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Chemical and Radiation Stability of a Proprietary Cesium Ion Exchange Material Manufactured from WWL Membrane and SuperLig® 644

G. N. Brown
K. J. Carson
J. R. DesChane
R. J. Elovich
P. K. Berry

September 1996

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

Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

Pretreatment of nuclear process wastes for the ion exchange removal of cesium and other radionuclides has been proposed as one method to minimize the amount of high-level radioactive waste (HLW) at Hanford. This study evaluated cesium-selective SuperLig® 644 (IBC Advanced Technologies, American Fork, UT) entrapped in a proprietary WWL web membrane (3M, St. Paul, MN) for chemical and radiation stability in simulated caustic neutralized current acid waste (NCAW), 0.5 M HNO_3, distilled water, and air. Following exposure from 0 to 2.0E+09 rad, the material was evaluated for cesium uptake in a series of 5 M sodium NCAW simulants with variable cesium concentrations. The following specific conclusions and recommendations resulted from the study:

- The radiolytic stability of the SuperLig® 644/WWL membrane appears to be sufficient for typical activities involving ion exchange pretreatment of radioactive cesium. Essentially no decrease in cesium selectivity or loading (K_d) was observed during cobalt-60 gamma irradiation of the material in either distilled water or 0.5 M HNO_3 up to 1.0E+09 rad. The cesium K_d decreased by a factor of two after exposure to 2.0E+09 rad in water or acid.

- Essentially no decrease in cesium K_d was observed during the irradiation of the SuperLig® 644/ WWL membrane in 5 M NCAW or ambient air up to 1.0E+08 rad. However, under these same conditions, the observed Cs K_d values decreased more than an order of magnitude between 1.0E+08 and 2.0E+09 rad. These high irradiation dose results portray an extreme situation and should not be considered representative of typical process conditions.

- Chemical stability of the SuperLig® 644/WWL membrane under caustic conditions is significantly lower than in ambient air or under neutral or acidic conditions. Even at low irradiation doses (e.g., 0 to 1.0E+07 rad), cesium K_ds of approximately 80 milliliter per gram (mL g^{-1}) were observed for the NCAW irradiated membrane. At these lower irradiation doses, cesium K_ds of approximately 300 mL g^{-1} were measured in other matrices. These results suggest that the material is less stable in caustic solution irrespective of the radiation exposure.

In terms of mechanical stability, samples of the membrane retained their physical form throughout the entire experiment and were only slightly brittle after exposure to 2.0E+09 rad. The reader is reminded that the material evaluated was a finely ground (400 mesh) particulate that was engineered to form a polymeric fiber (WWL), not the macroscopic form of the SuperLig® 644 ion exchange resin (20 to 50 mesh).

Pacific Northwest National Laboratory[a] (PNNL) conducted this study for the Efficient Separations and Processing (ESP) Crosscutting Program, Office of Science and Technology, U.S. Department of Energy. The work was completed under the Technical Task Plan Number RL3-6-C3-42, Test Sorbents; Industrial Contracts.

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Acknowledgments

Pacific Northwest National Laboratory (PNNL) is collaborating with universities, national laboratories, and industry to develop and test new materials for the pretreatment of nuclear wastes stored at Hanford. The experimental work described in this report would not have been possible without the encouragement and resources provided by Kurt D. Gerdes, Efficient Separations and Processing (ESP) Crosscutting Program, Office of Science and Technology, U.S. Department of Energy. The authors would like to acknowledge the contributions to this effort of W. Bonner, W. Kuhn, G. Johnson, and M. Telander at PNNL (Richland, WA); D. Boggs, K. Carlson, T. Fredrickson, T. Kafka, D. Seely, I. Shaw-Epperson, N. Stern, L. White, and E. Wisted at 3M (St. Paul, MN); and R. Bruening, R. Decker, and S. Izatt at IBC Advanced Technologies (American Fork, UT). This work was performed by researchers at PNNL under the Technical Task Plan (TTP) RL3-6-C3-42.

Contents

Summary iii

Acknowledgments v

1.0 Introduction 1.1

1.1 Background 1.1

1.2 Historical Results 1.1

1.3 Objectives 1.2

1.4 Scope 1.2

2.0 Experimental 2.1

2.1 Ion Exchange Material Selection 2.1

2.2 Simulant Solution Selection 2.1

2.3 Cesium Batch Distribution 2.2

2.4 Sample Irradiation 2.3

3.0 Results and Discussion 3.1

3.1 Irradiation in Water 3.1

3.2 Irradiation in 0.5 M HNO_3 ..... 3.3

3.3 Irradiation in Air 3.3

3.4 Irradiation in NCAW 3.3

3.5 Cesium Batch Distribution as a Function of Irradiation Dose 3.7

4.0 Conclusions and Recommendations 4.1

5.0 References 5.1

Figures

3.1 Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in Distilled Water 3.2

3.2 Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in 0.5 M HNO_3 ..... 3.4

3.3 Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in Air 3.5

3.4 Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in NCAW 3.6

3.5 Cesium Batch Distribution Ratio as a Function of Irradiation Dose at a Na/Cs Ratio of 1.0E+05 for Samples of the SuperLig® 644/WWL Web Irradiated in Various Solutions ... 3.8

Table

2.1 NCAW Simulant Composition 2.2

1.0 Introduction

1.1 Background

The contents of Hanford's 177 underground storage tanks (UST) include a mixture of sludge, salt cake, and alkaline supernatant liquid. The insoluble sludge fraction of the waste consists of metal oxides and hydroxides and contains the bulk of the ^{90}Sr waste component and many transuranic (TRU) radionuclides. The salt cake, which resulted from extensive evaporation of aqueous solution, consists primarily of dried sodium salts. The supernate consists of concentrated aqueous solutions of sodium nitrate/nitrite salts with smaller quantities of hydroxide, aluminum, potassium, carbonate, sulfate, and phosphate. The bulk of the water-soluble radionuclides such as ^{137}Cs are likely contained in the supernatant solution, interstitial liquid, and salt cake fractions.

Although the pretreatment and disposal strategy for the USTs is still being defined, one of the first steps in most pretreatment scenarios will be a solids/liquid separation of the pumpable waste liquor, followed by ion exchange removal of cesium from the resulting supernatant liquid. Next, a salt cake dissolution and sludge wash step will be initiated, followed by another solids/liquid separation. Most of the cesium is expected to be in the aqueous liquids from these processes and it is these solutions that are the focus of the cesium ion exchange removal process. This process is being designed with the goal of removing enough cesium so the resulting low-level waste (LLW) will meet the U.S. Nuclear Regulatory Commission's (NRC) 10 CFR 61 Class A Limit for ^{137}Cs (1 Ci m^{-3}). However, a greater cesium decontamination and the removal of ^{90}Sr, ^{99}Tc, and TRUs from the alkaline supernate may be required to meet the NRC incidental waste designation or another designation.

1.2 Historical Results

The technology for cesium decontamination of high-level alkaline wastes and sludge wash waters has been investigated at the Pacific Northwest National Laboratory[a] (PNNL) in Richland, Washington (Bray 1989; Bray et al. 1990; Bray et al. 1992; Bray et al. 1993a; Bray et al. 1993b; Brown et al. 1995c; Kurath et al. 1994); the Westinghouse Savannah River Company (WSRC), Aiken, South Carolina (Bibler et al. 1989; Bibler 1991 and 1994; Bray et al. 1990); and the West Valley Nuclear Services Company, Inc. (WVNS) in West Valley, New York (Bray et al. 1984; Kurath et al. 1989; Bray and Hara 1991).

Many ion exchange materials have demonstrated the ability to remove cesium from various simulated solutions (Marsh et al. 1994a; Marsh et al. 1994b; Marsh et al. 1994c; Marsh et al. 1995). Brown et al. (1995b) investigated the column ion exchange removal of radioactive cesium from actual and simulated 101-SY and 103-SY tank waste using CS-100. In addition, several inorganic and organic

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materials have been evaluated for cesium and strontium selectivity using small-scale batch distribution experiments with actual and simulated 101-AW tank waste (Brown et al. 1996).

The chemical and radiolytic stability of several organic ion exchange resins (e.g., SuperLig® 644, resorcinol-formaldehyde [R-F], and CS-100) has been evaluated in simulated alkaline waste solutions using ^{60}Co gamma radiation (Bibler and Crawford 1994; Brown et al. 1995a; Bryan et al. 1993; Carlson et al. 1995; Hubler et al. 1995).

1.3 Objectives

Experimental studies are being conducted by PNNL under the Efficient Separations and Processing (ESP) Crosscutting Program to 1) develop and evaluate newly conceived novel materials for the recovery of cesium, strontium, and technetium from alkaline wastes; 2) determine the loading and elution efficiency of these processes; 3) determine the physical life cycle (including radiation and chemical stability) of these materials; and 4) determine if basic ion exchange data can be applied to a broad range of tank wastes. The goal is to provide the technology to produce a LLW effluent with radioactivity levels that are low enough to permit further treatment in the LLW glass vitrification facility. Ultimately, each process must be evaluated in terms of removal efficiency, process chemical consumption and recycle, chemical and radiation stability of materials, compatibility with other process streams, secondary waste generation, process and maintenance costs, final material disposal, and impact upon the volume and quality of the high-level waste (HLW).

The purpose of this report is to describe the chemical and radiolytic stability of the newly synthesized SuperLig® 644 sequestering agent (IBC Advanced Technologies, American Fork, UT), which has been engineered to form a proprietary web-like matrix called SL644/WWL (3M, St. Paul, MN). Material stability was evaluated under realistic process flow sheet solutions including a simulated Hanford alkaline waste supernate, dilute nitric acid, distilled water, and ambient air.

1.4 Scope

The work described in this report involves the evaluation of a cesium selective material (SuperLig® 644/WWL) based on the results obtained from chemical and radiolytic stability experiments conducted within the material. The material was irradiated to a total dose of 2.0E+09 rad at 1.6E+06 rad hr^{-1} in either a simulated neutralized current acid waste (NCAW) supernatant liquid, 0.5 M HNO_3, distilled water, or ambient air. The samples were analyzed for ion exchange selectivity and capacity by cesium batch distribution (K_d) experiments in a simulated NCAW solution containing variable concentrations of cesium and 5.0 M sodium. Prior to the ion exchange K_d contact, the initial sodium-to-cesium molar ratio (Na/Cs) ranged from 500 to 500,000. These K_d data were used to determine the relative stability of the material tested.

2.0 Experimental

Previous experimental work completed prior to Fiscal Year 1996 for cesium recovery included the evaluation of several ion exchange materials (e.g., SuperLig® 644, R-F, and CS-100) in simulated and actual waste matrices using bench-scale column loading and elution tests, small-scale batch distribution contacts, and chemical and radiolytic stability experiments. Brown et al. (1995a) exposed three organic ion exchange resins (SuperLig® 644, R-F, and CS-100) to either ^{60}Co gamma radiation or oxygen in simulated alkaline waste solutions to evaluate each material's relative radiolytic and oxidative stability. Radiolytic stability was deduced by periodic gas generation and cesium batch K_d analysis of each material. Stability toward oxidation was inferred by O_2 uptake measurements, nuclear magnetic resonance (NMR), and cesium batch K_d analysis of the exposed materials.

The focus of the work reported here is to further investigate the chemical and radiolytic stability of engineered SuperLig® 644 entrapped in a web-like polymer matrix and to compare the subsequent results to those obtained previously (Brown et al. 1995a) for the ion exchange resin in the absence of the WWL web matrix. The data described in this report can be found in the PNNL laboratory record book (LRB) BNW55765. Because of the limited availability of actual radioactive waste, simulated solutions have been used for the current stability testing.

2.1 Ion Exchange Material Selection

The removal of radioactive cesium by column ion exchange is considered to be the baseline pretreatment process for cleanup of a majority of the waste currently stored at Hanford. In addition to SuperLig® 644, several materials are capable of removing cesium from highly alkaline solutions, including organic ion exchange resins (CS-100 and R-F), inorganic zeolites (IE-95, IE-96, TIE-96), and other materials (Crystalline Silico-Titanates [CSTs]). The SuperLig® 644 polymer resin is the latest version of the covalently bound SuperLig® macrocycle family of sequestering ligands from IBC Advanced Technologies (American Fork, UT), and has been shown to be highly selective for cesium even in the presence of excess sodium or potassium (Brown et al. 1995c). The SuperLig® 644 polymer has recently become available engineered in a web-like matrix produced by 3M (St. Paul, MN). In the current study, this SuperLig® 644/WWL web material was evaluated for chemical and radiation stability under a variety of solution conditions.

2.2 Simulant Solution Selection

The NCAW simulant described in Table 2.1 was chosen for the current ion exchanger irradiation/ stability experiments. This composition is considered to be representative of much of the Hanford tank wastes. It has been previously shown that this composition replicates the chemical environment of actual NCAW tank wastes for radiation and chemical stability experiments (Brown et al. 1995a; Bryan et al. 1993; Carlson et al. 1995). Numerous cesium batch K_d and column loading tests have been completed

Table 2.1. NCAW Simulant Composition

Species	Molarity, M
Na	5.00
K	0.12
Rb	5.00E-05
Cs	Variable
Al	0.43
SO_4	0.15
OH (Total)	3.40
OH (Free)	1.68
CO_3	0.23
NO_2	0.43
NO_3	1.67
F	0.089
PO_4	0.025

using this formulation (Bray et al. 1992; Kurath et al. 1994; Brown et al. 1995c). Cesium was absent from the NCAW simulant during material irradiation, but was present during the batch K_d determinations at an initial concentration, which varied from 1.0E-02 to 1.0E-05 M as cesium nitrate.

2.3 Cesium Batch Distribution

The batch K_d ($K_d = [Cs]_{solid} \div [Cs]_{liquid}$) is an equilibrium measure of the overall ability of the solid phase ion exchange material to remove an ion from solution under the particular experimental conditions that exist during the contact. In most batch K_d tests, a known quantity of ion exchange material is placed in contact with a known volume of solution containing the particular ions of interest. The material is allowed to contact the solution for a sufficient time to achieve equilibrium at a constant temperature, after which the solid ion exchange material and liquid supernate are separated and analyzed. In this report, the batch K_ds were achieved by contacting 0.07 g of the web material with 10 mL of supernate liquid. The equation for determining the K_d can be simplified by determining the concentration of the analyte before and after contact and calculating the quantity of analyte on the ion exchanger by difference (Equation 1).

$$K_d = \frac{(C_i - C_f)}{C_f} * \frac{V}{M*F} \qquad (1)$$

C_i is the initial amount or activity of the ion of interest in the feed solution prior to contact, C_f is the amount or activity after contact, V is the solution volume, M is the exchanger mass, and F is the mass of dry ion exchanger divided by the mass of wet ion exchanger (F-factor). K_d (normal units are mL g^{-1}) represents the theoretical volume of solution (mL) that can be processed per mass of exchanger (dry weight basis) under equilibrium conditions. Lambda, the theoretical number of bed volumes of solution that can be processed per volume of exchanger, is obtained by multiplying K_d by the exchanger bed density, ρ_b (g of resin per mL of resin) as shown in Equation 2.

$$\lambda = K_d * \rho_b \tag{2}$$

The lambda value provides a method for comparing the ion exchange performance of a wide variety of materials on a volume basis (e.g., in an ion exchange column). Brown et al. (1995c) reported that this method is biased against low bed density materials (e. g., SuperLig® 644 $\rho_b = 0.22$ g mL^{-1}; R-F $\rho_b = 0.45$ g mL^{-1}).

The experimental equipment required to complete the batch K_d determinations included an analytical balance, a constant temperature water bath, an oven for F-factor determinations, a variable speed shaker table, 20-mL scintillation vials, 0.2-µm syringe filters, the appropriate ion exchanger, and simulant solutions. Prior to initiating the batch contact, each material was irradiated to the specified dose in the specified solution (e.g., cesium-deficient NCAW simulant, 0.5 M HNO_3, distilled water, or air), rinsed with copious amounts of distilled water, and then air dried for a minimum of 48 hours. Approximately 0.07 g of each material was contacted with 10 mL of the stock NCAW solutions (e.g., 5 M Na with variable cesium concentrations). The sample bottles were placed into a 25°C constant temperature bath and shaken lightly for 72 hours. The samples were then filtered with a 0.2-µm pore filter to separate the resin material from the solution and the resulting liquid was analyzed for cesium content by ^{137}Cs gamma counting.

2.4 Sample Irradiation

To evaluate the chemical and radiation stability of the SuperLig® 644/WWL sorbent, samples were contained in glass scintillation vials and exposed to high dose ^{60}Co gamma radiation. The sealed system provided a convenient system for exposing the resin to either an NCAW simulant, 0.5 M HNO_3, distilled water, or ambient air during the radiolysis process. A single piece of the sorbent web weighing approximately 0.6 g was triple rinsed and placed into 15 mL of the appropriate irradiation solution. No attempt was made to evacuate or purge the entrained oxygen or air from the sample containers. Following irradiation, the samples were rinsed with copious amounts of distilled water and air dried for a minimum of 48 hours.

Small subsample (0.07-g) portions of the sorbent were evaluated at each irradiation dose for cesium uptake using the ^{137}Cs batch distribution coefficient K_d (see Section 2.3). The remainder of the irradiated sample for each solution was dried at 105°C for 24 hours and the fraction (F-factor) of easily removed water was calculated. The samples were contacted with fresh NCAW simulant containing a variable

initial cesium concentration (Na/Cs from 500 to 500,000). The obtained distribution coefficients are used as an indicator of the material cesium loading and an approximation of the radiation damage to the resin material. The materials were exposed to a total radiation dose of approximately 2.0E+09 rad at 1.6E+06 rad hr^{-1} over the course of several weeks.

The Gamma Irradiation Facility is operated by PNNL and contains 37 stainless steel irradiation tubes positioned in a 7-ft-dia. by 13-ft-, 8-in.-deep stainless steel tank. Two arrays of ^{60}Co with a combined inventory of 32 KCi are located near the bottom of the tank. For radiation shielding purposes, the tank is completely filled with water and a 3.5-ft-tall concrete wall surrounds the top of the tank. The irradiation tubes, which are sealed on the bottom, vary in length and diameter from 16 ft to 18 ft and 1.8 in. to 6 in., respectively. The irradiation flux of the tubes ranges from approximately 2.0E+06 rad hr^{-1} to 2.0E+02 rad hr^{-1}. The uniform flux region varies from ~6 in. for the tubes closest to the sources to greater than 12 in. for the tubes farthest away from the sources. All flux measurements of the tubes are traceable to the National Institute of Standards and Technology.

All materials and test systems were manually lowered into the irradiation tubes and were left in the tubes for the specific amount of time (6.25E-01, 6.25E+00, 6.25E+01, 6.25E+02, 1.25E+03 hour) to achieve the required exposure (1.0E+06, 1.0E+07, 1.0E+08, 1.0E+09, 2.0E+09 rad). There is no activation associated with the gamma irradiation, so the materials were transported to other facilities for Cs batch K_d evaluation (Section 2.3) after they were removed from the tubes. The temperature of the test was approximately 15°C to 20°C based on the recorded ambient temperatures in the gamma facility.

3.0 Results and Discussion

IBC Advanced Technologies has developed a new class of sequestering agents (SuperLig®) that can selectively remove various radionuclides (cesium, strontium, etc.) from high ionic-strength alkaline solutions based on molecular recognition technology. Previous tests have demonstrated that one of these materials in resin form (SuperLig® 644) can remove cesium from simulated Hanford tank waste even in the presence of excess sodium and potassium (Brown et al. 1995c). To more accurately assess the potential use of this technology for pretreating nuclear process wastes, the oxidative and radiolytic stability of SuperLig® 644 was tested and compared to that of two current baseline materials (Brown et al. 1995a).

More recently, IBC Advanced Technologies and 3M (St. Paul, MN) have collaborated to develop a novel form of the SuperLig® 644 material. In this case, the organic resin is ground to a fine uniform particle size and entrapped in a web-like fibrous matrix designated WWL. Due to the unique nature of this novel material, the chemical and radiolytic stability has been questioned. The increased surface area-to-mass ratio of the smaller particles raises the potential for reduced stability; conversely, there is also a possibility for an increase in stability because of the greater stability of the inert fibrous matrix. For these reasons, the SuperLig® 644/WWL material was exposed to ^{60}Co gamma radiation in a variety of solutions over several weeks. The irradiated samples were analyzed for cesium selectivity in a simulated NCAW solution by batch distribution (K_d) analysis.

3.1 Irradiation in Water

Cesium batch distribution (K_d) results as a function of equilibrium Na/Cs ratio are displayed in Figure 3.1 for samples of the SuperLig® 644/WWL membrane irradiated in distilled water. Each data curve represents a known irradiation dose from 0 to 2.0E+09 rad. It is evident from the data that irradiation in water to 1.0E+09 rad does not appreciably change the cesium K_d results. Even after irradiation to 2.0E+09 rad, the cesium K_d decreased by less than a factor of two.

The K_d data are presented as a function of the equilibrium Na/Cs ratio (i.e., the Na/Cs ratio remaining after material and solution contact) so the reader can easily extrapolate back to the original composition of the NCAW feed solution. Data displayed in this manner allow evaluation of material loading and extrapolation to column ion exchange performance. The samples were evaluated for cesium uptake in a series of standard NCAW simulants with variable cesium content (500 to 500,000 Na/Cs) as described by previous researchers (Bibler et al. 1989; Bray et al. 1990; Bray et al. 1992; Brown et al. 1996; Kurath et al. 1994).

At a final Na/Cs ratio of 5.0E+04 (e.g., the composition of the standard NCAW solution used previously by Brown et al. [1995c]), the cesium K_d was determined to be approximately 220 mL g^{-1} for the nonirradiated samples. The value decreased slightly to 170 mL g^{-1} after irradiation to 2.0E+09 rad. Multiplying this value by the estimated SuperLig® 644/WWL bed density

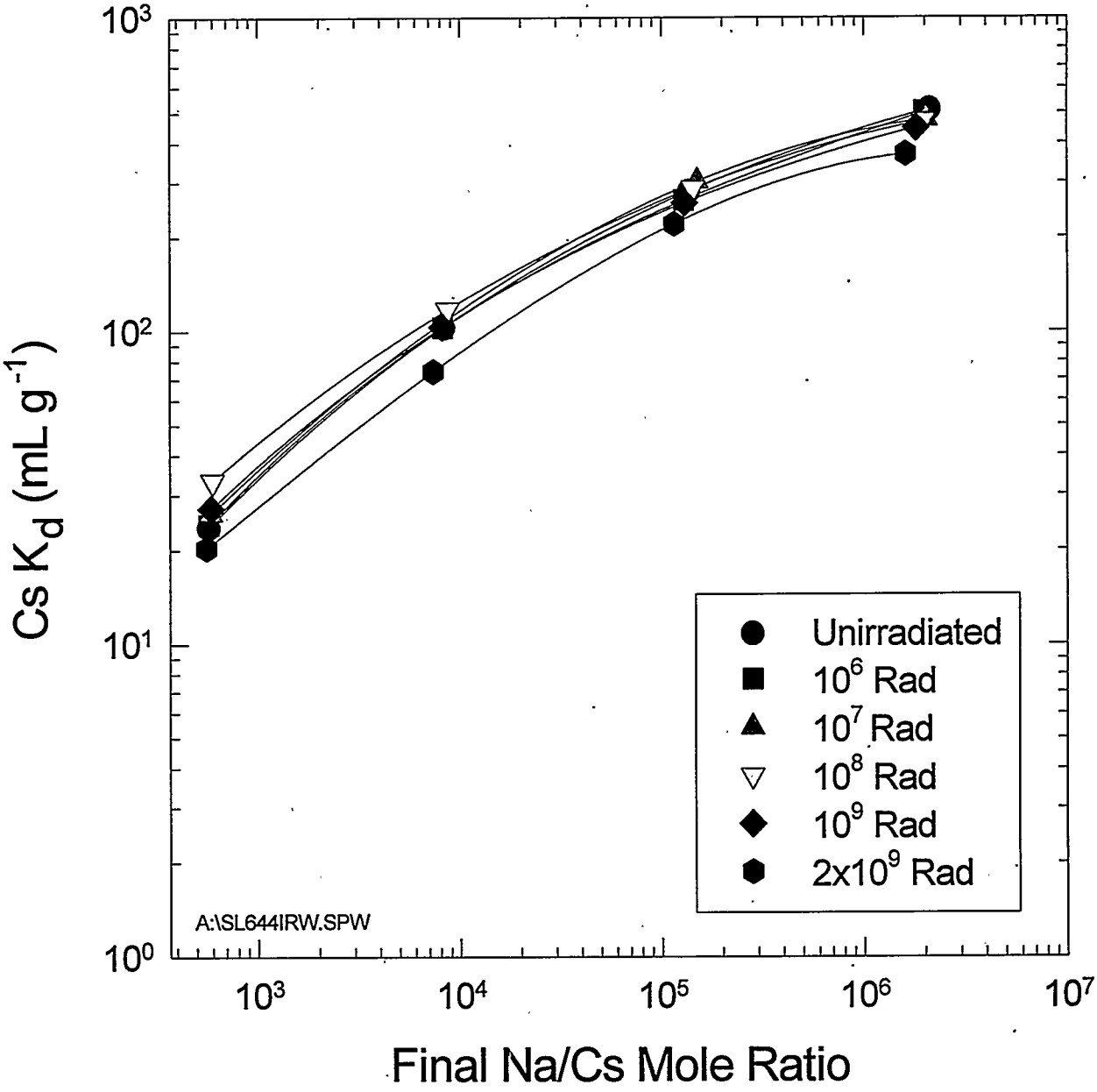

Figure 3.1. Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in Distilled Water

(ρ_b = 0.347 g mL^{-1}) yields a lambda (λ) value of nearly 80 column volumes (CV). When the fractional amount of SuperLig® 644 loaded into the WWL membrane is taken into account, this value compares favorably to the 110 CV value reported earlier for nonirradiated SuperLig® 644 resin (Brown et al. 1995c). The percentage of active particle in the WWL membrane is known to vary between 60% and 95%, depending upon production conditions.

3.2 Irradiation in 0.5 M HNO_3

The cesium K_d results for the irradiation of the SuperLig® 644/WWL membrane in dilute nitric acid (0.5 M HNO_3) are displayed in Figure 3.2. The results are analogous and are nearly identical to those displayed in Figure 3.1 for irradiation in distilled water. The similarities are not surprising since the SuperLig® 644/WWL material is obtained from the supplier in the acid form. The manufacturer suggests that if the material is to be stored for an extended time, the acid form is preferred. The equilibrium pH was not measured during the experiment, but it is likely that the distilled water became slightly acidic after contact. At 2.0E+09 rad, the samples irradiated in either water or acid exhibited a moderate weight loss (6%) that was only slightly greater than the 2% weight loss observed for the lower irradiation doses.

3.3 Irradiation in Air

The cesium K_d results for the irradiation of the SuperLig® 644/WWL membrane in ambient air are displayed in Figure 3.3. The results are analogous but more extreme than those described above for the irradiation in dilute nitric acid or distilled water. After irradiation, the cesium K_d results at a Na/Cs ratio of 1.0E+05 drop significantly from approximately 300 mL g^{-1} at 1.0E+07 rad to less than 30 mL g^{-1} at 2.0E+09 rad. Evidently, exposure of the material to either dilute acid or distilled water solution during the irradiation process may protect the resin from chemical degradation. Several protection mechanisms can be postulated, including free radical transfer from the resin to a water molecule or a reduction in oxygen availability in solution versus in air. The air-irradiated sample actually displayed a minor weight gain (0.8% and 2.1%) at 1.0E+09 and 2.0E+09 rad, respectively.

3.4 Irradiation in NCAW

The cesium K_d results for irradiation of the SuperLig® 644/WWL membrane in simulated NCAW solutions are displayed in Figure 3.4. The results are analogous and surprisingly similar to those described for irradiation in ambient air. However, after irradiation, the cesium K_d results drop significantly from approximately 70 mL g^{-1} at 1.0E+07 rad to less than 1 mL g^{-1} at 2.0E+09 rad. Also evident is the significant reduction in cesium K_d for the nonirradiated material stored in NCAW when compared to storage and irradiation in either distilled water, dilute nitric acid, or ambient air. Evidently, the material is less stable toward radiation in the NCAW than in air or other solutions.

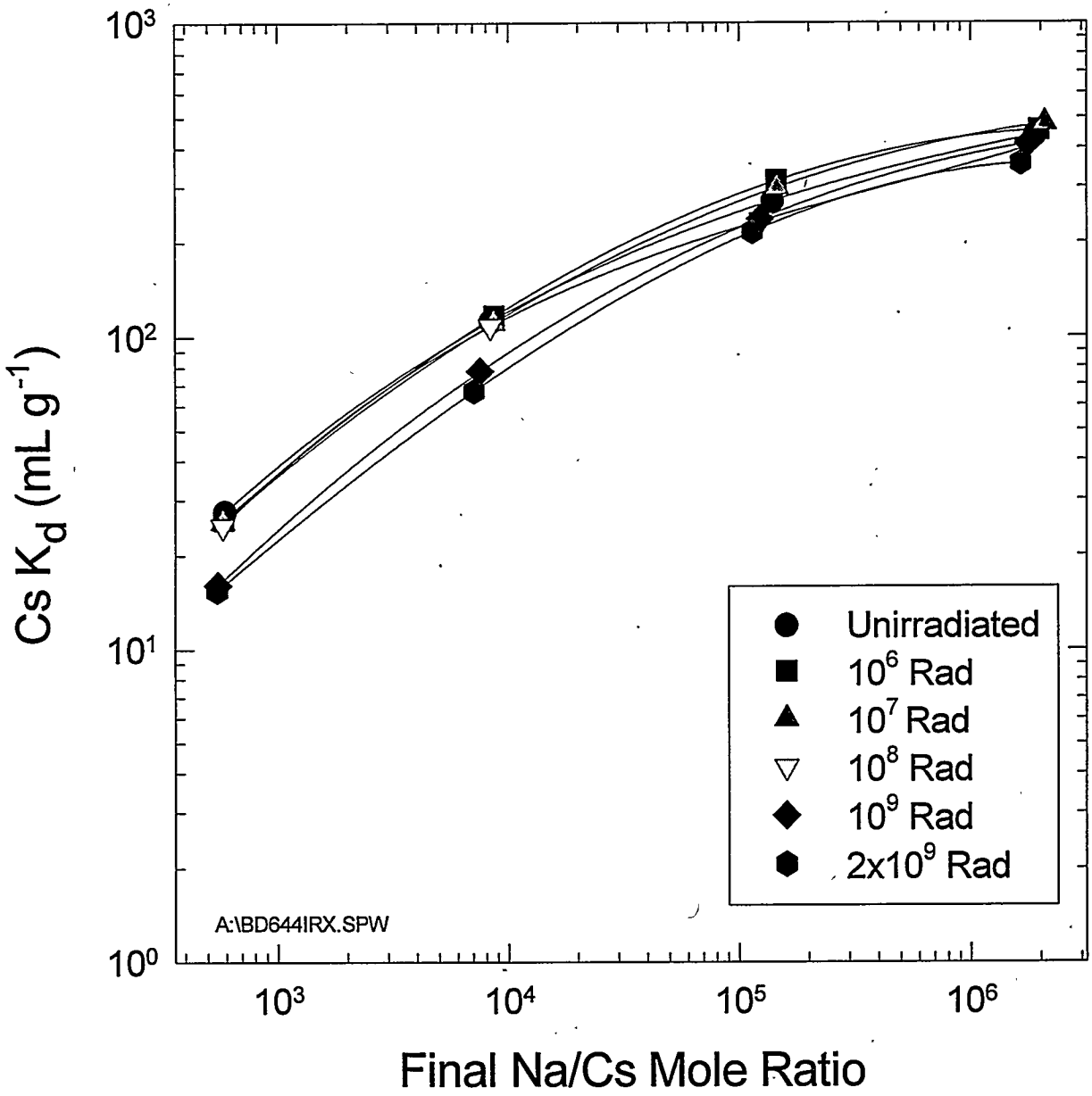

Figure 3.2. Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in 0.5 M HNO_3

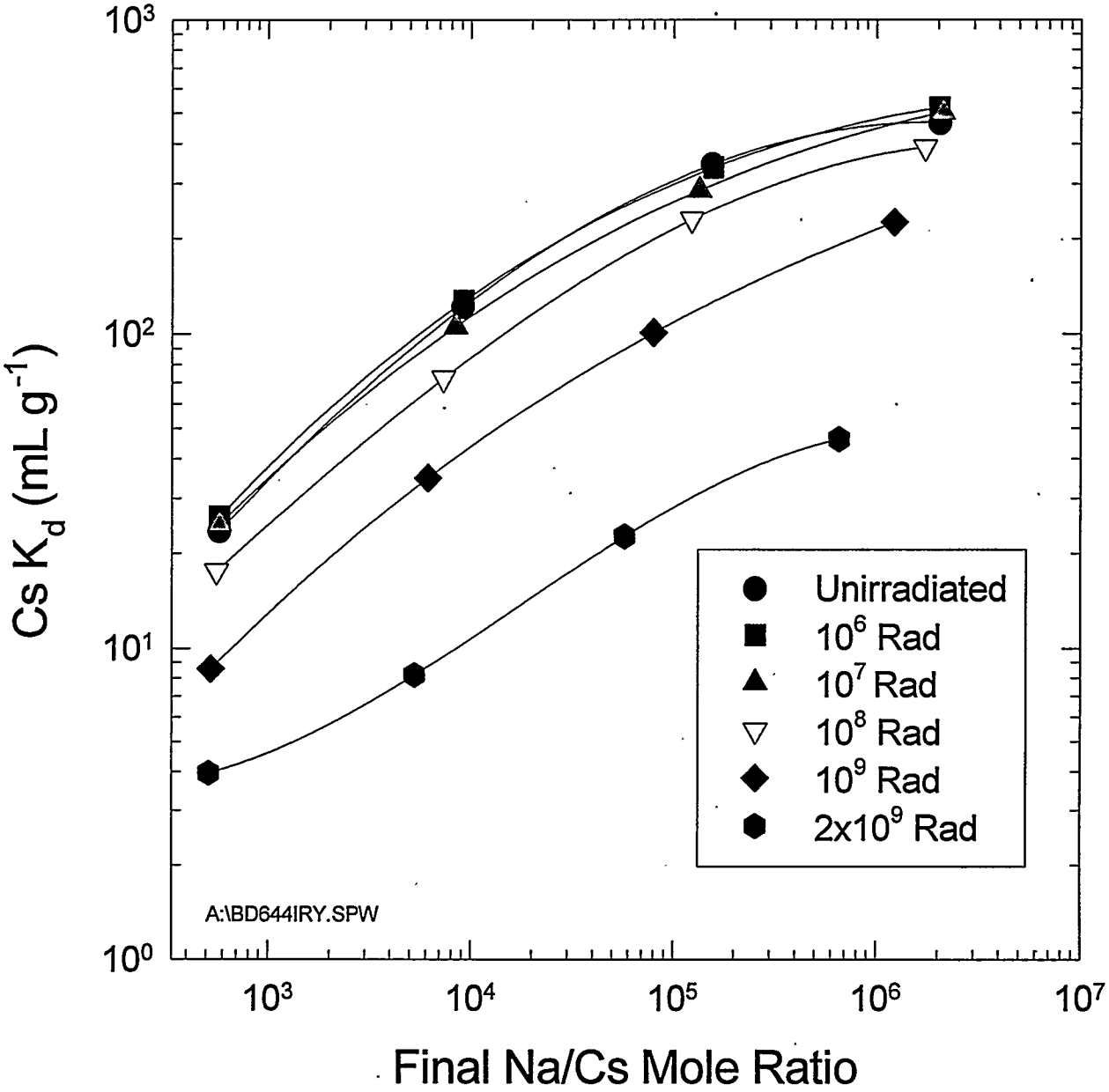

Figure 3.3. Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in Air

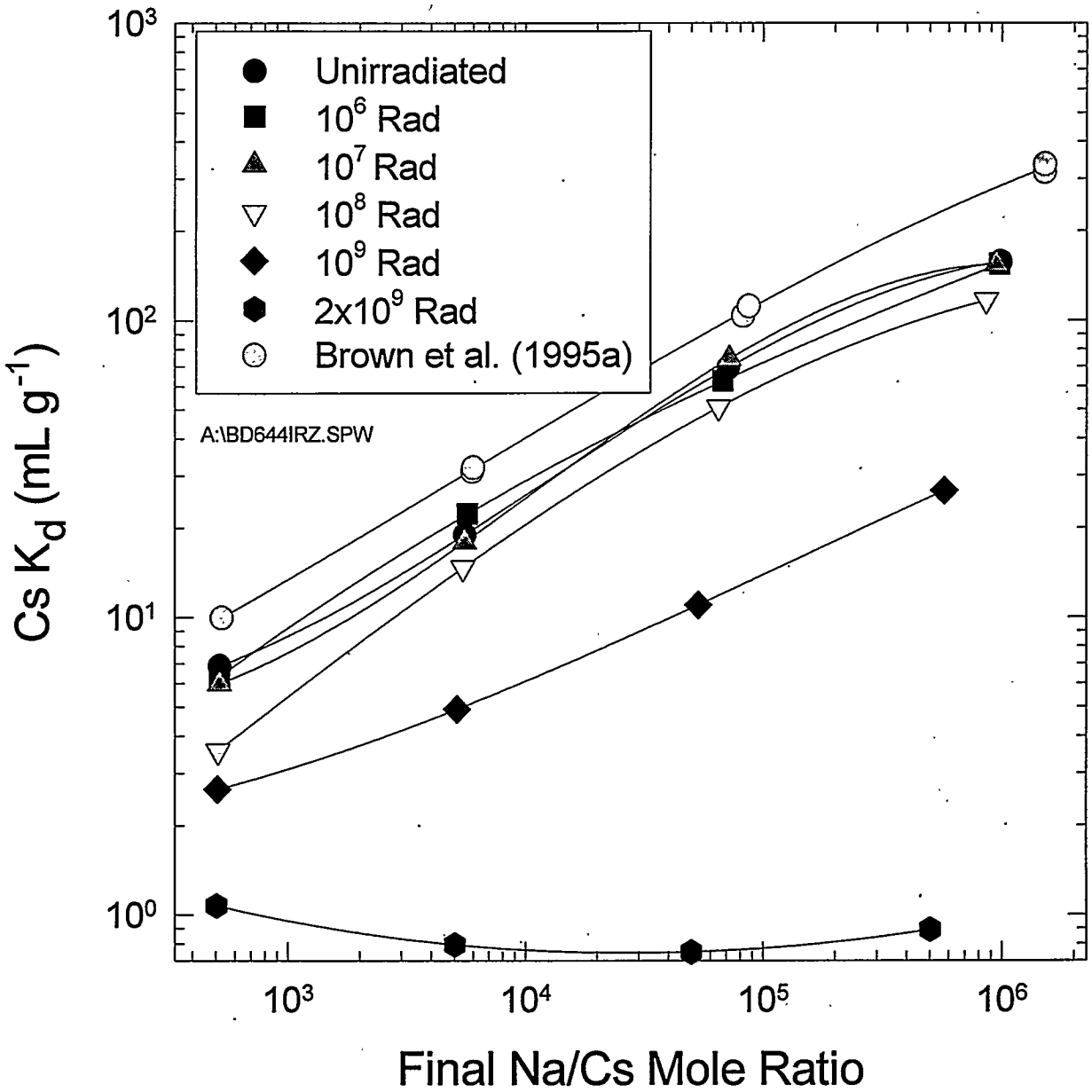

Figure 3.4. Cesium Batch Distribution Ratio as a Function of Na/Cs Ratio for Samples of the SuperLig® 644/WWL Web Irradiated in NCAW

A comparison of these results to those described previously (Brown et al. 1995a) for a pure SuperLig® 644 resin irradiated at 1.0E+09 rad is also displayed in Figure 3.4. The previous K_d results are significantly higher than the current results. For example, in the current experiment, a cesium K_d value of 14 mL g^{-1} (measured in NCAW at 1.0E+05 Na/Cs) was determined for the SuperLig® 644/WWL membrane irradiated in an NCAW solution to 1.0E+09 rad. Using the same irradiation solution, the SuperLig® 644 polymeric resin yielded a K_d value of 110 mL g^{-1} (Brown et al. 1995a).

Several important differences between the two experiments could account for the lower cesium K_d results. First, in the current tests, no attempt was made to exclude oxygen or entrained air from the samples. In the previous gas generation experiments (Brown et al. 1995a), the samples were explicitly purged with ultra high purity argon so the generation of various gases could be accurately determined. Secondly, the SuperLig® 644 material is known to swell in caustic solutions, an effect that may enhance diffusion-limited degradation mechanisms as compared to the unswelled sorbent (e.g., irradiated in air, water, or acid). Thirdly, the SuperLig® 644/WWL membrane contained smaller active particles (>400 mesh) than the polymeric resin (20 to 50 mesh), which might also explain the differences. Also, the current membrane samples were irradiated at a higher volume-to-mass phase ratio (V/M = 30) than the resin samples (V/M = 6) reported by Brown et al. (1995a). Finally, a significant post-irradiation weight increase (50%) from approximately 0.5 g to 0.75 g was observed for these samples and was only partially compensated by the F-factor (0.88) correction. Evidently, the sorbent picked up one or more of the components from the NCAW that could not be easily rinsed out of the web. In this case, the actual sorbent mass would be lower than expected and a reduced cesium K_d would be calculated.

Previous column loading experiments in NCAW at 5.0E+04 Na/Cs (Brown et al. 1995c) have indicated a loading of between 80 and 90 CV for the nonirradiated SuperLig® 644 resin. In the current experiment (Figure 3.4), it was estimated that only 21 CV could be processed ($\lambda = \rho_b \times K_d = 0.347 * 60$) for the SuperLig® 644 membrane. From Section 3.3, samples that were not exposed to NCAW demonstrated a greater performance ($\lambda = \rho_b \times K_d = 0.347 * 300 = 104$). It is likely that this difference is due to the additional 8-week exposure to the NCAW solution prior to the batch K_d experiment.

3.5 Cesium Batch Distribution as a Function of Irradiation Dose

Cesium batch K_d results for samples of the SuperLig® 644/WWL membrane are displayed in Figure 3.5 as a function of irradiation dose. The data are displayed at a constant equilibrium Na/Cs ratio of 1.0E+05 (5.0E-05 M cesium). The chosen Na/Cs ratio was entirely arbitrary but represents an approximate estimate of the cesium concentration expected in many of the Hanford waste tanks. The samples were irradiated with an external ^{60}Co source at 1.6E+06 rad hr^{-1} in a variety of aqueous solutions. The maximum dose was 2.0E+09 rad and required nearly eight weeks of exposure.

From the data it is evident that the K_d values are nearly constant for all solution conditions at exposures below 1.0E+08 rad. A slight reduction in K_d is noted for the samples irradiated up to 2.0E+09 rad in distilled water and dilute nitric acid. However, this degradation appears to be minor when compared to the results for the material irradiated in air or NCAW. In these cases, a significant

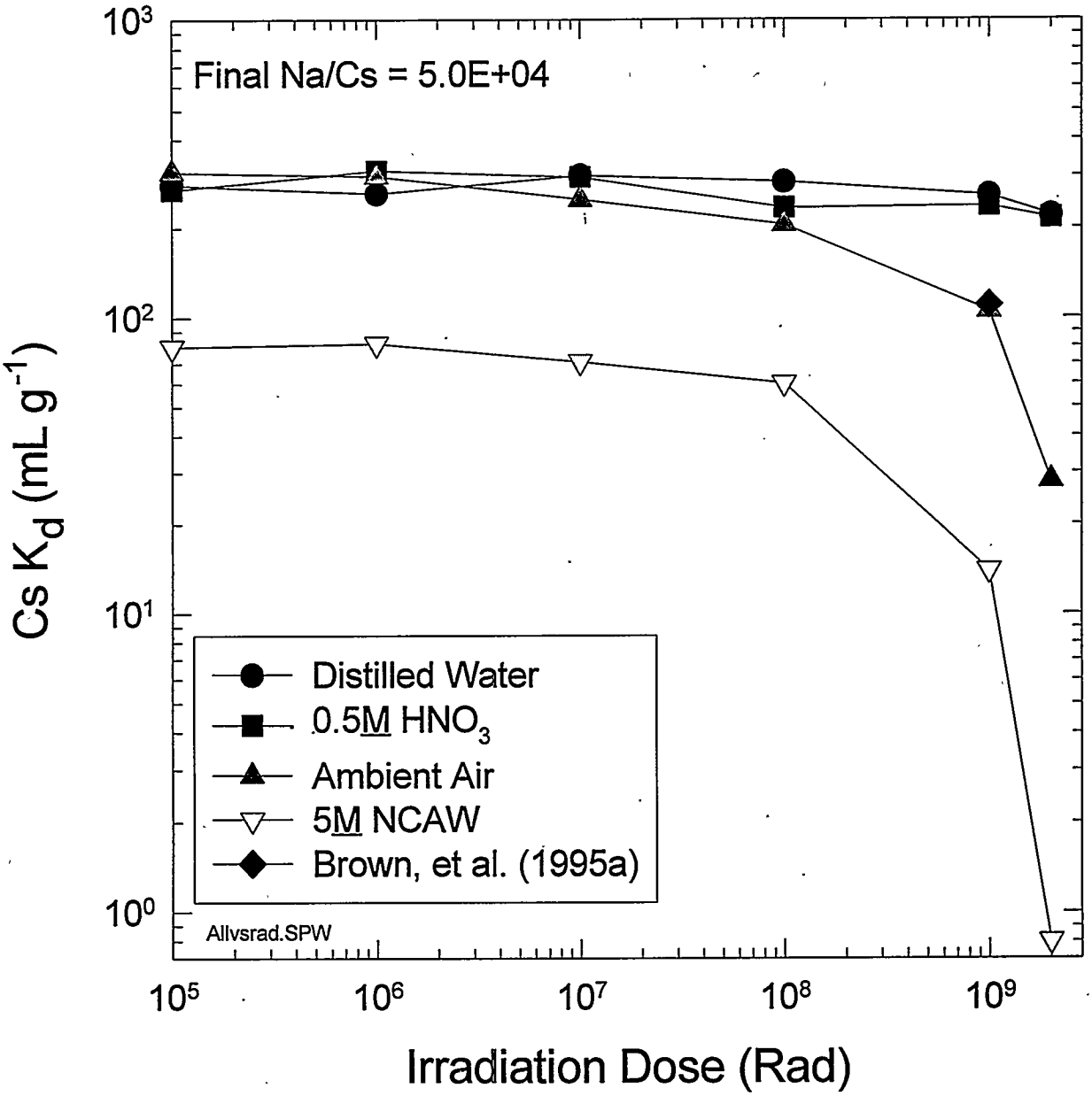

Figure 3.5. Cesium Batch Distribution Ratio as a Function of Irradiation Dose at a Na/Cs Ratio of 1.0E+05 for Samples of the SuperLig® 644/WWL Web Irradiated in Various Solutions

reduction in the measured K_d value occurred at exposures greater than 1.0E+08 rad. For the air-irradiated sample, K_d decreased more than an order of magnitude from approximately 300 to less than 30 mL g^{-1}. For the NCAW-irradiated sample, a reduction from 80 mL g^{-1} to less than 1 mL g^{-1} was observed. Degradation at these extreme conditions is not unexpected and has been observed previously for the SuperLig® 644 and other organic resins (Brown et al. 1995a).

However, the reduction is minor considering that most organic materials display extreme degradation at these high exposures. The SuperLig® 644/WWL web retained its physical form over the entire exposure range and was only slightly brittle at the highest doses (over 1.0E+09 rad). These doses are based on irradiation with an external ^{60}Co source and may or may not accurately predict resin exposure during actual waste processing. However, Penwell et al. (1994) estimated that a maximum radiation field of 2.7E+05 rad hr^{-1} would exist at the surface of an NCAW-loaded R-F ion exchange column. The NCAW solution is expected to contain the highest cesium concentration and therefore exhibit the highest radiation field. Assuming a loading cycle of 50 hours (e.g., full loading at 300 CV and a process flowrate of 6 CV hr^{-1}) and a total of 10 loading cycles before resin disposal, a conservative maximum for material exposure would be 1.4E+08 rad. The time-averaged radiation dose would be approximately one-half this value since the resin is only fully loaded at the end of the cycle. Therefore, it is estimated that 1 to 2 months of continuous operation would be required to reach the 1.0E+08 rad dose.

One interesting observation is that the NCAW-irradiated material consistently yields lower K_d values than materials irradiated in other solutions. Even the nonirradiated sample that was exposed to the NCAW simulant exhibited a much lower value (80 mL g^{-1}) compared to the other solutions (300 mL g^{-1}). This effect is attributed to the reduced chemical stability of the SuperLig® 644 material in caustic solutions (Brown et al. 1995a). During the radiation testing, all samples were exposed to the respective solutions for the entire 8 weeks of the experiment. The samples were prepared prior to the start of the irradiation process and were irradiated in reverse order so that all the materials would be removed simultaneously at the end of the test. Therefore, the SuperLig® 644/WWL webs were exposed to the NCAW, distilled water, dilute nitric acid, and air for the entire 8-week irradiation period, irrespective of the dose received. This method allows the separation of chemical degradation (solution exposure) from radiolytic degradation as well as nonsolution degradation (air exposure).

If the NCAW-irradiated samples had been freshly prepared such that the resin was only exposed to the NCAW solution during irradiation, one might observe degradation of the resin based on chemical and radiation damage and would not be able to separate the two effects. With these conditions, the chemical degradation of the SuperLig® 644/WWL is clearly differentiated from the radiolytic stability. If the nonirradiated samples had been placed into the NCAW solution just prior to determining the cesium K_d, one would expect the K_d data to be much closer to that measured for irradiation in ambient air (e.g., constant 250 to 300 mL g^{-1} between 0 and 1.0E+08 rad). No evaluation of resin storage methods (e.g., storage under nitrogen or other inert cover gas) was attempted.

Unfortunately, the data suggest that prolonged storage in a caustic solution will enhance chemical degradation of the resin. Due to safety considerations and the concern regarding the potential for organic-nitrate oxidation reactions, cation exchange resins are often converted to the sodium form with

sodium hydroxide to prepare them for storage. Historically, runaway organic-nitrate oxidation reactions have caused significant damage to ion exchange systems. For this reason, following elution with nitric acid, it is recommended that the organic resin be converted to the sodium form as quickly as possible.

4.0 Conclusions and Recommendations

The data among the tests are consistent and indicate the relative stability of the SuperLig® 644/WWL membrane under a variety of chemical and radiolytic conditions. The material was irradiated in ambient air, distilled water, dilute nitric acid, and highly caustic tank waste simulant (NCAW). Following exposure from 0 to 2.0E+09 rad, the material was evaluated for cesium uptake in a series of 5 M sodium NCAW simulants with variable cesium concentrations.

- Essentially no decrease in cesium selectivity or capacity (K_d) was observed during the irradiation of the SuperLig® 644/WWL membrane in distilled water or 0.5 M HNO_3 up to a total gamma irradiation (cobalt-60) dose of 2.0E+09 rad. The stability of the SuperLig® 644/WWL membrane, as measured by the cesium K_d on a mass equivalent basis, is essentially equivalent to that of the SuperLig® 644 resin in the absence of the WWL membrane.

- Essentially no decrease in cesium selectivity or capacity (K_d) was observed during the irradiation of the SuperLig® 644/WWL membrane in 5 M NCAW or ambient air up to 1.0E+08 rad.

- Following irradiation of the SuperLig® 644/WWL membrane in either 5 M NCAW or ambient air, the observed Cs K_d values decreased more than an order of magnitude between 1.0E+08 and 2.0E+09 rad. These high irradiation dose results portray upper bounding limits and should not be considered representative of typical ion exchange process conditions.

- The cesium K_d results for the SuperLig® 644/WWL membrane irradiated in 5 M NCAW were significantly lower than those results obtained in other solutions (0.5 M HNO_3, distilled water, or ambient air). K_ds of approximately 80 mL g^{-1} were observed for the NCAW-irradiated membrane and about 300 mL g^{-1} for irradiation in the other matrices. These results suggest that the material is less stable in the caustic NCAW solution than in the other matrices, irrespective of the radiation exposure.

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