Measurement of Carbon-14 Activity in Spent Ion-exchange Resin of Wolsong Nuclear Power Plant

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Abstract

Measurement of spent resin activity was initiated in 2004 in order to develop the C-14 removal technology for safe disposal. As part of this program, spent resins were sampled and measured in the in-station resin storage tank 2 at Wolsong Nuclear Power Plant Unit 1. At the time of sampling, the resins had been in storage tank from 3 to 23 years. Total 72 resin samples were sampled, which were collected from both man-hole (68 samples) and test-hole (4 samples) in the in-station resin storage tank 2. They were separated into liquid, activated carbon, zeolite, and spent resin. The spent resins were oxidized with sample oxidizer and analyzed for C-14. Ten of collected mixed resin samples were separated by density into cation and anion resins using a sugar solution. The C-14 concentration in anion exchange resin was approximately 2 times higher than in the mixed resin. The average concentration of C-14 in the cation/anion mixed exchange resin was 460 GBq/m^3 from test-hole and 53.1 GBq/m^3 from man-hole. We have found that concentration of C-14 in the spent resin is about from 0.4 to 1,321 GBq/m^3. So it could be a problem, when dispose of at a repository, since there is a disposal limit of 222 GBq/m^3. This means we should develop the C-14 removal technology.

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 C-14 [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 C-14 generated from CANDU nuclear power plants is much higher than those from reactors of other types [3]. It is estimated that CANDU nuclear power plants produce about 2.6 TBq/GWe (70 Ci/GWe) per year. Because CANDU nuclear power plant produce up to 10 m^3 spent resin, it is necessary to know the activity of the spent resin when we dispose of at the repository. The production of C-14 occurs in the Moderator (MOD), Primary Heat Transport System (PHTS), Annulus Gas System (AGS), and Fuels. Approximately 94 % of total C-14 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 in the nuclear power plant facility.

Korea regulates the concentration of C-14 in the radwaste as 222 GBq/m^3 (6 Ci/m^3). The Nuclear Regulatory Commission (NRC) has also required in 10CFR61 that C-14 be measured in nuclear power plant radwastes, and has specified C-14 concentration limits for class A and C wastes as 296 GBq/m^3 (8 Ci/m^3).

If activity of spent resin exceeds the C-14 concentration limit of disposal, the activity should be reduced for safe disposal. As preliminary steps for legal disposal of spent resins, we must precisely evaluate the C-14 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.

This paper describes the results from a program undertaken to analyze C-14 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 C-14. The average concentration of C-14 in the mixed exchange resin (cation and anion exchange resin) was determined. A comparison of the C-14 concentrations in mixed and anion exchange resin was obtained.

Experiment

1. Resin Sampling Technique

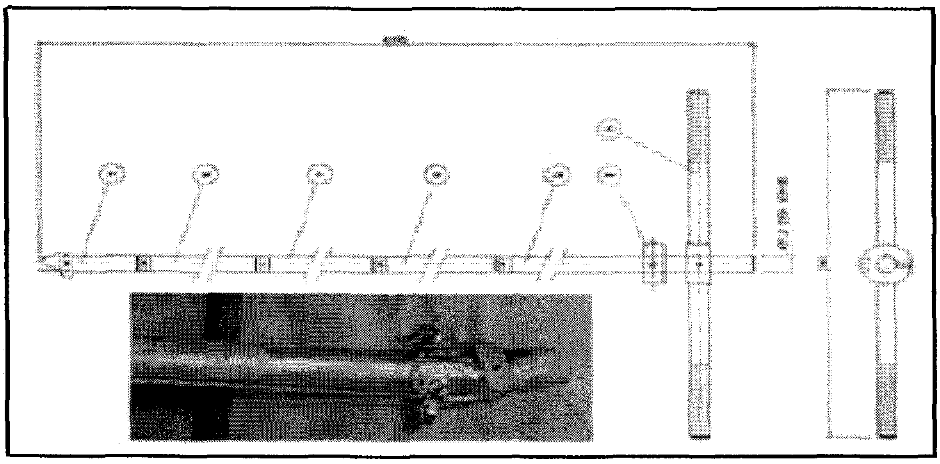

Fig. 1 Spent Resin Sampler for in-station storage tank within Wolsong NPP

Commercially available grain samplers were referred for 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 length 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, an overall length of approximately 5.5 m was required. The 15 cm finger was, therefore, equipped with four extensions each approximately 1.5 m long. Concrete caps of manhole and test-hole on the in-station storage tank structure were first removed to sample the spent resin. The finger was assembled with the extension pieces, and then, the sampler being lowered through the manhole or test-hole into the in-station storage tank. 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. Lead blankets were used to minimize dose uptake during sampling.

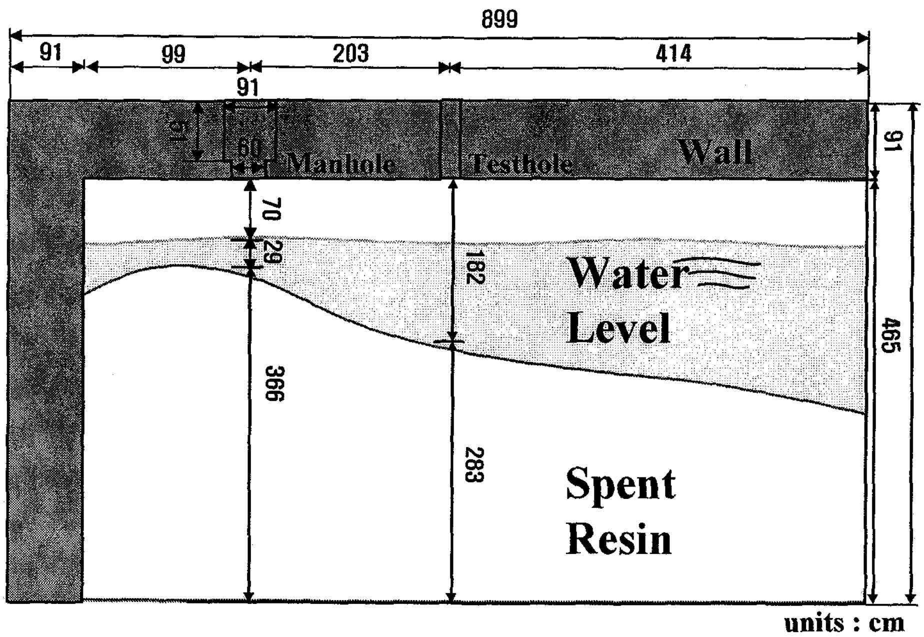

Fig. 2 Schematic of spent resin storage tank 2 at Wolsong NPP Units 1

The in-station storage tank 2 at Wolsong Nuclear Power Plant Unit 1 was contained about 145 m^3 of resin, 20 m^3 of granular activated carbon, and 11 m^3 of zeolite on December 2002 (the contents in the tank were at the 85 % level; see Fig. 2). No resin shipments have taken place from this tank since the Unit 1 start-up in April 1985. It was desirable to estimate the age distribution of resin in the tank in order to correlate the measured radioactivity data and age. The elevation of resin was estimated considering the resin levels in the Units 1 tank 2 to be at the 85% levels (i.e. water above the resin bed was assumed to represent 5% of the level in the tank). As shown in Fig. 3, the axis of the inserted sampler would be laterally displaced from the vertical axis of the tank. This assessment was based on the physical dimensions and location of the sampler within the tank during sampling, and the dimensions and shape of the tank. The resin elevation in the tank was estimated to be 340 cm. Based on the overall resin storage duration of 17 years for the Unit 1 tank 2, and taking into account the elevation of discharged resins, the average resin accumulation rates in these tanks were estimated to be 20 cm/y. These rates were then applied to the elevations shown in Fig. 3 to estimate the corresponding age of resin samples.

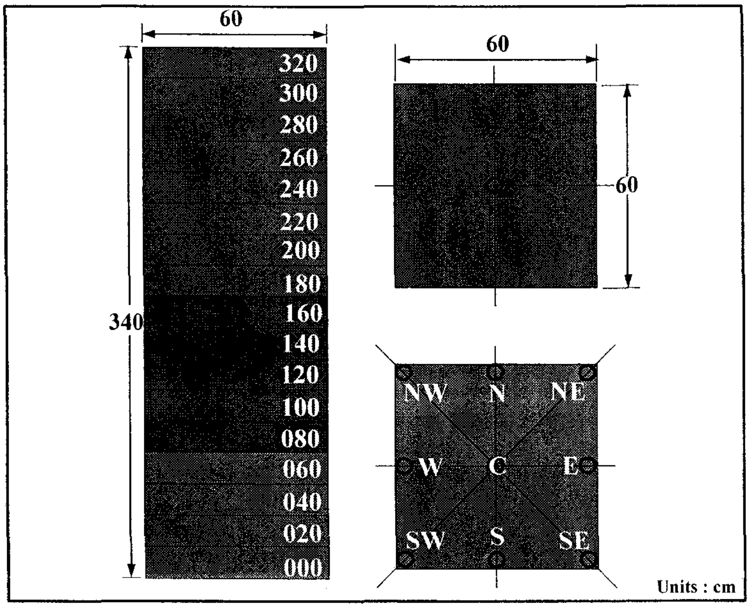

Fig. 3 Sampling position of Wolsong NPP spent resin from manhole.

2. Separation

Prior to C-14 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 zeoite) 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 C-14 concentration.

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 C-14 concentrations reported in this study are given in terms of activity per volume of wet resin (Bq/m^3).

Ten of collected mixed resin samples were separated by density into cation and anion resins using a sugar solution. To perform this separation, the resin sample was transferred into a 250 ml separation funnel using 10 ml of distilled deionized water (DDW) and added 190 ml of 40% (w/w) sugar (sucrose) solution. 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 sugar solution and the cation fraction was drained from the separation funnel. The cation and anion fractions were rinsed with 10 ml of DDW.

3. Oxidation with Sample Oxidizer

The appropriative 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.

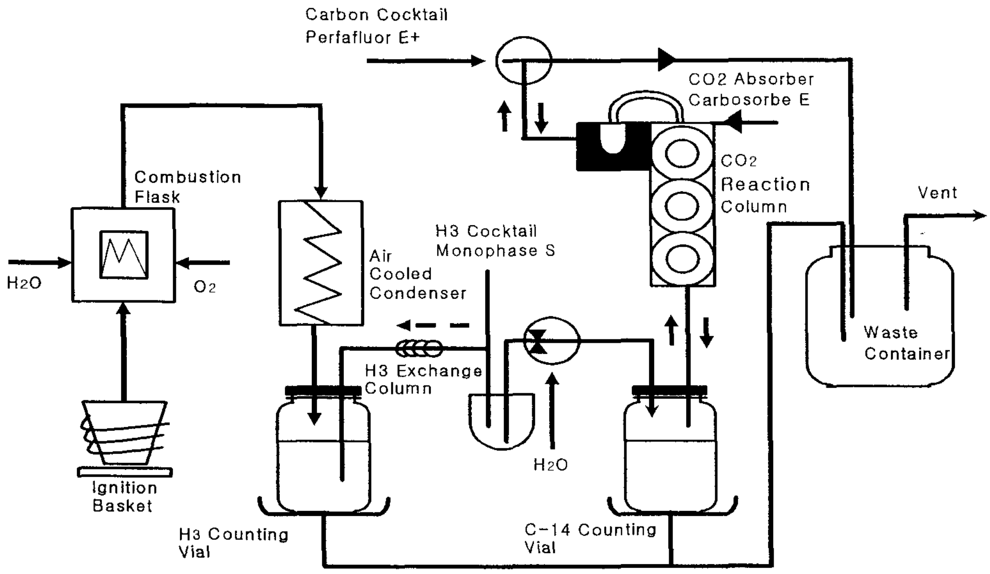

Fig. 4 Schematic diagram of sample oxidizer (PerkinElmer, M307)

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 C-14, were oxidized to gaseous carbon dioxide. The carbon dioxide passed through the tritium collection system and into the C-14 collection system. The carbon dioxide was trapped in a column filled with a carbon dioxide absorbent. This chemical, CARBO-SORB E, trapped the radioactive carbon dioxide and formed a carbamate, to be flushed into the C-14 counting vial using the C-14 scintillator, PERMAFLUOR E+ as a rinsing media.

Carbon-14 measurement

The distribution of C-14 concentration with elevation in the Wolsong NPP's spent resins smpled from manhole and testhole is shown in Table 1 and Table 2, respectively. The average concentration of C-14 at the bottom of tank was about 170 GBq/m^3 for the cation/anion mixed resin. The average C-14 concentration at the elevated level from 20 to 40 cm was 380 GBq/m^3, which was the highest. The concentration of C-14 was generally decreased with elevation.

Table 1. Horizontal and vertical C-14 concentration distribution of manhole.

Unit : GBq/m^3

Elevation	C	E	W	S	N	SE	NE	SW	NW
M000	89	270	52	259	218	174	192	181	67
M020	207	78	126	59	167	1321	448	707	313
M040	37	448	144	41	566	224	1069	418	481
M060	78	33	59	22	71	23	374	59	243
M080	7	152	34	19	30	-	-	-	-
M100	1	12	3	6	190	-	-	-	-
M120	4	20	0.4	6	83	-	-	-	-
M140	1	8	3	27	2	-	-	-	-
M160	1	3	1	11	6	-	-	-	-
M180	4	-	-	-	-	-	-	-	-
M200	3	-	-	-	-	-	-	-	-
M220	5	-	-	-	-	-	-	-	-
M240	1	-	-	-	-	-	-	-	-
M260	0.4	-	-	-	-	-	-	-	-
M280	0.4	-	-	-	-	-	-	-	-
M300	0.4	-	-	-	-	-	-	-	-
M320	89	-	-	-	-	-	-	-	-

Table 2. Horizontal C-14 concentration of testhole

Unit : GBq/m^3

Elevation	C
T000	187
T100	1003
T200	426
T300	229

A comparison of the C-14 concentrations of the cation/anion mixed resins sampled from manhole and test-hole is given in Fig. 5. The average concentration of C-14 was 460 GBq/m^3 from test-hole and 53.1 GBq/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 C-14 in the system.

The concentrations of C-14 for the cation/anion mixed and anion resins are given in Fig. 6. The average concentration of C-14 was 546 GBq/m^3 for cation/anion mixed resin and 1,431 GBq/m^3 for anion resin. The concentration of C-14 for the anion resins were higher than those found for the cation/anion mixed resins; the C-14 concentration in anion resin was approximately 2 times higher than in the cation/anion mixed resin. This explains that most of the C-14 is in the anion resin because cation-to-anion ratio of Wolsong NPP's spent resin is almost 1: 1.

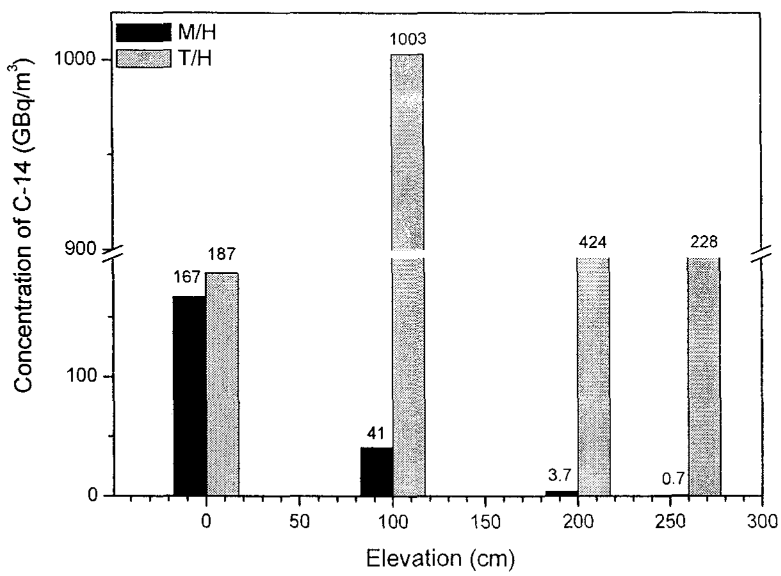

Fig. 5. Concentration of C-14 in the Wolsong NPP's spent resins sampled from man-hole and test-hole

Conclusion

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 C-14 in the cation/anion mixed resin was 460 GBq/m^3 from test-hole and 53.1 GBq/m^3 from man-hole. The C-14 concentration in anion resin was approximately 2 times higher than in the cation/anion mixed resin. The C-14 concentration was generally decreased with elevation.

We have found that concentration of C-14 in the spent resin is about from 0.4 to 1,321 GBq/m^3. So it could be a problem, when dispose of at a repository, since there is a disposal limit of 222 GBq/m^3. In the result, we must separate them into anion and cation resins and then eliminate the C-14 from the anion resins for legal disposal of spent resins.

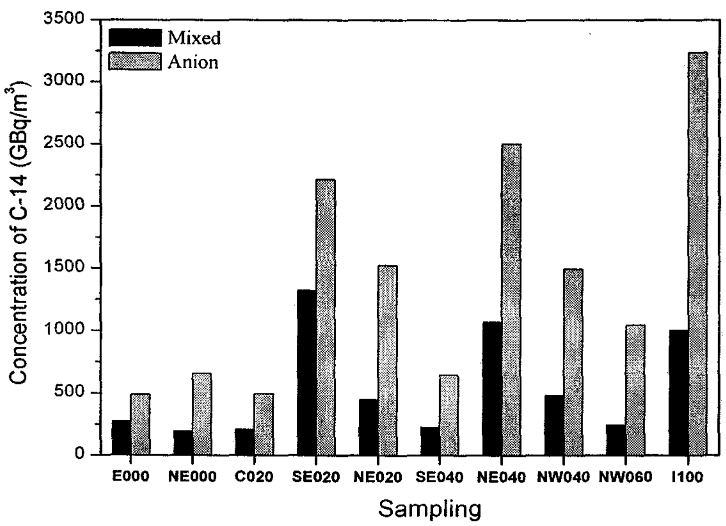

Fig. 6. Concentration of C-14 in the Wolsong NPP's spent resin; mixed resin and anion bead fraction.

References

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