INTRODUCTION

Ion exchange resins are commonly relied upon to purify water, but when the resin itself is exposed to degradative conditions, the resin may contribute impurities as well as remove them. In non-nuclear applications, unless the degradation is severe, the release of material may not be noticed. In purification of coolant from nuclear reactors, however, where the effluent is subsequently irradiated in a neutron flux, the presence of even trace amounts of some impurities eluted from the resins becomes evident through the appearance of induced activities. In addition to contributing to radioactivity problems in the reactor complex, such impurities released into the reactor recirculating system can alter the solution composition required to reduce metal corrosion and/or the radiolytic decomposition of the water. The radiolytic and oxidizing environment within the resin bed, due to the buildup of radioactive materials there, is conducive to resin degradation.

At Savannah River, impurities contributed from the ion exchangers became apparent when $^{35}S$ and sulfate ion were found in the $D_2O$ coolant-moderator and $^{32}P$ was found on surfaces of the aluminum-clad fuel elements. These impurities were traced to the beds of cation exchange resin (sulfonic acid type) that were used at that time to supplement and conserve the mixed beds in the purification of the $D_2O$, which is maintained slightly acid to reduce the corrosion of the aluminum cladding. The $^{35}S$ and $^{32}P$ were attributed to neutron activation of natural sulfur released from the cation resin beds. Use of cation resin beds has since been discontinued to eliminate problems associated with their use.

Other conditions in the reactor also attributed to radiolysis of the deionizer resins are: (1) the prevalence of $CO_2$ in the reactor system, from radiolytic oxidation of organic material released from the
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resin beds, and (2) unusually low degree of utilization calculated for the mixed-bed deionizers. The low utilization suggests that exchange capacity is being exhausted by a mechanism other than removal of ions from the feed.

This work was done to assess the problem of resin radiolysis in Savannah River reactors and in other nuclear reactor coolant systems.

**DISCUSSION**

**BACKGROUND**

The coolant-moderator system of the Savannah River reactors has been described. A bypass stream of D_2_0 is filtered and deionized in a purification cell that consists of 25 ft^3 of resin flanked upstream and downstream by asbestos leaf-type filters. The mixed-bed deionizers contain a mixture in the ratio of one exchange equivalent of "Amberlite" IR-120-D to two exchange equivalents of "Amberlite" IRA-400-OD. Beds of "Amberlite" IR-120-D alone were also once used, but are not in use at the present time.

The rate of flow through a cell is nominally 20 to 80 gallons per minute. Each mixed-bed deionizer processes 3 to 5 million gallons during a service time on-line of three to four months.

The radioactivity in the D_2_0 is comprised of both fission products and neutron activation products. The resin beds retain more than 90% of the radioactivity.

**CALCULATED ESTIMATE OF DOSE**

An estimate was made of the beta-gamma dose received by the deionizer resins from representative amounts of four prominent radionuclides found in the moderator: Na_24, Np_239, Mn_56, and I_133. The calculated dose during normal service is given in Table I, with the source and approximate concentration of each nuclide.

These results show that Na_24 is a major contributor to the dose, and that the total dose is greater than 10^7 rad. The actual dose will be considerably higher because short-lived isotopes were not included in the estimate; these will be abundant in the deionizer feed, which has less than five minutes decay time out of the reactor. As shown in Figure 1, the total dose is sufficient to damage the resin and to reduce the exchange capacity of the mixed-bed deionizers.

**MEASUREMENT OF ON-LINE DOSE RATE**

Measurements of the beta-gamma dose rate to the resins during deionizer operation were made with polyacrylamide dosimeters placed

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at intervals from the top to the bottom of the deionizers. One set of measurements was made with the dosimeters outside the deionizer vessel. A second set was made in which the dosimeters were placed in a well within the deionizer. Measurements made on four deionizers are presented in Figure 2 to illustrate the range of dose rates encountered.

The beta rays were shielded out in all measurements, and it is estimated that the beta dose will be at least as large as the gamma dose. Extrapolation of these results to 100 days of operation confirms that during service the top of the resin bed will receive a total beta-gamma dose of $10^7$ to $10^8$ rad, or enough to damage the resin.

EXAMINATION OF WATER FROM DEIONIZERS

The postulated release of dissolved material from the deionizers into the reactor system could not be confirmed through direct analysis of the deionizer effluent. The concentrations of C, S, and N (the expected elements) were below the limits of detection of the analytical methods available.

However, heavy water that is recovered from the exhausted deionizers several months after they are taken out of service was suitable for analysis. This water had been in contact with the resin for several months, and the soluble material had concentrated in the aqueous phase. Although the results of themselves could not indicate the amount of material the resins had released during service, potential release of soluble material from various types of deionizers could be compared. Had a partially exhausted deionizer been placed back on-line after a period of stagnation, similar material would have entered the reactor system.

The results of analyses of water recovered from several exhausted deionizers are shown in Figure 3. The cation resin beds processed more water to exhaustion than did the mixed resin beds, and consequently they received the greater dose. The cation beds released sulfur compounds, but the mixed-beds were more self-contained. With larger volumes treated a tendency of the mixed-beds to release sulfur compounds is detectable. The mixed-bed that had treated the largest volume, which was continued in use after it had been converted to the D-DCO$_3^-$ form, released as much sulfur as the cation resins. Between 60 and 70% of the sulfur was present as sulfate. In general the pH decreased and the amount of dissolved solids increased with the sulfur concentration. The nitrogen and carbon concentrations were below 5 and 100 ppm, respectively, the maximum sensitivity of the analytical methods used.

EXAMINATION OF EXHAUSTED DEIONIZER RESIN

The most direct information regarding radiation damage to the deionizer resins can be obtained through examination of the exhausted deionizer resins themselves. For this purpose a core of resin has been
removed from a mixed-bed deionizer. The results of this examination will be presented at the Symposium.

CONCLUSIONS

Long-term exposure to radioactivity inherent in reactor coolant purification systems may result in radiation damage to deionizing resins. For many reactor systems, radiation damage will not be acute because of the high threshold dose required.

In the Savannah River reactors, however, two conditions contribute to the attainment of sufficient dose for damage: a high neutron flux, which promotes the production of induced activities, and the presence of aluminum, the precursor of Na$^{24}$, which makes a major contribution to the total dose.

Even when radiation damage occurs, a mixed-bed resin will not pollute the water significantly during normal use, but rather it will retain its ionic degradation products (sulfonic acids and amines). In doing so, however, some of the exchange capacity is consumed, and the efficiency of utilization of the resin bed for water purification is reduced.

Use of only cation or anion exchange resin for reactor coolant purification, rather than a combination of both, is inadvisable because the resins in general will not retain their own degradation products, which will therefore be released to the system.

ACKNOWLEDGMENT

The contribution of the many people in the Works Technical Department of the Savannah River Plant, who made dose rate measurements and obtained samples of resin and water for examination, is gratefully acknowledged.

REFERENCES

2. F. B. Longtin. "Impurities in Moderator of Savannah River Plant Reactors," paper to be presented at this Symposium.
3. E. W. Baumann, to be published.
<table>
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<th>Nuclide</th>
<th>Half-life</th>
<th>Mode of Formation</th>
<th>Nominal Concentration, microcuries/ml</th>
<th>Total Beta-Gamma Dose Absorbed by Resin, rad/cc</th>
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<td>Na$^{24}$</td>
<td>15 h</td>
<td>Al$^{27}$(na)</td>
<td>0.5</td>
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<tr>
<td>Np$^{239}$</td>
<td>2.3 d</td>
<td>U$^{238}$(nγ)U$^{238}$(-β)</td>
<td>0.5</td>
<td>$6 \times 10^6$</td>
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<tr>
<td>Mn$^{55}$</td>
<td>2.6 h</td>
<td>Mn$^{55}$(nγ)</td>
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<td>$1 \times 10^6$</td>
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<tr>
<td>I$^{133}$</td>
<td>21 h</td>
<td>Fission Product</td>
<td>0.05</td>
<td>$1 \times 10^5$</td>
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<tr>
<td><strong>Total</strong></td>
<td></td>
<td></td>
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FIG. 1 EFFECT OF GAMMA RADIATION ON EXCHANGE CAPACITY

FIG. 2 ON-LINE DOSE RATE MEASUREMENTS ON MIXED-BED DEIONIZERS
FIG. 3 COMPOSITION OF WATER FROM EXHAUSTED DEIONIZERS