Carbon-14 Analysis of Wolsong Nuclear Power Plant's Spent Resins

a Young-Ku Choi, a Jang-Sik Won, b Dong-Hoon Kim, a Ho-Yeon Yang, a Kyoung-Doek Kim a Nuclear Environment Tec. Institute, KHNP Co Ltd., P. O. Box 149, Yuseong, Daejeon, kmvp09@hotmail.com b Wolsong Nuclear Power Site NPP#1, KHNP Co Ltd., 260 Naa-ri, Yangnam-meon, Gyeong-ju, Gyeong-buk

1. Introduction

There are four CANDU commercial power reactors in Wolsong, Korea. The reactors are all owned by Korea Hydro & Nuclear Power Co. Ltd. (KHNP). The operation of the power reactors produces a number of spent resin waste streams. These originate from cleanup systems and decontamination facilities. In general, the spent resins are slurried out of the service columns and then stored in in-station resin storage tanks.

The spent resins generated from the moderator and PHT purification systems comprise the largest fraction of the radioactive resin waste. They are classified as intermediate level waste, largely because of their C-14 content; the moderator resins, in particular, contain elevated levels of C-14[1]. A program to sample spent resins was initiated in 2004 in order to develop the C-14 removal technology for the treatment of them. As part of this program, spent resins were sampled from instation resin storage tank #2 at Wolsong Nuclear Generating Station Units 1. At the time of sampling, the resins had been in storage at in-station tank for periods ranging between 3 and 23 years.

This paper describes the results from a program undertaken to analyze of C-14 in the spent resins produced from the nuclear operations of Wolsong Nuclear Power Plant. The 72 resin samples were sampled from the in-station storage tank at Wolsong Nuclear Generating Station Units 1. Resin samples were collected from both man-hole (68 samples) and test-hole (4 samples) at the top of in-station resin storage tank. They were separated into shipping liquid, activated carbon, zeolite, and spent resin. Spent resins were oxidized and analyzed for C-14.

2. Methods and Results

2.1 Resin Sampling Technique

Commercially available grain samplers were referred for resin sampling. The sampler (see Fig.1) consists of five stainless steel tubes; they were connected and disconnected with bolts. The finger which was opened or closed by take-up of the inner wire has a conical tip to facilitate penetration. The largest of the sampler is approximately 6 m long. Concrete caps of man-hole and test-hole were first removed from the storage structure to access the spent resin. This exposed the upper surface of the resin with shipping liquid. The resin sampler was then inserted into the resin bed and a sample obtained. Lead blankets were used to minimize dose uptake during sampling. The maximum dose rate, in contact with the resin sampler, was about 8 mR/h. For sampling resin from the in-station storage tanks at Wolsong, 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. For sampling, the finger was assembled with the extension pieces, and then, the sampler being lowered through the man-hole into the in-station storage tank. During sampling, the tritium level in the room was about 2.9 DAC.

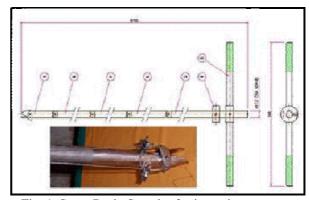


Fig. 1. Spent Resin Sampler for in-station storage tank within Wolsong NPP

2.2 Separation

Prior to the start of the 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 associated shipping liquid and precipitated materials were removed, each resin was analyzed by liquid scintillation counting (LSC) to determine C-14. To avoid the confusion associated with reporting activities based on wet resin weights, all resin samples were weighed prior to the start of 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/m³). Ten resins of collected resin samples were separated by density into anion and cation bead components 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 with a glass rod to facilitate separation, resulting in the anion beads floating to the

top layer and the cation beads remaining in the bottom layer. The anion fraction was removed from the sugar solution by aspiration of the beads using a pipet, and the cation fraction was drained from the separation funnel. The cation and anion fractions were rinsed with 10 ml of DDW.

2.3 Oxidation with Sample Oxidizer

The weight of unseparated and anion fraction resins was determined of 0.8 g for combustion in a sample oxidizer (PerkinElmer, M 307). The combustion section consists of a combustion flask, ignition basket, flask plug, flask heater with thermostat adjustment, and a small storage cylinder for O2 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. 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. 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.

2.4 Carbon-14 analysis

A comparison of the C-14 concentrations of the unseparated resins sampled from man-hole and test-hole

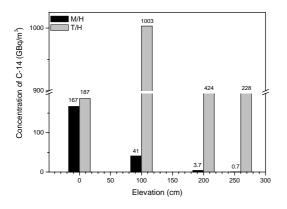


Figure 2. Concentration of C-14 in the Wolsong NPP's Unseparated Spent Resin; man-hole and test-hole sampling

is given in Fig. 2. The average concentration of C-14 on the unseparated resin was 460 GBq/m³ from test-hole and 53.1 GBq/m³ 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.

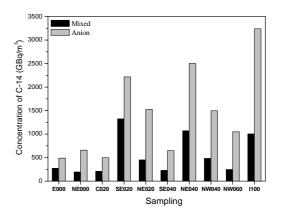


Figure 3. Concentration of C-14 in the Wolsong NPP's Spent Resin; mixed resin and anion bead fraction.

The concentration of C-14 for the unseparated resins and anion bead components is given in Fig. 3. The average concentration of C-14 was 546 GBq/m³ for unseparated resin and 1431 GBq/m³ for anion bead component. The concentration of C-14 for the anion bead components were higher than those found for the unseparated resins; the C-14 concentration in anion bead components was approximately 2 times higher than in the unseparated resin. This is explained that the cation-to-anion ratio of Wolsong NPP's Spent Resins is 1: 1.

3. Conclusion

Spent resins were sampled from in-station resin storage tank #2 at Wolsong Nuclear Generating Station Units 1. Resin samples were collected from both manhole (68 samples) and test-hole (4 samples) at the top of in-station resin storage tank. The average concentration of C-14 on the unseparated resin was 460 GBq/m³ from test-hole and 53.1 GBq/m³ from man-hole. The C-14 concentration in anion bead components was approximately 2 times higher than in the unseparated resin.

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[1] D. L. Moir, K. J. Ross, J. D. Chen, A. W. Tarr, E. L. Bialas, W. A. Boivin, D. C. Doern, D. L. Hoffman, and J. P. M. Ross, Determination of 14C in Spent Moderator Ion-Exchange Resin from Bruce Nuclear Generating Station A, J. Radioanal. & Nucl. Chem., Vol. 185, p. 69, 1994.