Radionuclide Inventory of Past Radioactive Waste in NPPs

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Dry active waste arising from PWR- and PHWR-type NPPs have to be shipped to a permanent disposal facility. For permanent disposal of LILWs, the radionuclide inventories should be evaluated using the scaling factor, which was determined between 2004 and 2008. Therefore, it is necessary to examine the applicability of the scaling factor to the dry active waste (DAW) drums generated prior to 2004. The samples that were assayed were obtained from DAW drums generated prior to 2004, and were pretreated to make-up a 20 ㎖ solution, separated radiochemically, and counted by radiation counters. In addition, using the database, the applicability of the scaling factor to the old DAW drums generated before 2004 was examined through a comparison between the measured radioactivity for a specific β-emitting radionuclide in the past DAW drums and the estimated radioactivity by the scaling factor. As a result, the radioactivity ratios of specific radionuclides were distributed within the identified data ranges of the scaling factor, which showed great significance at a 95% confidence level.

1. Introduction

Since the completion of a disposal site, the Korean Radioactive Waste Agency (KORAD) has accepted approximately 7,000 waste drums each year. It is necessary to quickly develop and secure technologies and facilities that can prove the compliance of the waste acceptance criteria for permanent disposal. This is to prevent a shortage of drums disposed of at the disposal site owing to the delay of the current technological development. To permanently dispose of the waste drums to the disposal site, it is necessary to comply with the disposal regulations based on the KORAD "Waste Delivery Criteria" and the Korea Atomic Energy Agency Notice "Waste Acceptance Criteria". In the case of all waste drums that have been generated during the past (prior to 2004), it must be verified whether the Waste Delivery/Acceptance Criteria for the drums is satisfactory. Therefore, the validity of the scale factor must be quickly verified through the establishment of a "Radionuclide inventory DB" for the past waste drums. The past waste drums are being stored in an interim storage of each Nuclear Power Plant site and currently reach approximately 55,000 drums [1].

In this study, the samples to be assayed were obtained from DAW drums generated prior to 2004, and were analyzed through radiochemical pretreatment and radionuclide separation [2]. Using the “database, the applicability of the scaling factor to the past DAW drums was examined through a comparison between radionuclide concentrations analyzed directly in the past DAW drums and radionuclide concentrations estimated by the scaling factor.

2. Experimental

2.1 Sampling of the past waste drums

The samples of the super compressed dry active waste were transferred to the Radioactive Waste Form Examination Facility (RWFEF) in the Korea Atomic Energy Research Institute (KAERI) from the Hanul NPP site. RWFEF is a facility used to identify the inventory of the specific radionuclides. In order to analyze the concentrations of some radionuclides, the RWFEF opened each drum to get representative samples, and sent to the radionuclide analysis laboratory.

2.2 Radiochemical analysis of the radionuclides

The samples, which were weighed and ashed in a furnace, were put into a 125 ㎖ digestion vessel. The ashed samples were dissolved completely using a HNO_3/HCl/HF mixed solvent. Subsequently, they were placed in a microwave oven (Ethos PLUS Milestone Microwave Digestion), and irradiated with a maximum energy of 400 W for 20 to 50 minutes in order to be dissolved in the mixed solvent. A representative sampling is very important to reduce the sampling error from the inhomogeneous distributions of the nuclides. Sequentially, a single portion of the digested sample was sequentially used throughout the procedures before the γ-spectroscopic measurement. The samples were classified into volatile difficult-to-measure (DTM) nuclides such as ^{3}H, ^{14}C, ^{99}Tc, and non-volatile nuclides. ^{3}H and ^{14}C in the radwastes were extracted into gaseous $^{14}CO_2$ and liquefied HTO through a wet oxidation and distillation method simultaneously [3]. ^{99}Tc was separated and purified with an ion exchange resin and counted by Gas flow Proportional Counter. ^{55}Fe from the completely dissolved samples were separated using an ion exchange resin and extraction chromatography [4].

2.3 Application of Scaling Factor

A scaling factor is a general method to estimate DTM nuclides in the waste drums. In order to develop the scaling factor, analytical samples were extracted representatively among the LILWs generated from the PWR and PHWR-type NPPs between 2004 and 2008. Eventually, the first scaling factor was derived through a radiochemical and statistical analysis. The applicability of the scaling factor to the past DAW drums generated prior to 2004 was examined through a comparison between the measured radioactivity for a specific β-emitting radionuclide in the past DAW drums and the estimated radioactivity by the scaling factor.

3. Results and Conclusions

In the past drums, the radionuclide concentrations of ^{60}Co and ^{137}Cs showed wide radioactivity distributions in the ranges of 2.1E-1 to 1.4E+3 Bq/g and 2.9E+0 ~ 1.3E+4 Bq/g, respectively. The concentration distributions of those radionuclides in the PWR-type NPP was over ten-fold higher than the case of the PHWR-type NPP. Among the target beta emitting radionuclides, ^{3}H and 14C radionuclides were distributed within the ranges of 6.6E+0 ~ 1.2E+3 and 6.6E-2 to 3.3E+0 Bq/g, respectively. ^{55}Fe and ^{99}Tc radionuclides as representatives of activation and fission products were distributed within concentration ranges of 1.2E+2 ~ 2.5E+2 and 1.9E-2 ~ 4.7E-2 Bq/g, respectively.

From these radiochemical analysis results, the applicability of the scale factor was confirmed through a comparison of the radioactivity ratio of the target radionuclide to the key radionuclide between the scaling factor results and radionuclide concentration results generated prior to 2004. As a result, both radioactivity ratios of ^{3}H, ^{14}C, ^{55}Fe, and ^{99}Tc were distributed within the identified data ranges, which had great significance at a 95% confidence level.

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