Removal of Legacy Low-Level Waste Reactor Moderator Deionizer Resins Highly Contaminated with Carbon-14 from the "Waste with no Path to Disposal List" Through Innovative Technical Analysis and Performance Assessment Techniques

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ABSTRACT

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 and due to the relatively high carbon-14 (C-14) contamination (i.e., on the order of 740 gigbecquerel (GBq) (20 curies) per deionizer) were considered a legacy "waste with no path to disposal". Considerable progress has been made in consideration of a disposal path for the legacy reactor deionizers. Presently, 48 – 50 deionizers being stored at SRS have "no path to disposal" because the disposal limit for C-14 in the SRS's low-level waste disposal facility's Intermediate Level Vault (ILV) is only 160 GBq (4.2 curies) per vault.

The current C-14 ILV disposal limit is based on a very conservative analysis of the air pathway. The paper will describe the alternatives that were investigated that resulted in the selection of a route to pursue. This paper will then describe SRS's efforts to reduce the conservatism in the analysis, which resulted in a significantly larger C-14 disposal limit. The work consisted of refining the gas-phase analysis to simulate the migration of C-14 from the waste to the ground surface and evaluated the efficacy of carbonate chemistry in cementitious environment of the ILV for suppressing the volatilization of C-14. During the past year, a Special Analysis was prepared for Department of Energy approval to incorporate the results of these activities that increased the C-14 disposal limits for the ILV, thus allowing for disposal of the Reactor Moderator Deionizers. Once the Special Analysis is approved by DOE, the actual disposal would be dependent on priority and funding, but the deionizers will be removed from the "waste with no path to disposal list."

INTRODUCTION

In the 50's, 60's and 70's, SRS Reactors were operating to produce plutonium, tritium, and other radionuclides. During the operations, deionizers were used in the reactor systems to control the chemistry of the moderator (heavy water). The deionizers trapped an appreciable amount of carbon-14. C-14 is a long-lived radionuclide (half life of 5730 years) which is highly mobile in the environment. As the reactors were shut down, the deionizers (about 50 of them) were taken out of service, packaged and stored at each reactor site.

These mixed bed deionizers were used to control the chemistry of the SRS reactor moderator and coolant to prevent corrosion of the stainless steel reactor components and to control the oxide formation on the aluminum cladding of the fuel and target assemblies. The heavy water moderator and coolant were continually deionized. The deionizers were run to depletion after treating about 28 million liters (7.5 million gallons) of heavy water and then replaced as complete units. Thermal neutrons in the reactor operation transformed the oxygen-17, present as a constituent in the water of the reactor, into C-14 at a rate dependent upon the flux of the reactor. Based on calculations from the neutron reaction and the chemistry of the system a value of 740 Gbq (20 curies) per deionizer was estimated as a bounding C-14 value for each deionizer. Characterization of the waste material contained within each vessel is described in a SRS memorandum (Gibbs, 2005). The other isotopes of concern in the deionizers are tritium, cesium-137, strontium-90 and low concentrations of alpha isotopes. In addition to the relative high amount of C-14 in each deionizer, a radiation field of about 0.01 Gray per hr (1 Rad per hour) has been measured at the container surface. About 50 of these deionizers were removed from the reactors and stored in or near the reactor sites. The resin in a deionizer weighs 2,000 kilograms and the vessel is 1.4 meters (4.5 feet) tall and 1.1 meters (3.5 feet) in diameter, constructed of stainless steel (Fig. 1 and Fig. 2). Each deionizer vessel containing the resin has been dedeuterized, dewatered, and had adsorbents added.

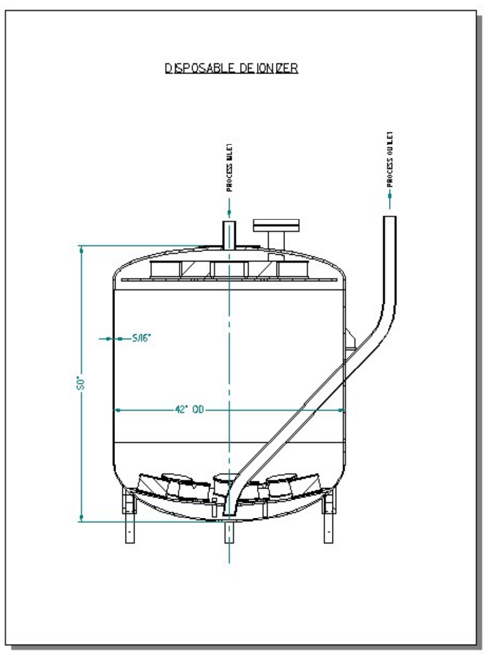

Fig. 1. Reactor deionizer drawing

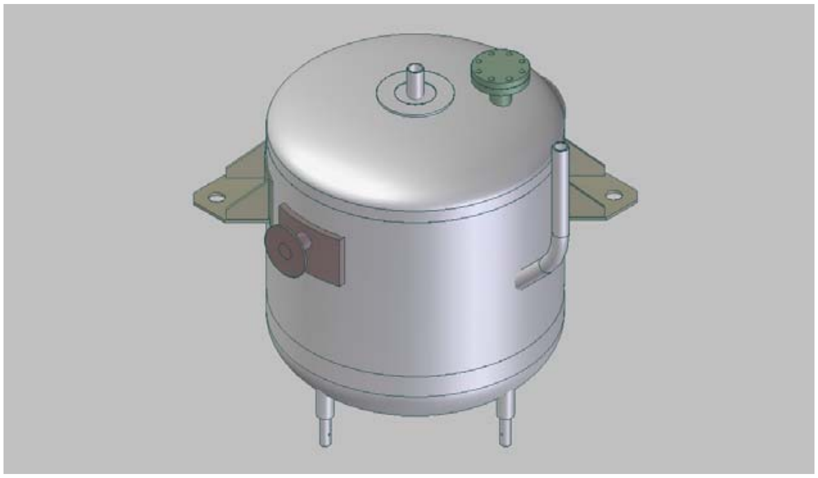

Fig. 2. Reactor deionizer pictorial

Due to the approximately 0.01 Gray per hour (1 Rad per hour) dose rate, the appropriate disposal location for the deionizers was the ILV at the SRS since that was the disposal facility designated for remote handled waste. With the exception of C-14, the other isotopes are within the waste acceptance criteria for disposal at the ILV. The disposal limit for C-14, however, was only 160 GBq (4.2 curies) for the entire vault. At 740 GBq (20 curies) per deionizer multiplied by 50 deionizers, it was clear that this waste greatly exceeded the criteria for disposal. In fact, the deionizers were considered a "waste with no path to disposal" for decades.

PERFORMANCE ASSESSMENT

On September 6, 2005, the Waste Management Area Project achieved another important milestone in the management of radioactive waste. DOE approved the Performance Assessment Special Analysis for the disposal of the 50 legacy reactor moderator deionizer resin columns that have been in storage for decades at SRS. The problem of disposal had gone unsolved for many years and had essentially become accepted as unsolvable to the point that a building was constructed in E-Area to store the deionizer resin columns in shielded casks until some means of treatment or disposal was identified.

In 1992 the Defense Nuclear Facility Safety Board in their study of Low-Level Waste across the DOE complex discussed the Reactor Deionizers in their Recommendation 94-2 that resulted in inclusion of the deionizers in a 1996 DOE Complex-Wide Review of DOE's LLW Management vulnerabilities. Over the years several technical groups were brought in to suggest alternative approaches to the treatment and disposal, but none of the alternatives were deemed to be cost effective or technically proven for implementation. As a result, the deionizers remained in storage at the L-, K-, C-, R-, and P-Reactor sites.

DOE Order 435.1 (Radioactive Waste Management) was implemented in 2000 and the deionizers were identified as a legacy "waste with no path to disposal", but again the plans to develop a disposal path were not implemented due to priority and funding constraints. The work that was proposed for study at the time involved sampling the deionizer columns (an expensive and hazardous undertaking), and disposal schemes that included such exotic techniques as overpacking the deionizers vessels with calcium to fix the C-14 as calcium carbonate, which would then stabilize the C-14 in an insoluble form.

At the beginning of FY2005, the Waste Management Area Project took on the challenge to find a relatively inexpensive, yet feasible method of disposal. With the encouragement of DOE staff, Waste Management Area Project engineers began the task of characterizing the waste forms using reactor physics and calculations from existing records, and put together a team of experts from the Spent Fuel project and the Savannah River National Laboratory (SRNL). The creative juices started to flow.

Solving the disposal problem revolved primarily around gaining a better understanding of the behavior of C-14 in the E-Area Low-Level Waste disposal environment since C-14 was the primary contaminant of concern. These deionizers contain relatively large amounts of C-14 (i.e., on the order of 740 GBq (20 curies) per deionizer) and the disposal limit for C-14 in the ILV was only 160 GBq (4.2 curies) per vault. The ILV is the most robust disposal environment available in E-Area, so it was thought that this facility would be the most likely to be able to safely dispose of this waste. The C-14 ILV disposal limit was based on a very conservative analysis of the air pathway. SRNL undertook studies of the C-14 to gain a better understanding of the gas-phase behavior of the resins encapsulated in the concrete which resulted in a significantly larger C-14 limit. The studies refined the gas-phase analysis to simulate the migration of C-14 dioxide ($^{14}CO_{2(g)}$) from the waste to the ground surface and evaluated the efficacy of carbonate chemistry in the ILV environment for suppressing the volatilization of C-14. SRNL found that irrespective of the amount of C-14 placed in the waste inventory, the same gaseous concentration occurs in the adjacent air space. This occurred because the carbonate equilibrium between the solid and the pore water and in turn between the pore water and air space in a cementitious environment is solubility controlled. Furthermore, since any exposure via the atmospheric pathway is greatly limited by these solubility constraints within the grout encapsulation disposal environment, no atmospheric limit needed to be applied to the Intermediate Level Vault for C-14. In other words, no matter how much C-14 was in the waste inventory, only a set, relatively small amount was able to move into the air space as gas due to the solubility control limitation. Consequently, the groundwater pathway disposal limit for C-14 in the Intermediate Level Vault became the most restrictive limit among the various pathways. This new limit allows for disposal of all the Reactor Moderator Deionizers.

A more detailed explanation of this phenomena is as follows. Kaplan (Ref.4) indicates that irrespective of the amount of C-14 placed in the inventory, the same gaseous C-14 concentration occurs in adjacent air space. This is depicted in Fig. 3. In this schematic, it shows a sealed container with a C-14 solid source material surrounded by water and an air space above the water. Equilibrium is established between the solid and the water, and between the water and air. The equilibrium between the solid and air in a cementitious environment is solubility controlled. In practical terms this means that the aqueous C-14 concentration slowly increases with increases in solid phase C-14 waste until the solubility limit is reach, beyond which the aqueous C-14 concentration does not increase any more. Kaplan (Ref.4) based the maximum aqueous C-14 values released from resins from long-term field studies conducted during the

1970's on the SRS (Fig. 4). In these studies, four field lysimeters were established and 0.28 cubic meter (1 cubic foot) of moderator deionizer resin, bearing 13 GBq (0.35 Curies) C-14 was placed in each lysimeter and the leachate chemistry was periodically monitored for nine years. Leachate C-14 concentrations ranged from 0.002 megebecquerel (MBq) per liter to 0.033 MBq per liter (0.06 to 0.89 µCi/L) over the nine years of monitoring (Ref.5), the highest value was used in these calculations to provide a conservative estimate. This value accounts 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 concrete, as was the deionizer vessels being modeled. As such, these pore water values represent much greater C-14 values than would be expected at the ILV.

Based on these measured aqueous C-14, as carbonate, values, thermodynamic calculations were conducted to predict the associated maximum vapor phase C-14 values that were used in the Performance Assessment Special Analysis performed by Hiergesell and Swingle (Ref. 1). Therefore, the aqueous and gaseous concentration of C-14 will never exceed those used in the performance assessment special analysis, irrespective of inventory disposed in the ILV because the limit is based on solubility constraints. Therefore, there is no need for an atmospheric C-14 limit for the Intermediate Level Vault.

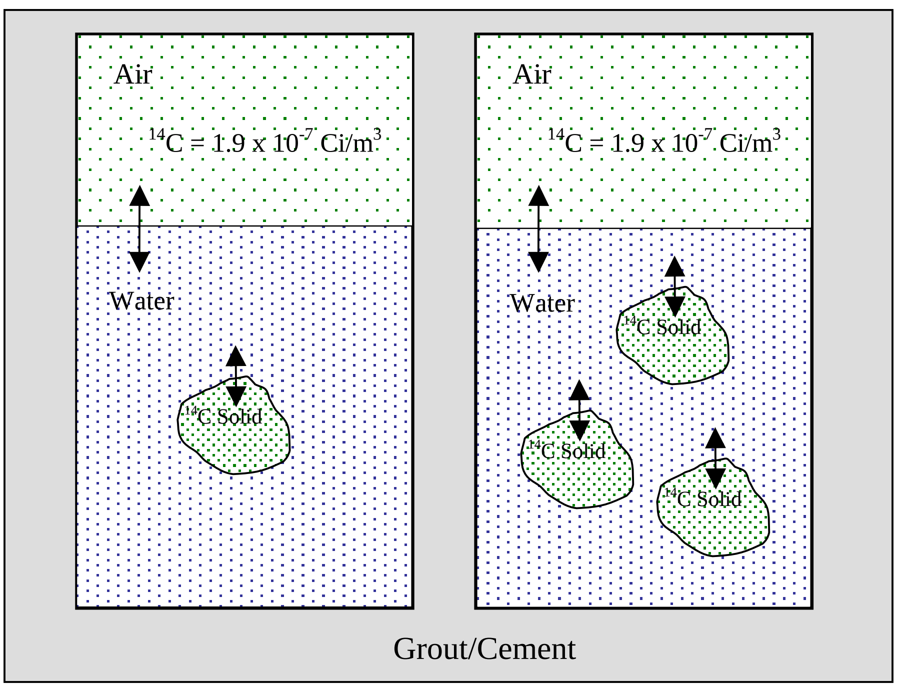

Fig. 3. Relationship between C-14 solid source and C-14 air concentration in a grout/cement

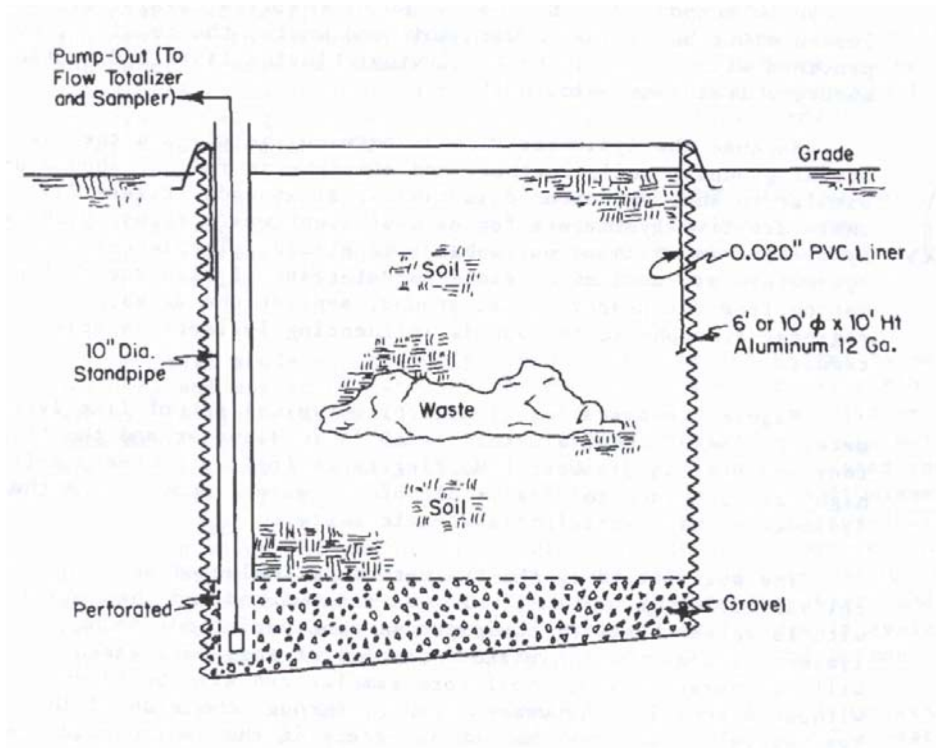

Fig. 4. Lysimeter cross section (waste consisted of 0.028 cubic meter (1-ft^3) of moderator deionizer resin, bearing 13 GBq (0.35 Ci) C-14

The Performance Assessment Special Analysis (Ref. 1) describes the analysis conducted to evaluate the potential magnitude of gaseous release of C-14 from the ILV over the 1,000-year performance assessment (PA) period of interest and the atmospheric pathway dose that this release might result in. The analysis considers the diffusion of C-14 upward from the ILV waste zone through the overlying closure cap material to determine emanation rates at the land surface. The peak emanation rate was then used to estimate the exposure of the Maximally Exposed Individual (MEI) at both the SRS boundary and at a location 100meters downwind from the ILV. The exposure standard for the MEI is stated in USDOE Order 435.1 as 0.1 mSv/yr (10 mrem/yr).

A 1-dimenional numerical model was employed to simulate a bounding (conservative) case to estimate the maximum plausible emanation rate of C-14 at the land surface. The C-14 source term for use in this model was developed in the separate investigation described above and in Ref. 4. The dose received by the MEI was determined using Dose Factors calculated for the Intermediate Level Vault (Ref. 2).

CONCLUSION

The Performance Assessment Special Analysis performed by Hiergesell and Swingle (Ref. 1) was approved by the Department of Energy on September 6, 2005 and at long last, the deionizers have been removed from the “waste with no path to disposal” list. Planning is now underway to remove the deionizers from the reactor areas where they have been stored for many years and ship them to the SRS Low-Level Waste Facility for disposal in the Intermediate Level Vaults.

REFERENCES

1. Hiergesell, R.A., and Swingle, R.F. 2005. Special Analysis: *Atmospheric Dose Resulting from the Release of C-14 from Reactor Moderator Deionizers in a Disposal Environment*, WSRC-TR-2005-00331, Westinghouse Savannah River Company, Aiken, SC.

2. Flach, G.P., and R.A. Hiergesell. 2004. *Special Analysis: Revision of Intermediate Level Vault Disposal Limits*, WSRC-TR-2004-00346, Westinghouse Savannah River Company, Aiken, SC.

3. Gibbs, A. 2005. *Characterization of Moderator Deionizers,* OBU-TRU-2005-00058. Westinghouse Savannah River Company, Aiken, SC.

4. Kaplan, D.I. 2005. *Estimate of Gaseous ^{14}Carbon Concentrations Emanating from the Intermediate-Level Vault Disposal Facility*, WSRC-TR-2005-00222, Rev. 0, Westinghouse Savannah River Company, Aiken, SC 29808.

5. McIntrye, P. F. 1987. 1987 *Monitoring Report for the Defense Waste Lysimeters,* DPST-87-568. Westinghouse Savannah River Company, Aiken, SC.