

Analysis of ^{14}C in the Spent Resin from Wolsong NPP

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1. Introduction

There are four CANDU commercial Nuclear Power Plants in Wolsong site, Korea. The operation of the power reactors produces a number of spent resin waste streams. These originate from clean-up systems and decontamination facilities. The spent resins generated from the moderator and primary heat transfer purification systems comprise the largest fraction of the radioactive resin waste. They are classified as low and intermediate level waste, largely because of their C-14 content; the moderator resins, in particular, contain elevated levels of ^{14}C [1]. In general, the spent resins are slurried out of the service columns and then stored in in-station resin storage tanks [2].

The amount of ^{14}C generated from CANDU nuclear power plants is much higher than those from reactors of other types [3]. The production of ^{14}C occurs in the Moderator (MOD), Primary Heat Transport System (PHTS), Annulus Gas System (AGS), and Fuels. Approximately 94 % of total ^{14}C production is from moderator system, about 95 % of that is removed and retained by the liquid purification ion exchange resin in CANDU Nuclear Power Plant [4]. At present, the ion exchange resin wastes from CANDU power plants are stored untreated at the spent resin storage site in the nuclear power plant facility.

The Nuclear Regulatory Commission (NRC) has required in 10CFR61 that ^{14}C be measured in nuclear power plant radwastes, and has specified ^{14}C concentration limits for class A and C wastes. As preliminary steps for legal disposal of spent resins, we must precisely evaluate the ^{14}C concentration in the spent resins and then separate them into anionic and cationic components to make their preliminary partial decontamination more feasible and less hazard.

Especially, spent resins contaminated with ^{14}C radioisotope which has long half-life of 5,730 years influences the strategy for the disposal of spent. It is recommended that disposal concentration limit of spent resin loaded with ^{14}C is 8 Ci/m³ according to US 10 CFR 61.8. Therefore, the removal of ^{14}C from spent resin and its concentration to solid sorbents become a desirable feature which can be disposed of as conventional low level waste. Acid stripping and thermal stripping methods are under development for the removal of ^{14}C from spent resins [5, 6].

This paper describes the results from a program undertaken to analyze ^{14}C in the spent resins produced from the nuclear operations of Wolsong Nuclear Power Plant. Total 72 resin samples were sampled from the

in-station storage tank at Wolsong Nuclear Power Plant Unit 1. Resin samples were collected from both man-hole (68 samples) and test-hole (4 samples). They were separated into liquid, activated carbon, zeolite, and spent resins which were oxidized and analyzed for ^{14}C . The average concentration of ^{14}C in the mixed exchange resin (cation and anion exchange resin) was determined. A comparison of the ^{14}C concentrations in mixed and anion exchange resin was obtained.

2. Methods and Results

In this section some of the techniques used to resin sampling, ^{14}C analysis from spent resin are described.

2.1 Resin Sampling

The sampler (see Fig.1) was consisted of main control part, finger, and four extension stainless steel tubes: they were connected and disconnected with bolts. The largest of the sampler connected of all is approximately 6 m long. The finger which was opened or closed by take-up of the inner wire has a conical tip to facilitate penetration.

For sampling spent resins from the in-station storage tank 2 at Wolsong Nuclear Power Plant Unit 1, concrete caps of manhole and test-hole were first removed from the in-station storage tank structure to sample the spent resin. The maximum dose rate, in contact with the resin sampler, was about 8 mR/h and the tritium level in the room was about 2.9 DAC during sampling.



Fig. 1. Spent Resin Sampler for in-station storage tank within Wolsong a NPP.

2.2 Separation

Prior to ^{14}C analysis, the liquid in which the resin had been shipped was carefully decanted into a bottle. Each shipping solution was filtered through Mesh 20 (0.5 mm) and Mesh 12 (0.71 mm) to remove precipitated materials (activated carbon and zeolite) and/or resin fines. After the shipping liquid and precipitated materials were removed, each resin was analyzed by liquid scintillation counter (LSC) to determine the ^{14}C concentration.



Fig. 2. Separation of resins from mixed samples.

To avoid the confusion associated with reporting activities based on wet resin weights, all resin samples were weighed prior to analysis, and this weight was converted to a volume using the bulk resin density. All ^{14}C concentrations reported in this study are given in terms of activity per volume of wet resin (Bq/m^3).

Mixed resin samples were separated with density into cation and anion resins by Fluidized Bed. To perform this separation, the resin sample was transferred into a separation bed using distilled deionized water. The solution was stirred to facilitate separation, resulting in the anion beads floating on the top layer and the cation beads remaining in the bottom layer. The anion fraction was removed from the solution and the cation fraction was drained.

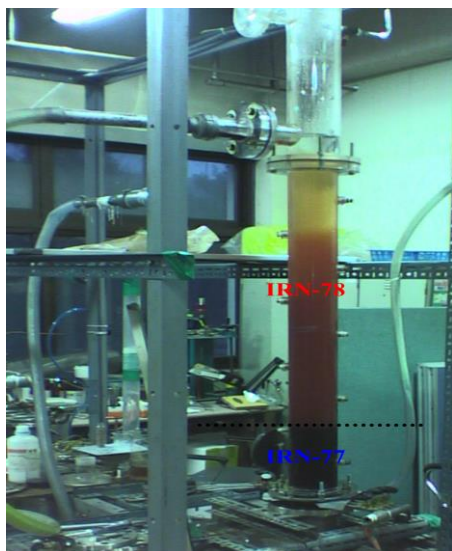


Fig. 3. Separation of cation and anion resins.

2.3 Oxidation with Sample Oxidizer

The appropriate weight of mixed and anion resins was determined of 0.8 g for combustion with a sample oxidizer (PerkinElmer, M307). As shown in Fig. 4, the combustion section consists of a combustion flask, ignition basket, flask plug, flask heater with thermostat adjustment, and a small storage cylinder for O_2 delivery. Each resin sample to be oxidized was transferred into a COMBUSTO-CONE which in turn is placed into the oxidizer's removable ignition basket, mounted to the flask plug assembly. The combustion system burned the sample in an oxygen atmosphere.

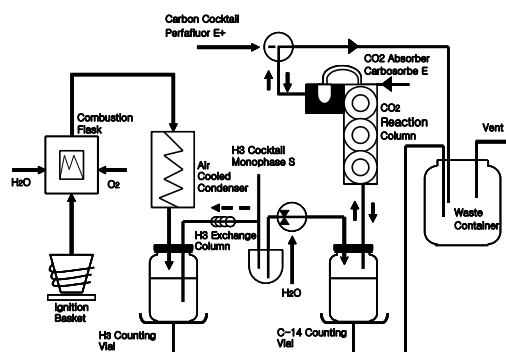


Fig. 4. Schematic diagram of sample oxidizer (PerkinElmer, M307).

The resin sample was ignited by the ignition basket, which is a platinum heating coil capable of developing high temperatures. The combustion time is set for 4 minutes. During the combustion process, all isotopes of carbon, including ^{14}C , were oxidized to gaseous carbon dioxide. The carbon dioxide passed through the tritium collection system and into the ^{14}C collection system. The carbon dioxide was trapped in a column filled with a carbon dioxide absorbent. This chemical, CARBOSORB E, trapped the radioactive carbon dioxide and formed a carbamate, to be flushed into the ^{14}C counting vial using the ^{14}C scintillator, PERMAFLUOR E+ as a rinsing media.

2.4 Analysis of ^{14}C

The distribution of ^{14}C concentration with elevation in the Wolsong NPP's spent resins sampled from manhole is shown in Fig. 5. The concentration of ^{14}C at the bottom of tank was about $170 \text{ GBq}/\text{m}^3$ for the cation/anion mixed resin. The ^{14}C concentration at the elevated level from 20 to 40cm was $380 \text{ GBq}/\text{m}^3$, which was the highest. The concentration of ^{14}C was generally decreased with elevation.

A comparison of the ^{14}C concentrations of the cation/anion mixed resins sampled from manhole and test-hole is given in Fig. 6. The average concentration of ^{14}C was $460 \text{ GBq}/\text{m}^3$ from test-hole and $53.1 \text{ GBq}/\text{m}^3$ from man-hole. The differences in the results obtained by two sampling suggest some inhomogeneity

exists in the resin storage tank. It is possible that resins from test-hole were originated in the higher proportion of ^{14}C in the syste

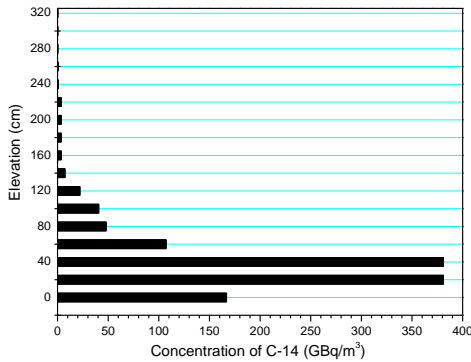


Fig. 5. Concentration of ^{14}C in the Wolsong NPP's spent resins sampled from manhole.

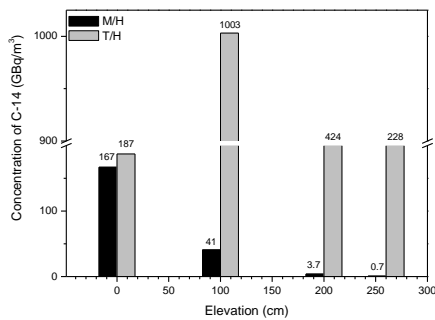


Fig. 6. Concentration of ^{14}C in the Wolsong NPP's spent resins sampled from man-hole and test-hole.

The concentrations of ^{14}C for the cation/anion mixed and anion resins are given in Fig. 7. The average concentration of ^{14}C was 546 GBq/m³ for cation/anion mixed resin and 1,431 GBq/m³ for anion resin. The concentration of ^{14}C for the anion resins were higher than those found for the cation/anion mixed resins; the ^{14}C concentration in anion resin was approximately 2 times higher than in the cation/anion mixed resin. This explains that most of the ^{14}C is in the anion resin because cation-to-anion ratio of Wolsong NPP's spent resin is almost 1: 1.

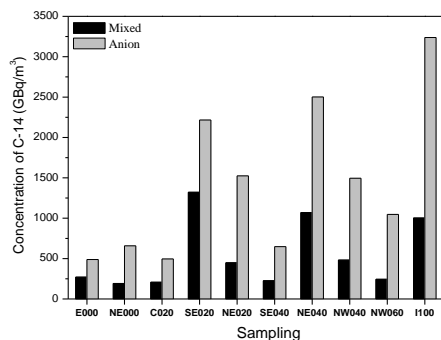


Fig. 7. Concentration of ^{14}C in the Wolsong NPP's spent resin; mixed resin and anion bead fraction.

3. Conclusions

Spent resins were sampled from in-station resin storage tank 2 at Wolsong Nuclear Power Plant Unit 1. Commercially available grain samplers were referred for resin sampling. Resin samples were collected from both man-hole (68 samples) and test-hole (4 samples). The maximum dose rate, in contact with the resin sampler, was about 8 mR/h and the tritium level in the room was about 2.9 DAC. Each resin sample was oxidized with sample oxidizer (Perkin Elmer, M307). The average concentration of ^{14}C in the cation/anion mixed resin was 460 GBq/m³ from test-hole and 53.1 GBq/m³ from man-hole. The ^{14}C concentration in anion resin was approximately 2 times higher than in the cation/anion mixed resin. The ^{14}C concentration was generally decreased with elevation. In the result, we must separate them into anion and cation resins and then eliminate the ^{14}C from the anion resins for legal disposal of spent resins.

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