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Disposal of Deionizer Vessels Highly Contaminated with ^{14}C Carbon at SRS

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INTRODUCTION

At the Savannah River Site (SRS), nuclear production reactors used deionizers to control the chemistry of the reactor moderator during their operation to produce nuclear materials primarily for the weapons program. These deionizers were removed from the reactors and stored as a legacy waste with no path to disposal due to the relatively high ^{14}C contamination (i.e., on the order of 20 curies per deionizer for 48-50 deionizers) and the low disposal limit of 4.2 Ci previously established for the Intermediate Level Vault (ILV). The ILV is considered most appropriate facility within which to dispose these

items due to the method of solidifying waste items with cementitious material inside concrete vaults. In previous analyses the ^{14}C ILV disposal limit was established at 4.2 Ci resulting from the use of a very conservative method to analyze the dose received from atmospheric releases of gaseous ^{14}C . This investigation implemented a more rigorous evaluation of the physical and chemical processes influencing the release and migration of gaseous ^{14}C (as CO_2) to obtain a more realistic estimate of atmospheric dose and to determine new ILV disposal limits.

ANALYSIS

The analysis consisted of 2 separate studies, the first (Ref. 1) establishing the chemical constraints in the disposal environment which determine the concentration of $^{14}\text{CO}_2$ in the vapor filled pore space of the waste zone and overlying materials. The second study (Ref. 2) consisted of developing a 1-D numerical model, which utilized a source term consistent with the first study, to evaluate the migration of $^{14}\text{CO}_2$ within the vapor-filled pore space from the waste zone to the land surface. Finally, a recommendation was made to revise the ^{14}C disposal limit for the ILV based on the findings of both studies.

The first study (Ref. 1) indicated that within the disposal environment an equilibrium concentration of ^{14}C is established between the solid and the water, and between the water and air. This equilibrium between the solid and air in a cementitious environment is solubility controlled. In practical terms this means that aqueous ^{14}C concentrations slowly increases when exposed to solid phase ^{14}C waste until the solubility limit is reached, beyond which the aqueous ^{14}C concentration does not increase any more. This calculation of the maximum aqueous ^{14}C concentration is based on values obtained from long-term field studies conducted during

the 1970's at the SRS (Ref. 3). The field studies included four field lysimeters, each containing 1-ft³ of moderator deionizer resin bearing 0.35 Ci ^{14}C imbedded in soil. The leachate chemistry was periodically monitored for nine years. Leachate ^{14}C concentrations ranged from 0.06 to 0.89 $\mu\text{Ci/L}$ over the nine years of monitoring, and the highest value was used in calculating equilibrium concentrations of $^{14}\text{CO}_2$, thus providing a conservative estimate. This value accounted for desorption from resin and re-adsorption to soil or to resins. It is important to note that the resin was simply placed in the sediment and was not enclosed in a stainless steel container, which was in turn entombed within cementitious material, as was the deionizer vessels being modeled. As such, these pore water values represent much greater ^{14}C values than would be expected at the ILV.

The analysis concluded that irrespective of the amount of ^{14}C placed in the ILV waste inventory, the same gaseous concentration occurs in the adjacent air space. This is so because the carbonate equilibrium between the solid (resin) and the pore water, and in turn between the pore water and air space in a cementitious environment is solubility controlled. This concept is expressed

schematically in Figure 1. It is therefore concluded that the aqueous and gaseous concentration of ^{14}C will never exceed those calculated in this investigation irrespective of inventory disposed in the ILV because the limit is based on solubility constraints. The results of the investigation indicate that maximum vapor phase $^{14}\text{CO}_2$ concentration of $1.9\text{E-}07\text{ Ci/m}^3$ is realized in the vapor filled spaces in this environment.

Based on these measured aqueous ^{14}C , as carbonate, values, thermodynamic calculations were conducted to predict the associated maximum vapor phase $^{14}\text{CO}_2$ values that were used in the second study,

In the second part of this investigation (Ref. 2), the $^{14}\text{CO}_2$ flux at the land surface above an ILV was evaluated over the 1,000-year performance assessment (PA) compliance period. The primary mechanism for release of $^{14}\text{CO}_2$ at the land surface is diffusion within the vapor filled pore space of the ILV and overlying cover materials.

RESULTS AND CONCLUSIONS

The results are in terms of an emanation rate of ^{14}C activity at the land surface. There is a rapid buildup to the maximum emanation rate of $1.42\text{E-}8\text{ Ci/year}$ within 0.4 years. This rate continues steadily for the remainder of the simulation.

The maximum emanation rate was used to calculate the dose to the Maximally Exposed Individual (MEI) at a point 100 m from the ILV, which is the most restrictive exposure point. The dose to the MEI at this location was calculated to be $3.83\text{E-}05\text{ mrem/yr}$. which is miniscule compared to the exposure limit defined in USDOE Order 435.1 as 10 mrem/yr .

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A 1-D numerical model was constructed to simulate the diffusion rate and provide results that could be used to determine the atmospheric pathway dose. The model grid was constructed as a node mesh 3 nodes wide by 36 nodes high. The grid extends upward only as far as the closure cap erosion barrier, anticipating that this is the cover thickness that will prevail over the majority of the PA compliance period.

The only radionuclide evaluated in this investigation is ^{14}C , which exists as part of the CO_2 molecule within the model domain. Since CO_2 exists as a gas in the vapor-filled pores and cracks, air was taken to be the fluid within which transport occurs. Air-diffusion was the only transport mechanism simulated in the model and advective air-transport was assumed to be negligible. Transport was allowed to proceed only through air-filled pore space and, therefore, residual pore water was treated as if it was part of the solid matrix material within the flow field. The source term was a constant concentration of $1.9\text{E-}07\text{ Ci/m}^3$ in the air spaces above the resin leachate in the waste zone.

Considering the tiny dose to the MEI calculated in this analysis, no atmospheric pathway limit needed to be applied to the Intermediate Level Vault for ^{14}C . This is justified in that no matter how much ^{14}C is disposed in the ILV disposal environment, only a set, relatively small quantity can move into the air space as gas due to the solubility control limitation. Consequently, the groundwater pathway disposal limit for resin-based ^{14}C in the Intermediate Level Vault became the most restrictive limit among the various pathways, this limit being $2.6\text{E+}06\text{ Ci}$, which easily allows for disposal of all the Reactor Moderator Deionizers.

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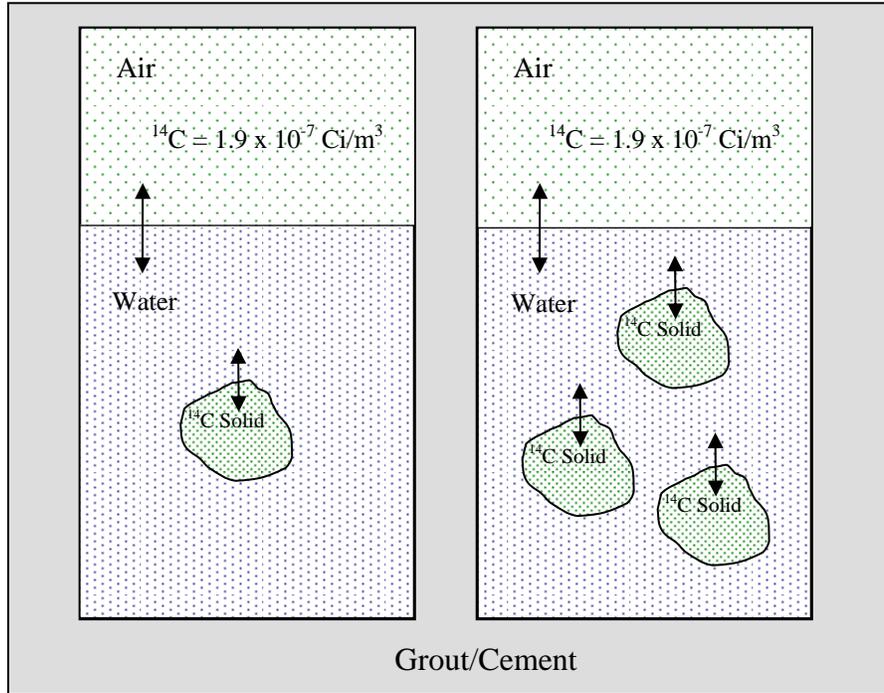


Figure 1. Relationship Between ^{14}C Solid Source and ^{14}C Air Concentration in a Grout/Cement Environment