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WSRC-TR-91-566

CARBON-14 REMOVAL FOR DISPOSAL OF REACTOR DEIONIZER RESINS (U)

by WSRC Contract - W. H. Carlton

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October 31, 1991

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CARBON-14 REMOVAL FOR DISPOSAL OF REACTOR DEIONIZER RESINS (U)

INTRODUCTION AND SUMMARY

Disposal of depleted ion exchange resins from the primary system of the SRS reactors is complicated by the presence of Carbon-14.(1) Because Carbon-14 has a long half-life (5730 years) and high mobility in soils, burial of the resins is no longer a viable option. Consequently some 35 spent reactor deionizers have accumulated that are to be stored aboveground in H-Area for an indefinite period.(2) Spent deionizers containing Carbon-14 will continue to accumulate with operation of the present production reactors and would also accumulate from the proposed heavy water new production reactor.

Removal of the Carbon-14 from the resins would reduce the volume of Carbon-14 bearing waste and enable the resins to be disposed of as low-level waste. Studies at SRS have indicated that the Carbon-14 from reactor primary coolant is mostly retained by the resins as the bicarbonate anion. Thus Carbon-14 removal might be accomplished by an acidification operation with trapping of the carbon dioxide released, for separate disposal. Conversion of the bicarbonate from the resin to barium carbonate, for example, would reduce the volume of waste more than a hundredfold.

Displacement and recovery of Carbon-14 dioxide from reactor coolant deionizers by acid treatment has been reported by the Canadians. This memorandum recommends that a process be developed for Carbon-14 dioxide removal from SRS spent reactor deionizer resins, drawing on the Canadian experience.

DISCUSSION

Background

Production rates of Carbon-14 for different SRS reactor charges have been estimated.[3] Carbon-14 is produced primarily by the (n,α) reaction on Oxygen-17. The current best estimate for the Oxygen-17 concentration in the reactor system is its natural abundance of 0.039%. Carbon-14 is a pure beta emitter with a half-life of 5730 years.

Part of the Carbon-14 generated will be present as dissolved carbon dioxide in the moderator and will be retained as bicarbonate on the ion exchange resins of the deionizers. Small samples of spent deionizer resins have been analyzed for Carbon-14 by acidification of the resin, absorption of the CO_2 generated, and scintillation counting.[4] The resins analyzed contained 1.4E7 and 2.1E7 picocuries of Carbon-14 per mL resin respectively.[5]

Various estimates of the total amount of Carbon-14 in the deionizers have been made. The values from Reference 5, scaled up to 30 ft^3 resin in a deionizer, indicate that each deionizer would contain about 14 Ci of Carbon-14. Lysimeter studies[6] use 0.35 Ci/ft^3 as the source term, which corresponds to 11 Ci per deionizer.

A recent survey[7] of long-lived radionuclide concentrations in deionizer resins gives 20 Ci of Carbon-14 per deionizer as a conservative value. Other significant isotopes with >20-year half-lives were Strontium-90 (3.2 Ci), Cesium-137 (2.1 Ci), Samarium-151 (0.04 Ci), and alpha-emitters (Z>Pb) (0.08 Ci). The data from this survey are plotted in Figure 1, where it is apparent that Carbon-14 is the only long-lived isotope of significant concentration in the deionizers.

The removal and analysis procedures [4,5] assumed that the Carbon-14 is present on the resins primarily as the carbonate or bicarbonate, not as other carbonaceous ions, and not as exchanged carbon on the resin matrix. This is probably a reasonable assumption. Chemical analysis (which did not include Carbon-14) of depleted reactor deionizer resins showed that bicarbonate ion is a primary anion on the resins.[8,9] These studies also revealed that nitrate, which is present in the moderator as an acidifier to control aluminum corrosion, displaces bicarbonate from the resin exchange sites.

The Canadian Experience

Removal of Carbon-14 from spent deionizer resins from the CANDU reactors has been under investigation for about 10 years.[10] The progress of this work has been reported in publications and at conferences. The Canadian intent is to remove the Carbon-14 and immobilize it for separate storage to reduce storage requirements for Carbon-14 waste. The treated resins can then be disposed of as low or intermediate level wastes at lower cost. The Canadians recognize that the separation of the Carbon-14 from the resin wastes offers the potential for Carbon-14 enrichment to produce a marketable isotope, if there is a demand for the product. No attempt has been made to provide a complete bibliography in this memorandum.

The amount of Carbon-14 reported is about 200 μCi per mL resin. A significant proportion of this is presumed to be present as bicarbonate/carbonate.[10] The amount of Carbon-14 is about 10 times that at SRS, probably because no other major anions (like nitrate in the SRS reactors) are present to collect on the resin.

Acid stripping tests showed that more than 99% of the Carbon-14 (as carbon dioxide) was removed with agitated aeration in 2N hydrochloric acid with 2N NaOH the sorbent solution. Demonstration in pilot plant studies was reported to be about to get underway.[12] Further progress will be reported at a current conference.[13,14]

Recommendation

It is recommended that SRS develop a process for treating depleted reactor deionizer resins, building on the Canadian process and engineering experience. The fundamental idea of acid treatment and carbon dioxide sorption is a practical approach to the problem of removing Carbon-14 from the resins. This treatment would enable Carbon-14 separation and storage in a much smaller volume.

Details of a specific process are beyond the scope of this memorandum. However, one possible scenario would be to leave the resins in the present stainless steel deionizer shell and treat them in situ. The deionizers might be filled from the bottom with 5% nitric acid (currently used in RBOF for resin regeneration), which would displace carbon dioxide from the resins according to the equation:

$$\text{Resin-}HCO_3 + HNO_3 \text{ -----> Resin-}NO_3 + CO_2\text{(gas)} + H_2O$$

The carbon dioxide could be stripped from the solution by sparging, and trapped by a sorbent solution or solid.

The standard analytical chemistry procedure of sorbing carbon dioxide in a barium hydroxide solution is used as an example. Barium carbonate is produced by the reaction:

$$Ba(OH)_2 + CO_2 \text{ -----> } BaCO_3\text{(solid)} + H_2O$$

This barium carbonate could be filtered out for storage. The resins could be left in their original containers for disposal as low level waste. The nitric acid could be simultaneously decontaminated and regenerated for reuse by passage through a cation exchange resin in the acid (hydrogen) form.

Conversion of the bicarbonate from the resin into barium carbonate would reduce the Carbon-14 waste volume more than 100-fold. The 30 ft^3 of mixed resin in the deionizer contains about 25 ft^3 of anion exchange resin. Examination of a depleted deionizer[9] showed that about 20% of the resin was in the bicarbonate form, where the Carbon-14 would be held. For the anion resin exchange capacity of 1.1 equivalents per liter, this amounts to 156 equivalents of CO_2, or 30.7 kg of $BaCO_3$. With the $BaCO_3$ density of 4.3 kg/L, the volume of $BaCO_3$ to be stored from one deionizer would amount to 0.25 ft^3. This is 120 times less than the volume of the deionizer resin.

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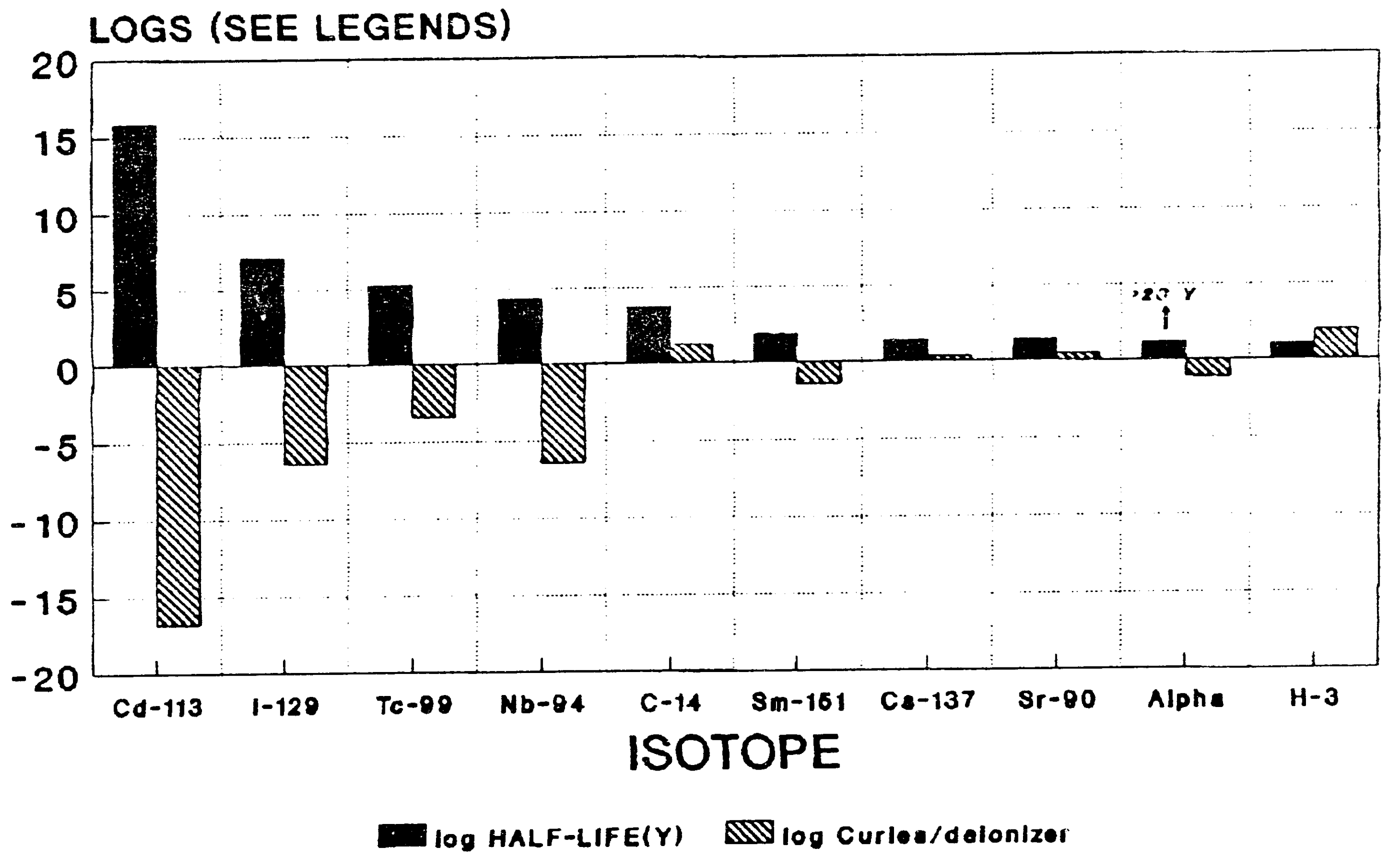
FIGURE 1
LONG-LIVED ISOTOPES IN DEIONIZERS
(From Reference 7)



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