02
^{60}Co	1.41E+07	1.84E+05	1.30E-02	2.75E+04
^{134}Cs	< 2.73E+04	4.37E+03	> 1.60E-01	> 6.55E+02
^{137}Cs	< 2.29E+04	< 5.41E+02	< 2.36E-02	< 8.11E+01
^{152}Eu	< 2.37E+04	< 1.13E+03	< 4.77E-02	< 1.70E+02
^{154}Eu	< 1.54E+04	< 1.00E+03	< 6.54E-02	< 1.51E+02
^{155}Eu	< 1.81E+04	< 7.69E+02	< 4.26E-02	< 1.15E+02
^{54}Mn	< 2.81E+04	< 6.67E+02	< 2.38E-02	< 1.00E+02
^{94}Nb	< 2.15E-01			
^{95}Nb	< 2.47E+04			
^{239}Np	< 2.19E+04			
^{214}Pb	< 3.28E+04			
^{226}Ra	< 2.37E+05	< 8.56E+02	< 3.60E-03	< 1.28E+02
^{103}Ru	< 1.61E+05	< 5.16E+02	< 3.20E-03	< 7.74E+01
^{106}Ru	< 1.91E+05	< 4.52E+03	< 2.37E-02	< 6.77E+02
^{125}Sb	4.14E+06	1.71E+04	4.15E-03	2.57E+03
^{235}U	< 1.45E+04	< 2.17E+03	< 1.50E-01	< 3.26E+02
^{65}Zn	1.48E+05	4.17E+03	2.81E-02	6.25E+02
^{95}Zr	< 4.40E+04	< 1.18E+03	< 2.69E-02	< 1.78E+02

Table A-4. (continued).

Nuclide	#487022-46 anion 25.8 g	IP1-93 #487022-46 anion 248.1 mL	Fraction Release	pCi/yr/g resin
Other radionuclides				
^{129}I	< 2.78E+02			
^{59}Ni	3.54E+05	4.14E+03	1.17E-02	6.22E+02
^{14}C	3.43E+04	< 5.31E+01	< 1.55E-03	< 7.97E+00
^{63}Ni	7.88E+07	3.75E+05	4.75E-03	5.62E+04
^{241}Pu	4.24E+04	1.27E+02	3.00E-03	1.91E+01
^{90}Sr	9.83E+02	< 2.33E+01	< 2.37E-02	< 3.49E+00
^{99}Tc	3.17E+03	< 3.00E+01	< 9.46E-03	< 4.50E+00

Table A-5. Deborating resin leach results (37 days).

Nuclide		#480722-36 22.38 g		IP2-37 #480722-36 259.1 mL		Fraction Release		pCi/yr/g resin
Alpha Spectrometry								
^{241}Am		2.42E+01	<	6.27E-01	<	2.59E-02	<	2.73E-01
^{242}Cm	<	2.06E+01	<	4.56E-01	<	2.21E-02	<	1.98E-01
^{244}Cm	<	3.69E+01	<	6.50E-01	<	1.76E-02	<	2.83E-01
^{238}Pu	<	4.70E+01	<	1.17E+00	<	2.49E-02	<	5.09E-01
$^{239/240}$Pu	<	2.64E+01		8.58E-01		3.25E-02		3.73E-01
^{233,234}U		2.14E+02		2.64E+00		1.23E-02		1.15E+00
^{235}U	<	2.16E+01	<	4.07E-01	<	1.89E-02	<	1.77E-01
^{238}U	<	2.15E+01	<	4.53E-01	<	2.11E-02	<	1.97E-01
Gamma Spectrometry								
^{108m}Ag	<	3.02E+03	<	4.33E+01	<	1.43E-02	<	1.88E+01
^{110m}Ag	<	2.37E+03	<	3.52E+02	<	1.49E-01	<	1.53E+02
^{214}Bi	<	1.27E+04						
^{144}Ce	<	1.59E+04	<	5.26E+02	<	3.31E-02	<	2.29E+02
^{58}Co	<	3.22E+03	<	5.78E+01	<	1.79E-02	<	2.51E+01
^{60}Co		2.26E+04		9.82E+02		4.34E-02		4.27E+02
^{134}Cs	<	4.07E+03		5.13E+02		1.26E-01		2.23E+02
^{137}Cs	<	6.56E+03		9.61E+03		1.47E+00		4.18E+03
^{152}Eu	<	6.47E+03	<	1.97E+02	<	3.05E-02	<	8.57E+01
^{154}Eu	<	4.16E+03	<	1.29E+02	<	3.10E-02	<	5.61E+01
^{155}Eu	<	4.21E+03	<	1.94E+02	<	4.61E-02	<	8.44E+01
^{54}Mn	<	3.29E+03	<	5.05E+01	<	1.54E-02	<	2.20E+01
^{94}Nb	<	2.78E-03						
^{95}Nb	<	3.00E+03						
^{239}Np	<	5.33E+03						
^{214}Pb	<	1.29E+04						
^{226}Ra	<	6.58E+04	<	2.37E+02	<	3.60E-03	<	1.03E+02
^{103}Ru	<	1.64E+03	<	1.10E+02	<	6.68E-02	<	4.78E+01
^{106}Ru	<	2.22E+04	<	7.23E+02	<	3.26E-02	<	3.14E+02
^{125}Sb		1.23E+05		2.38E+04		1.94E-01		1.03E+04
^{235}U	<	4.01E+03	<	5.36E+02	<	1.34E-01	<	2.33E+02
^{65}Zn	<	8.97E+03	<	1.32E+02	<	1.47E-02	<	5.74E+01
^{95}Zr	<	4.97E+03						

Table A-5. (continued).

Nuclide	#480722-36 22.38 g	IP2-37 #480722-36 259.1 mL	Fraction Release	pCi/yr/g resin
Other radionuclides				
^{129}I	< 3.00E+02			
^{59}Ni	< 2.11E+03	< 6.43E+01	< 3.05E-02	< 2.79E+01
^{14}C	5.62E+02	6.71E+01	1.19E-01	2.92E+01
^{63}Ni	7.50E+04	5.21E+02	6.95E-03	2.26E+02
^{241}Pu	< 3.04E+03	8.99E+00	2.95E-03	3.91E+00
^{90}Sr	< 5.53E+02	4.09E+00	7.41E-03	1.78E+00
^{99}Tc	< 0.00E+00	6.45E+00		2.80E+00

Table A-6. Deborating resin leach results (93 days).

Nuclide	#480722-36 23.17 g	IP2-93 #480722-36 250.2 mL	Fraction Release	pCi/yr/g resin
Alpha Spectrometry				
^{241}Am	2.50E+01	< 4.40E-01	< 1.76E-02	< 7.36E-02
^{242}Cm	< 2.13E+01	< 4.85E-01	< 2.28E-02	< 8.11E-02
^{244}Cm	< 3.82E+01	< 4.40E-01	< 1.15E-02	< 7.36E-02
^{238}Pu	< 4.87E+01	< 1.09E+00	< 2.24E-02	< 1.82E-01
$^{239/240}$Pu	< 2.73E+01	< 6.26E-01	< 2.29E-02	< 1.05E-01
^{233,234}U	2.22E+02	5.05E+00	2.28E-02	8.44E-01
^{235}U	< 2.23E+01	3.55E-01	1.59E-02	5.94E-02
^{238}U	< 2.22E+01	3.55E-01	1.60E-02	5.94E-02
Gamma Spectrometry				
^{108m}Ag	< 3.13E+03	< 7.61E+01	< 2.43E-02	< 1.27E+01
^{110m}Ag	< 2.46E+03	< 5.55E+02	< 2.26E-01	< 9.28E+01
^{214}Bi	< 1.31E+04	0.00E+00	0.00E+00	0.00E+00
^{144}Ce	< 1.65E+04	< 8.01E+02	< 4.86E-02	< 1.34E+02
^{58}Co	< 3.34E+03	< 8.56E+01	< 2.56E-02	< 1.43E+01
^{60}Co	2.34E+04	7.66E+02	3.27E-02	1.28E+02
^{134}Cs	< 4.22E+03	5.98E+03	1.42E+00	9.99E+02
^{137}Cs	< 6.79E+03	1.10E+04	1.61E+00	1.83E+03
^{152}Eu	< 6.70E+03	< 2.88E+02	< 4.30E-02	< 4.81E+01
^{154}Eu	< 4.31E+03	< 1.96E+02	< 4.56E-02	< 3.28E+01
^{155}Eu	< 4.36E+03	< 3.08E+02	< 7.06E-02	< 5.14E+01
^{54}Mn	< 3.41E+03	< 7.68E+01	< 2.26E-02	< 1.28E+01
^{94}Nb	< 2.87E-03			
^{95}Nb	< 3.10E+03			
^{239}Np	< 5.51E+03			
^{214}Pb	< 1.34E+04			
^{226}Ra	< 6.81E+04	< 3.43E+02	< 5.03E-03	< 5.73E+01
^{103}Ru	< 1.70E+03	< 1.83E+02	< 1.08E-01	< 3.06E+01
^{106}Ru	< 2.30E+04	< 1.04E+03	< 4.54E-02	< 1.74E+02
^{125}Sb	1.27E+05	2.23E+04	1.76E-01	3.73E+03
^{235}U	< 4.15E+03	< 7.83E+02	< 1.89E-01	< 1.31E+02
^{65}Zn	< 9.29E+03	< 1.63E+02	< 1.76E-02	< 2.73E+01
^{95}Zr	< 5.14E+03	< 1.31E+02	< 2.54E-02	< 2.18E+01

Table A-6. (continued).

Nuclide	#480722-36 23.17 g	IP2-93 #480722-36 250.2 mL	Fraction Release	pCi/yr/g resin
Other radionuclides				
^{129}I	< 3.10E+02			
^{59}Ni	< 2.18E+03	< 8.93E+01	< 4.10E-02	< 1.49E+01
^{14}C	5.82E+02	5.00E+01	8.60E-02	8.36E+00
^{63}Ni	7.76E+04	5.55E+02	7.16E-03	9.28E+01
^{241}Pu	< 3.15E+03	6.33E+01	2.01E-02	1.06E+01
^{90}Sr	< 5.72E+02	1.26E+02	2.20E-01	2.11E+01
^{99}Tc				

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10. SUPPLEMENTARY NOTES

11. ABSTRACT (200 words or less)

As part of a study being performed for the Nuclear Regulatory Commission (NRC), small-scale waste specimens were collected following an AP/CAN-DEREM full-system decontamination of the reactor vessel of the Indian Point 2 reactor vessel in March 1995. CAN-DEREM is a modified version of a reduction process developed by Atomic Energy of Canada Ltd. for that country's heavy-water CANDU reactors. The purpose of this program was to evaluate the radionuclide and chelating agent inventories of some of the decontamination ion-exchange resins produced during the decontamination process and to assess the leachability of the resins. The samples of anion, cation, and mixed-bed ion-exchange resins were acquired and tested because few data have been obtained on the radionuclide content and leachability characteristics of full-system decontamination ion-exchange resins. Results of this study indicate that both chelating agents and radionuclides did leach from the resin.

12. KEY WORDS/DESCRIPTORS (List words or phrases that will assist researchers in locating the report.)
SCADP/RELAP5; severe fuel damage; thermal hydraulics; LWR; coolant system; reactor core

Radioactive waste, waste form, leachability, leach index, leach rate, ion-exchange resins, radioactive material, CAN-DEREM, chelating agents, radionuclide leachability, decontamination, decontamination effectiveness, full system decontamination

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