Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Studies on Cesium Uptake by Phenolic Resins

S. K. Samanta[a]; M. Ramaswamy[a]; B. M. Misra[b]
[a] PROCESS ENGINEERING AND SYSTEMS DIVISION, BHABHA ATOMIC RESEARCH CENTRE, BOMBAY, INDIA [b] DESALINATION DIVISION, BHABHA ATOMIC RESEARCH CENTRE, BOMBAY, INDIA

To cite this Article Samanta, S. K. , Ramaswamy, M. and Misra, B. M.(1992) 'Studies on Cesium Uptake by Phenolic Resins', Separation Science and Technology, 27: 2, 255 — 267

To link to this Article: DOI: 10.1080/01496399208018877

URL: http://dx.doi.org/10.1080/01496399208018877

SEPARATION SCIENCE AND TECHNOLOGY, 27(2), pp. 255–267, 1992

Studies on Cesium Uptake by Phenolic Resins

S. K. SAMANTA and M. RAMASWAMY
PROCESS ENGINEERING AND SYSTEMS DIVISION

B. M. MISRA
DESALINATION DIVISION

BHABHA ATOMIC RESEARCH CENTRE
TROMBAY, BOMBAY 400085, INDIA

Abstract

The selective removal of cesium by phenolic ion-exchange resins from highly salted alkaline radioactive solutions was studied. The resins were synthesized by alkaline polycondensation of phenol, resorcinol, catechol, and a resorcinol–catechol mixture with formaldehyde and characterized for their moisture regain, ion-exchange ($H^+ \rightarrow Na^+$) capacity, and distribution coefficient (K_D) for cesium. The effects of open and sealed curing of the polymers on their properties were studied. The effect of Na^+, NaOH, and Cs^+ concentration on the uptake of cesium by resorcinol–formaldehyde resin was investigated, in particular. The chemical, thermal, and radiation stabilities of the polymers were also studied.

INTRODUCTION

Ion exchange has long been used as an efficient process for the removal of hazardous radionuclides like Cs-137 and Sr-90 from various radioactive waste streams generated at nuclear installations (*1*). From the waste management point of view, the most attractive feature of this technique is its ability to partition radioactive effluents into two separate components, viz., a small volume of concentrated waste containing most of the radioactivity and a relatively large volume of decontaminated effluent suitable for direct discharge. Higher volume reduction and decontamination factors are twin objectives, and efforts are underway for improving existing ion exchangers or developing new ones toward their realization.

Cs-137, due to its long half life (~30 years) and high energy gamma emission, has always been the focus of attention in the treatment of aqueous radioactive wastes. Though ion exchangers have been developed and are in use to remove Cs-137 from neutral or mildly acidic waste streams having low salt content, there are not many materials that can effectively operate in alkaline waste streams with high salt content such as those generated at nuclear fuel reprocessing plants. To be useful in such cases, the ion exchanger must not only be stable and effective in an alkaline environment, but it must also have a high selectivity for Cs^+ in the presence of the large concentrations of competing Na^+ ions ($[Na^+]/[Cs^+] = 10^6$–10^7) that are present in such wastes.

The effectiveness of a phenolsulfonic acid cation exchange resin (Amberlite IR-100) in removing Cs-137 from alkaline sodium salt solutions was first reported by Miller and Kline (*2*). They attributed the high selectivity for Cs^+ of this resin to the presence of phenolic —OH groups which ionize and start functioning only at high pH. The unusually high affinity of phenolic —OH groups for Cs^+ is also reflected in the use of phenolic compounds like 4-*sec*-butyl-2(α-methylbenzyl)phenol (BAMBP) for the solvent extraction of Cs^+ from aqueous solutions at pH values above 10 (*3*). Among the known commercially available cesium selective resins are Duolite CS-100 (phenolic–carboxylic), Duolite ARC-9359 (phenolic–methylenesulfonic), Duolite S-30 (phenolic), Duolite S-761 (phenolic), etc. Studies on processes based on the use of commercial phenolic resins for the treatment of low level (*4*, *5*) as well as high level (*6*, *7*) alkaline wastes are well documented. Several workers have also accomplished the laboratory synthesis of phenolic resins having high affinity for both cesium and strontium (*8–10*). While a phenolic compound like resorcinol is most commonly used to impart cesium selectivity, a chelating compound like iminodiacetic acid is added to give high selectivity for strontium.

In this paper we report the cesium uptake and other properties of some phenolic polymers prepared in our laboratory as per the general method of Pennington and Williams (*11*). Resorcinol, catechol, resorcinol–catechol, and phenol itself were condensed separately with formaldehyde to prepare the resins under study. The effects of initial water content of the reaction mixture and conditions of curing on the properties of the polymer products were also investigated.

EXPERIMENTAL

Materials

Resorcinol and catechol were purified by recrystallization. Phenol, sodium hydroxide, formaldehyde (37% aqueous solution), and other chemicals used for characterization, equilibration, kinetics, and column experiments were of AR quality. Cs-134, used as the tracer, was obtained from Isotope Division, Bhabha Atomic Research Centre.

Synthesis

The general method of Pennington and Williams (*11*) was followed. In all cases the phenolic compound was dissolved in the requisite volume of 6 *N* NaOH to give a phenolic compound/NaOH mole ratio of 1:1.5. This was followed by further dilution with distilled water and then a quantity of 37% formaldehyde solution, sufficient to give a phenolic compound/formaldehyde mole ratio of 1:2.5, was added. The total quantity of water in the final reaction mixture was enough to make the water/phenolic compound mole ratio equal to about 50. After adding the reactants, the mixtures were stirred and left overnight. After 24 h the mixtures were kept in an air oven at 100°C for curing up to 4 days. Variations in the above procedure included a lower water content of the reaction mixture (water/phenolic compound mole ratio changed to 25 as compared to 50 in the above procedure) and curing under sealed conditions so that no water was lost even when the mixture was heated at 100°C for 4 days. After curing, the product polymers, reddish-brown to black in color, were crushed, sieved to −20+50 ASTM mesh size particles, washed, and conditioned by subjecting them to two 1 *N* HCl/0.1 *N* NaOH cycles in a column with a water wash in between. The resins were finally converted to the H^+ form and washed thoroughly with water until they were neutral. The resins were then air-dried for several days and stored in stoppered plastic containers.

Moisture Regain

This was determined by noting the loss in weight on heating of a known weight of air-dry resin in an air oven at 100°C for 24 h.

Ion-Exchange Capacity

For determination of total ion-exchange capacity, a weighed quantity (about 0.5 g) of resin in the H^+ form of known moisture regain was equilibrated overnight with 100 mL of 0.1 *N* NaOH solution containing 5% NaCl. The amount of NaOH consumed in the $H^+ \rightarrow Na^+$ reaction was found by titrating the remaining NaOH in the supernatant with 0.1 *N* HCl solution using phenolphthalein as indicator.

Distribution Coefficient

As a measure of the resin's affinity for cesium and for comparison between various resins, a standard procedure was followed for the measurement of distribution coefficients (K_D). Some resin (0.1 g) of known moisture content was equilibrated overnight with 50 mL of a standard equilibration solution (1.0 *M* $NaNO_3$ + 0.1 *M* NaOH) containing Cs-134 tracer. Two milliliters of the clear supernatant was pipeted out and counted in standard glass tubes using a well-type NaI(Tl) gamma scintillation counter. The distribution coefficient was calculated by using the formula

$$K_D = [(C_i - C_f)/C_f][V/W_r][100/(100\text{-}W_m)]$$

where C_i and C_f are the initial and final counts in cpm/mL, V is the volume of the equilibration solution, W_r is the weight in grams of air-dry H^+-form resin taken, and W_m is the moisture regain (%wt/wt) of the resin.

Rate of Cesium Uptake

This was studied for the resorcinol–formaldehyde (R-F) resins only. To 0.8 g of resin (−80+100 mesh ASTM) taken in a 600-mL polyethylene beaker was added 400 mL of a solution containing 1.0 *M* $NaNO_3$, 0.1 *M* NaOH, 5.13×10^{-5} *M* $CsNO_3$, and Cs-134 tracer. The solution was stirred continuously with a PTFE-coated magnetic bar. At regular intervals, 2 mL of solution was withdrawn using a pipette and its activity determined by gamma counting.

Chemical Stability

Two grams each of the conditioned, H^+-form, air-dry resin was taken in a 250-mL stoppered conical flask and 200 mL of equilibrating solution (1.0 *M* $NaNO_3$ + 0.1 *M* NaOH) was added. The flasks were shaken on a platform shaker daily for 8 h for a total period of 4 days. After this, the resins were separated, washed with distilled water, converted back to the H^+ form with 1.0 *N* HCl, washed, and recharacterized.

TG and DTA

An ULVAC SINKU-RIKO thermal analyzer was used to record the thermogravimetric (TG) and differential thermal analysis (DTA) spectra of some of the resins. A heating rate of 10°C/min was used with about 30 mg of finely ground sample.

Effect of Gamma Irradiation

Samples of air-dried, H^+-form R-F resin were soaked in distilled water and irradiated to different doses with gamma rays from a Co^{60} source at a dose rate of about 0.1 Mrad/h. The maximum dose given was 20 Mrad. After irradiation, the resins were washed, air-dried, and characterized again for their moisture regain, ion-exchange capacity, and distribution coefficient for cesium.

Effect of Solution Composition on K_D

To find the effect of Na^+ concentration, distributions coefficients were determined from equilibrating solutions containing 0.1 *M* NaOH, 8.7×10^{-6} *M* $CsNO_3$, Cs-134 tracer, and varying $NaNO_3$ concentrations to give total Na^+ concentrations of 0.5, 1.0, 2.0, 3.0, and 4.0 *M*. To find the Cs^+ concentration effect, the equilibrating solution was 1.0 *M* $NaNO_3$ and 0.1 *M* NaOH with varying Cs^+ concentrations (added as the inactive salt $CsNO_3$) and Cs-134 as tracer. The range covered was from 0.5 to 10.0 ppm. Similarly, the effect of OH^- concentration was also studied in the range 0.01 to 2.0 *M* with the total Na^+ concentration kept constant at 4.0 *M*, using $NaNO_3$. Inactive $CsNO_3$ (8.7×10^{-6} *M*) and Cs-134 tracer were added as usual.

RESULTS and DISCUSSION

Phenol formaldehyde condensation under alkaline conditions essentially involves electrophilic attack of the highly reactive phenoxide ring by formaldehyde to give methylolphenols as the first step. In the presence of excess formaldehyde, further substitution leads to di- and trimethylol derivatives which, during heat curing, undergo a condensation reaction between methylol groups with the elimination of water to give an infusible, insoluble, amorphous, crosslinked polymer (*12*, *13*). Such a polymer essentially consists of the original phenolic nuclei (with the —OH groups intact) linked together with methylene linkages and has methylol end groups. It is the phenolic —OH groups which are mainly responsible for the ion-exchange properties of such resins. In our study, resorcinol, catechol, and phenol itself were chosen as the compounds for polymerization. On the basis of the above and assuming complete crosslinking, the repeating unit in such polymers would be of the following composition

$$(C_6H_{3-n})(OH)_n(CH_2)_{3/2}$$

where $n = 1$ for phenol and 2 for resorcinol as well as catechol. If all the —OH groups are accessible in the polymer, the theoretically expected $H^+ \rightarrow Na^+$ ion-exchange capacity in alkaline solutions would be 8.9 meq/g for phenol and 15.6 meq/g for the two dihydroxybenzene resins. Still higher capacities are to be expected if one considers the chelating tendency (*11*) of unreacted methylol groups, the possible presence of carboxyl groups (*14*) resulting from the oxidation of methylol groups, and the fact that all possible methylene crosslinks may not be able to form due to steric and other reasons. The results of characterization of the various resins along with the conditions of preparation are presented in Table 1. The various noteworthy features of the results are discussed below.

TABLE 1
Properties of the Phenolic Polymers Studied

Polymer[a]	H_2O/Ph[b]	Curing[c]	Moisture regain (%wt/wt)	Ion-exchange capacity [meq/g(dry)]	K_D^{Cs}[d] [mL/g(dry)]
R-F-1	50	Open	22.1	6.35	19,260
R-F-2	25	Sealed	42.0	6.90	16,150
C-F-1	50	Open	36.3	6.44	120
C-F-2	25	Sealed	41.3	7.10	180
C-R-F-1	50	Open	22.0	6.24	730
C-R-F-2	25	Sealed	39.1	7.69	1,090
P-F-1	50	Open	17.8	4.67	4,000
P-F-2	25	Sealed	24.9	5.26	2,890

[a] R-F, Resorcinol–formaldehyde; C-F, catechol–formaldehyde; C-R-F, catechol–resorcinol–formaldehyde; P-F, phenol–formaldehyde.

[b] H_2O/Ph: Mole ratio of water to phenolic compound.

[c] Curing for 4 days in an air oven at 100°C.

[d] K_D values determined in a solution containing 1.0 M $NaNO_3$ + 0.1 M NaOH + Cs-134 tracer.

In all cases, hard insoluble polymers were obtained. The ion-exchange capacities, although comparable to those found in the polystyrene–divinylbenzene-based conventional cation-exchange resins, are much lower than the theoretical values estimated above. The most likely reason seems to be the inaccessibility of many of the phenolic —OH groups in the cured resin. The low moisture holding capacities and consequent limited swelling of the resins would also seem to support the above explanation. Resorcinol gave a resin with the highest affinity for cesium. This was followed by the phenol resin. It is worth noting here that the conventional sulfonic and carboxylic resins give practically zero uptake of Cs^+ under similar conditions. Catechol resins, although having ion-exchange capacities comparable to those of resorcinol resins, showed very poor uptake of Cs^+. Steric hindrance, caused by the orthodihydroxy structure of catechol, appears to be the reason for this behavior. The cesium uptake by C-R-F resin appears to be primarily due to its resorcinol content. This uptake can presumably be increased further by increasing the molar proportion of resorcinol in the reaction mixture.

Curing under sealed conditions (*14*), ensuring that no water is lost, was attempted to find out if resins with superior properties could be prepared. The water/phenolic compound mole ratio was kept at 25 in this case because there is no loss of water. In the case of open curing, the above ratio was maintained as 50. The results in Table 1 show that sealed curing did result in somewhat higher moisture holding capacities for the resorcinol and catechol resins. The consequent higher swelling and increased accessibility of phenolic —OH groups is also reflected in higher ion-exchange capacities of sealed cured resins, though the effect is marginal. The distribution coefficients for cesium are, however, not very different.

The superiority of open-cured resin was best revealed in experiments designed to measure the rate of cesium uptake. The results are shown graphically in Fig. 1 for various R-F resins prepared with different initial water contents and cured under sealed as well as open conditions. All the preparations show a fast initial uptake of Cs^+ followed by slow attainment

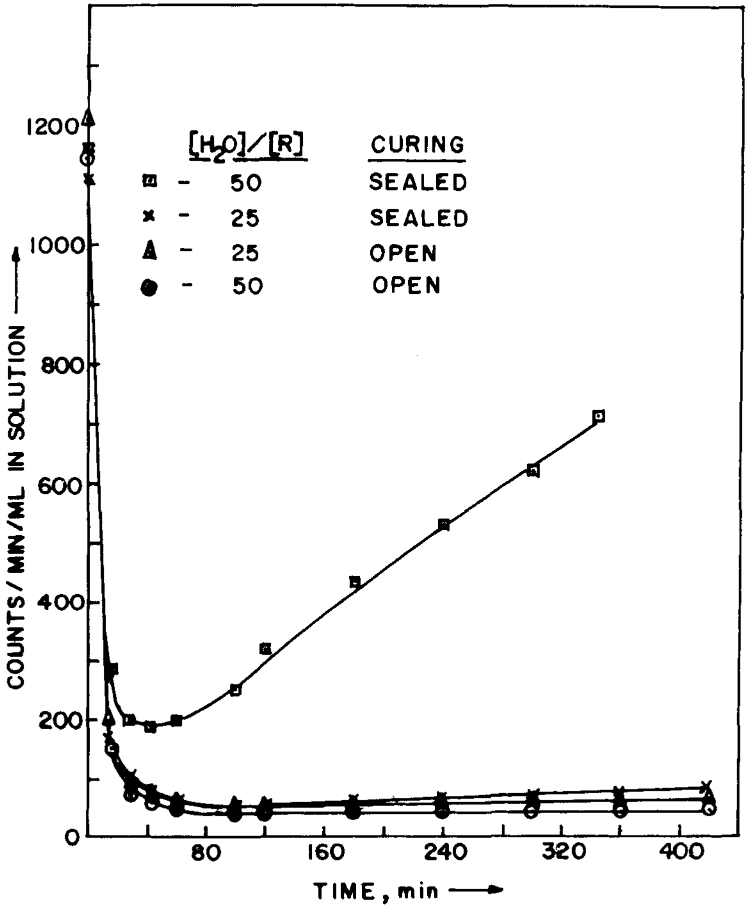
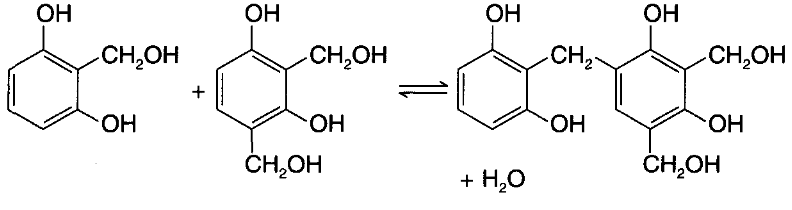


FIG. 1. Rate of cesium uptake by R-F resin.

and then near constancy of the equilibrium value except in the case of R-F resin with higher initial water content in the reaction mixture and cured under sealed condition. In this case the resin appears to release the Cs^+ into the solution slowly. R-F resin prepared as per Pennington and Williams' (*11*) general method was found to be the best. These results show that though sealed curing may give phenolic resins with slightly higher moisture and ion-exchange capacity, there is no improvement in the cesium distribution coefficient and, most important, the products are not stable enough to retain cesium sorbed from alkaline solutions. This lack of stability of sealed cured resins is probably the result of insufficient crosslinking, which in the case of R-F resins proceeds by the elimination of water between methylolphenolic units initially formed, e.g.,



The presence of excess water during sealed curing affects the progress of the above reaction in the forward direction and hence crosslinking is not as extensive as in the case of open curing where the water is allowed to escape.

Though phenolic resins show high affinity for Cs^+ as compared to Na^+, the distribution coefficient for Cs^+ gradually reduces in the presence of increasing Na^+ concentration. This was evident when K_D values for Cs^+ were measured in a 0.1-*N* NaOH solution containing varying amounts of $NaNO_3$. The results are shown graphically in Fig. 2 where log K_D has been plotted against log $[Na^+]$ and a straight line is obtained with a slope of -1.16. Ideally, uni-univalent ion exchange under similar conditions should give a slope of -1.0. However, our result is not very far from the ideal value and, within the limits of experimental error, could be construed as indicating ideal Na^+–Cs^+ ion exchange in this system.

The effect of $[OH^-]$ at constant $[Na^+]$ is interesting. Since phenolic resins contain the weakly acidic phenolic —OH group as the functional group responsible for ion exchange, it is expected that complete manifestation of their ion-exchange properties will be possible only when the —OH groups are ionized to the maximum extent, i.e., in alkaline solutions. Thus, when the NaOH concentration is increased from 0.01 to 1.0 *M* at a constant total Na^+ concentration of 4.0 *M*, the K_D value for Cs^+ increases, but on

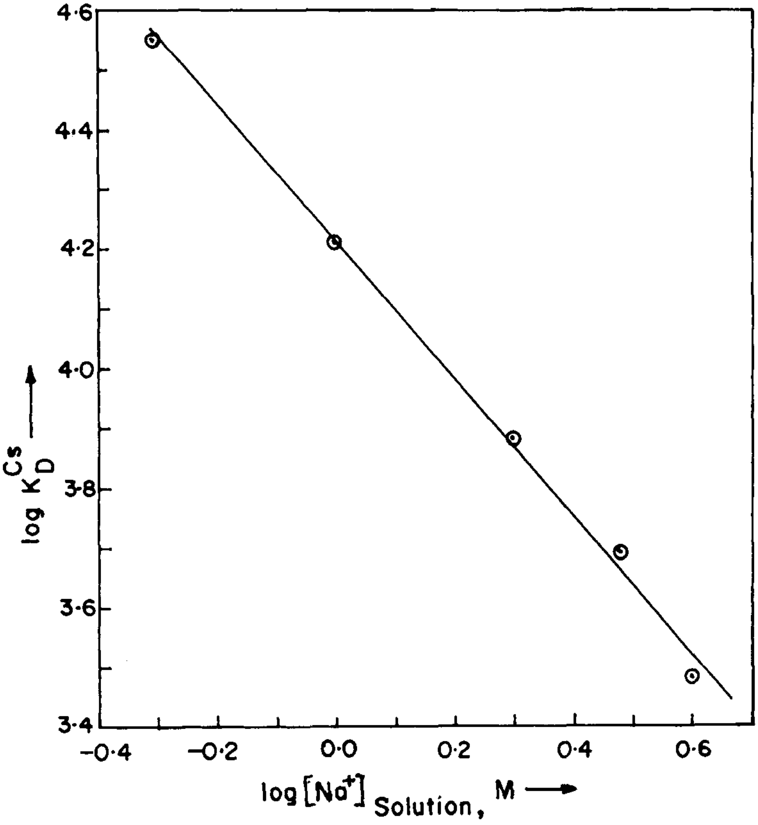


FIG. 2. Effect of Na^+ concentration on K_D^{Cs} of R-F resin.

further increase of NaOH concentration it remains more or less constant, as shown in Table 2. Thus, one may conclude that the benefit derived from increased ionization of the phenolic —OH groups reaches its peak at 1.0 *M* NaOH concentration and a further increase to 2.0 *M* does not give any additional advantage.

Figure 3 shows the effect of $[Cs^+]$ in solution on uptake by the resin. The linearity of the plot suggests that the Freundlich equation is applicable to this system:

$$[Cs^+]_r = K\,[Cs^+]_s^{1/n} \qquad (1)$$

where $[Cs^+]_r$ denotes the cesium concentration in the resin phase (mg/g), $[Cs^+]_s$ that in the solution at equilibrium (mg/L), and K and n are constants.

TABLE 2
Effect of NaOH Concentration on Uptake of Cesium by R-F Resin[a]

NaOH concentration (M)	K_D^{Cs} [mL/g(dry)]
0.01	1900
0.05	2930
0.10	3110
0.50	3330
1.00	4380
1.50	4350
2.00	4270

[a] Resin: Resorcinol-formaldehyde (open cured). Na^+: 4.0 M (total). NaOH: 0.01 to 2.0 M (with $NaNO_3$ to make up to 4.0 M). Cs^+: 8.7×10^{-6} M (as $CsNO_3$). Cs-134: Tracer.

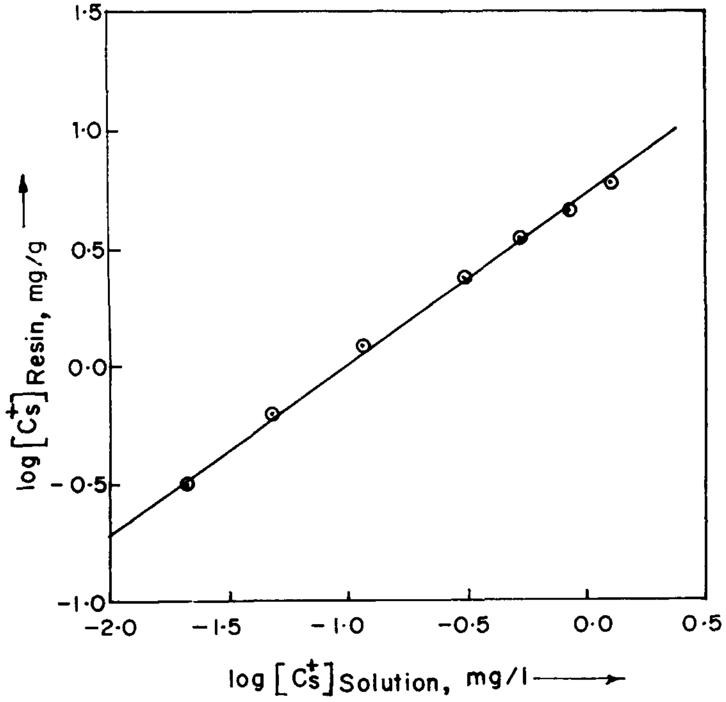


FIG. 3. Effect of Cs^+ concentration on its uptake by R-F resin.

The values of these constants can be calculated from the intercept and slope of the straight line in Fig. 3, and they are found to be $K = 5.01$ and $n = 1.41$.

According to definition, the distribution coefficient, K_D, can be written as

$$K_D = \frac{\text{Cs}^+ \text{ concentration in resin, mg/g}}{\text{Cs}^+ \text{ concentration in solution, mg/mL}}$$

$$= \frac{[\text{Cs}^+]_r}{[\text{Cs}^+]_s} \times 1000 \tag{2}$$

Substituting for $[\text{Cs}^+]_r$ from Eq. (1):

$$K_D = K[\text{Cs}^+]_s^{(1-n)/n} \times 1000 \tag{3}$$

Inserting the values of K and n in Eq. (3), we get the following expression relating the distribution coefficient, K_D, and the equilibrium concentration of Cs^+ in solution, $[\text{Cs}^+]_s$:

$$K_D = \frac{5010}{[\text{Cs}^+]_s^{0.29}} \tag{4}$$

This expression is valid for a solution containing 1.0 M NaNO_3, 0.1 M NaOH, and varying Cs^+ concentration in the range 0.02 to 1.34 mg/L. It also shows that the value of K_D increases with decreasing Cs^+ concentration, as observed in practice.

The chemical stability of the polymers was tested by contacting them with 1.0 M NaNO_3 + 0.1 M NaOH solution for nearly 100 h followed by recharacterization. The stability of open cured resorcinol–formaldehyde resin was particularly good with practically no change in moisture regain, ion-exchange capacity, or distribution coefficient for cesium.

The thermal analysis (TG/DTA) curves for all four types of prepared resins were found to be similar. In each case there is an endothermic loss of moisture at about 100°C (weight loss 10–15%). The broad exothermic peak in the DTA curves, starting at about 200°C, is indicative of oxidative degradation of the polymer matrix. A measure of the thermal stability of the polymers may be indicated by the temperature for 50% loss in weight as calculated from the TG curves. On this basis the polymers can be arranged in decreasing order of thermal stability as follows:

P-F(620°C) > R-F(490°C) > C-R-F(470°C) > C-F(450°C)

TABLE 3
Effect of Gamma Irradiation on R-F Resin[a]

Total dose (Mrad)	Sample condition	Moisture regain (%wt/wt)	Ion-exchange capacity [meq/g(dry)]	K_D^{Cs} [mL/g(dry)]
0	Unirradiated	27.73	6.37	30,000
5	Air-dry sample in water	15.13	6.77	26,000
10	Air-dry sample in water	15.25	6.50	22,000
20	Air-dry sample in water	15.35	6.57	26,500
20	Oven-dry sample, no water	8.81	7.08	13,700

[a] Sample: 5.0 g R-F resin in 6.0 mL distilled water. Source: Co^{60}. Dose rate: 0.1 Mrad/h.

Phenol formaldehyde polycondensates are known to be quite stable to irradiation by gamma radiation (*15*). The effects of gamma radiation to R-F resin in the H^+ form are shown in Table 3. The maximum dose given was 20 Mrad. It is seen that the moisture regain reduces on irradiation but the ion-exchange capacity and K_D values are not affected significantly except in the case of an oven-dried sample irradiated up to 20 Mrad. In this case, moisture regain and K_D values decrease drastically. Normally, one would have expected the radiation effect, if any, to be greater in the case of wet resins because of reactions with radiolytic products of water which are absent in the oven-dried resin. However, radiation may also result in additional crosslinks in a polymer, and this is what seems to happen in the case of the dry resin where the absence of water facilitates the crosslinking reaction. As a result, the moisture regain decreases. The increased crosslinking also hinders the diffusion of Cs^+ ions into the matrix, resulting in a significant drop in K_D^{Cs}.

CONCLUSIONS

Our studies indicate that simple phenolic polymers will be effective in removing cesium from alkaline radioactive wastes even in the presence of large quantities of sodium ions. Phenolic resins based on phenol, resorcinol, catechol, and a resorcinol–catechol mixture were synthesized in our laboratory by condensation with formaldehyde under alkaline conditions and curing by heating under sealed as well as open conditions. The resins with open curing showed better stability. Resorcinol–formaldehyde resin offered the highest uptake of cesium. The cesium uptake was found to proceed according to a near ideal uni-univalent ion-exchange mechanism. Data on cesium uptake under varying cesium concentrations could be fitted satisfactorily by the Freundlich equation. The effect of increasing NaOH in solution was to increase cesium uptake, but not indefinitely. DTA results showed exothermic oxidative degradation above 200°C. Irradiation of water-soaked resorcinol–formaldehyde resin with Co^{60} gamma rays up to 20 Mrad did not significantly affect its properties.

Acknowledgments

The authors thank Dr. P. N. Moorthy for providing the gamma irradiation facility and Dr. S. Brat for arranging the TG/DTA of polymer samples. The encouragement received from Mr. S. V. Kumar is also gratefully acknowledged.

REFERENCES

1. H. G. Swope, "Treatment of Radioactive Wastes," in *Ion Exchange Technology* (F. C. Nachod and J. Schubert, eds.), Academic Press, New York, 1956, p. 458.
2. H. W. Miller and G. E. Kline, *J. Am. Chem. Soc.*, *73,* 2741 (1951).
3. W. W. Schulz and L. A. Bray, *Sep. Sci. Technol.*, *22,* 191 (1987).
4. J. T. Roberts and R. R. Holcomb, *A Phenolic Resin Ion Exchange Process for Decontaminating Low-Radioactivity-Level Process Water Wastes,* ORNL-3036, 1961.
5. J. M. Chilton, *An Evaluation of the Low Level Waste Treatment Plant at ORNL and Suggested Changes in the Design and Operation,* ORNL-5618, 1980.
6. J. R. Wiley, *Ind. Eng. Chem., Process Des. Dev.*, *17*(1), 67 (1978).
7. R. M. Wallace and R. B. Ferguson, *Sci. Basis Nucl. Waste Manage.*, *3,* 75 (1981).
8. M. A. Ebra, R. M. Wallace, D. D. Walker, and R. A. Wille, *Ibid.*, *6,* 633 (1982).
9. J. R. Kaczvinsky, J. S. Fritz, D. D. Walker, and M. A. Ebra, *J. Radioanal. Nucl. Chem.*, *91,* 349 (1985).
10. J. R. Kaczvinsky, J. S. Fritz, D. D. Walker, and M. A. Ebra, *Ibid.*, *116,* 63 (1987).
11. L. D. Pennington and M. B. Williams, *Ind. Eng. Chem.*, *51,* 759 (1959).
12. W. A. Keutgen, "Phenolic Resins," in *Encyclopedia of Polymer Science and Technology,* Vol. 10, Wiley, New York, 1969.
13. K. J. Saunders, *Organic Polymer Chemistry,* Chapman & Hall, London, 1973.
14. J. R. Parrish and R. Stevenson, *Anal. Chim. Acta,* *70,* 189 (1974).
15. K. K. S. Pillay, *J. Radioanal. Nucl. Chem.*, *102*(1), 247 (1986).

Received by editor December 4, 1990