ASSESSMENT OF THE IMPACT OF DECAY CORRECTION IN THE DOSE-TO-CURIE METHOD FOR LONG-TERM STORED RADIOACTIVE WASTE DRUMS

K. H. Hwang, S. C. Lee, S. H. Kang, K. J. Lee
Korea Advanced Institute of Science and Technology (KAIST)

T. W. Kim, K. D. Kim, M. J. Song
Nuclear Environment Technology Institute (NETEC)

ABSTRACT

Regulations and guidelines require the detailed information about the characteristics of radioactive waste drums. Therefore it is important to know the accurate radionuclide inventory of radioactive waste. However, evaluation of radionuclide concentrations in the drummed radioactive waste is difficult. The gamma scanning method can be used for the assessment of gamma emitting radionuclides. In Korea, a radioactive waste assay system has been operated at Korean nuclear power plant (KORI site) since 1996 and the consolidated scaling factor (SF) concept has played a dominant role in the determination of difficult-to-measurable (DTM) radionuclide concentration. However, if the radioactivity in the radwaste drum is extremely low or high, the radionuclide activity of the drum cannot be evaluated properly. Gamma scanning is also time-consuming and imposes an economic burden to the utilities. In order to overcome these difficulties, dose-to-curie conversion (DTC) method, which is supplementary to the gamma scanning method, has been used to assess the radionuclide activity of radwaste drums in KORI site. Since then, a research for the improvement of existing methodology has been performed in 2000. A further detailed research has been progressed for more reliable assessment and continuous renewal of data since 2003. The scope of research is expanded and planned such as following. First, assay target nuclides (22 radionuclides) and NPPs (all of the Korean NPPs including Pressured Heavy Water Reactors) are increased. In addition, more reliable sampling (actual waste samples) and measurement techniques have been performed and utilized. Conformation of correlation pairs based on Korean analyzed data will be provided. Through these amendments, accuracy and reliability of assessment will be improved.

There are long-term stored LLW drums more than 60,000 in Korea NPPs. Some of them cannot be analyzed by a gamma scanning method. It is, thus, necessary to apply the dose-to-curie method for these LLW drums. Due to the difficulty of broad radiochemical analysis for long-term stored drums, current radiochemical analyzed database will be used on the

assumption that the trend (SF and fractional abundance of gamma emitter) is similar when it was generated. Therefore, SF and fraction abundance need to be corrected, which is based on the historical records. In this study, the impact of decay correction in the activity determination is performed based on the assumed historical records. From the case study, it is concluded that DTC method needs to involve the decay correction, which is based on the historical record. Therefore, decay correction algorithm is reflected to the DTC method.

INTRODUCTION

In Korea, the Enforcement Decree of the Korean Atomic Energy Act (Articles 88) requires the detailed information about the radioactive waste package. The measurement of concentration and total activity of radionuclide contained in radwaste drum is very important for the accurate and efficient management of radioactive waste in NPPs. An established waste characterization program in KORI site measures the concentration of gamma-emitting radionuclides directly using the gamma assay system and/or indirectly using the dose-to-curie conversion method. The concentration of other relevant radionuclides is estimated indirectly by relating DTM radionuclides to other Easy-To-Measurable (ETM) radionuclides. SFs are generated by use of sample data that are gathered from the radiochemical analyses of waste samples collected from the different waste stream. The determination of activity is conducted by radionuclide assay system and SF method.

However, some problems are remained. Some important radionuclides are not included in this program. Other PWRs except NPPs of KORI site and PHWRs are not considered. It also needs to more number of samplings and reliable sampling procedures for the improvement of reliability. For that reason, it is in progress to establish an assay system using more reliable methodology for updating the performance of Korean nuclear waste management. Korean hydro & nuclear power co. Ltd. (KHNP) organizes the overall project with partial cooperation with Korea power engineering company Inc. (KOPEC) [tomographic gamma Scanner system], Korea atomic energy research institute (KAERI) [radiochemical analysis of samples] and Korea advanced institute of science and technology (KAIST) [scaling factor evaluation and dose-to-curie conversion program].

In this paper, the status of radionuclide activity determination method in KORI site and new system are briefly introduced. In addition, sampling analyzed data set is assessed for the determination of target gamma-emitting nuclides and fractional abundance in DTC method. The impact of decay correction is also assessed from the viewpoint of the variation of fractional abundance and activity of each gamma emitting radionuclide in a typical radwaste drum.

Status of Radionuclide Activity Determination Method in Kori System and New System

At the end of 1993, Korea electric power research institute (KEPRI) organized the overall project to design and install the radioactive waste assay system with partial cooperation with KAERI and KAIST [1, 2]. With careful considerations, KORI NPP was selected as a candidate site for assay system. Radioactive waste assay system was installed and started operation during the mid 1996. In this research, activity determination was conducted by radioactive waste assay system for the key nuclides and SF method for DTM nuclides. A further detailed research has been progressed for more reliable assessment and continuous renewal of data since 2003. The project of new radioactive waste assay system is started in 2003 and will be finished at the end of 2005. Principal changes of new system compared to KORI system are assay target DTM radionuclides and NPPs. Comparison between KORI system and new system is summarized in Table I. [3]

Table I. Comparison of KORI System and New System

	KORI System	New System
Gamma assay method	SGS (Segmented Gamma Scanning) & DTC method	TGS (Tomographic Gamma Scaling) & DTC method
Target DTM radionuclide	H-3, C-14, Fe-55, Co-60, Ni-63, Sr-90, Nb-94, Tc-99, Cs-137 and gross alpha	H-3, C-14, Fe-55, Ni-59, Ni-63, Co-60, Sr-90, Nb-94, Tc-99, I-129, Cs-137, [Pu-238, 239, 240, 241], Am-241, [Cm-242, 244] and gross alpha
Target NPPs	KORI site	All Korean NPPs (16 PWRs and 4 PHWRs)
Waste types and representative sampling	A) Spent filter (RCS Letdown filter) B) Concentrates (Radwaste Evaporator Concentrate) C) Spent resin (Primary Mixed Bed Resin) D) DAW (Dry Active Waste)	A) Spent filter (RCS Letdown filter) B) Concentrates (Radwaste Evaporator Concentrate) C) Spent resin (Primary Mixed Bed Resin) D) DAW (Dry Active Waste) E) Sludge

Analysis of Sampling Radiochemical Analysis Data Set

The representative sampling is in progress for the evaluation of scaling factor and application of DTC method. In this study, cumulative sampling data set is analyzed and it is focused on gamma radionuclide data set for the determination of fractional abundance of gamma emitting radionuclides. Information of sampling data set is summarized in table II. It is nearly impossible to obtain the reliable representative sampling for overall radwaste drums, especially, for the long-term stored drums. Therefore, fractional abundances of gamma nuclides are derived from the current sampling data set and should be corrected by the use of the historical record, which is mainly the time period for decay correction.

Table II. Sampling Data Set of Gamma Emitting Radionuclides

<table>
<tr><th>Waste Type</th><th>Detailed Classification</th><th>Sampling Site
(Number of Sample)</th><th>List of Nuclide</th></tr>
<tr><td>DAW (18)</td><td>Cotton (7)
Paper (5)
Vinyl (6)</td><td>Kori (6)
Ulchin (9)
Yonggwang (3)</td><td rowspan="5">Co-60
Cs-137
Ag-110m
Ce-144
Co-57
Co-58
Cr-51
Cs-134
Fe-59
Mn-54
Nb-95
Sb-125
Zr-95</td></tr>
<tr><td>Concentrates (5)</td><td>-</td><td>Kori (2)
Ulchin (2)
Yonggwang (1)</td></tr>
<tr><td>Spent resin (5)</td><td>High level (2)
Low level (3)</td><td>Kori (1)
Ulchin (1)
Yonggwang (3)</td></tr>
<tr><td>Spent filter (1)</td><td>-</td><td>Ulchin (1)</td></tr>
<tr><td>Sludge (4)</td><td>-</td><td>Kori (2)
Ulchin (1)
Yonggwang (1)</td></tr>
</table>

Establishment of the Decay Correction Method in DTC Method and its Application

DTC method estimates the concentration of gamma-emitting radionuclides in a waste package based on the dose rate. The dose rate is measured at a specified distance and geometrical position from the waste package. This information, combined with the data on the relative abundance of the gamma-emitting radionuclides, is used. The radionuclide-specific CFs are determined using the MCNP-4C in this study [4, 5].

The basic equations are same as following:

$$C_{total} = \frac{D}{\sum_{i=1}^{n} f_i \times CF_i} \qquad \text{(Eq. 1)}$$

$$C_i = C_{total} \times f_i \qquad \text{(Eq. 2)}$$

where:

C_{total} = Total activity (mCi)
C_i = Radionuclide specific activity (mCi)
D = Average surface dose rate (mR/hr)
f_i = Fractional abundance of the i_{th} radionuclide
CF_i = Radionuclide specific dose-to-curie conversion factor for a given waste drum configuration (mR/hr/mCi)

Decay correction can be done from the historical record. Modified equations are same as following.

$$f_i' = \frac{f_i \times e^{-\lambda_i T}}{\sum_{i=1}^{n} f_i \times e^{-\lambda_i T}} \quad \text{(Eq. 3)}$$

$$C_{total}' = \frac{D}{\sum_{i=1}^{n} f_i' \times CF_i} \quad \text{(Eq. 4)}$$

$$C'_i = C_{total} \times f_i' \quad \text{(Eq. 5)}$$

where:
C'_{total}, C'_i = Modified total activity and radionuclide specific activity, respectively (mCi)
f'_i = Modified fractional abundance of the i_{th} radionuclide
λi = Decay constant (sec^{-1})
T = Decay correction time (= Measured time – Generation time) (sec)

From the survey of status of long-term stored drum in Korean NPPs, it is concluded that there exist 22 different radwaste drums, which can be applicable to DTC method as a complement method for gamma assay system. They are classified by the drum type, contained radwaste type and solidification process. To show the impact of decay correction in the determination of gamma activity, a case study is conducted in a typical drum. In this case, concrete drum type II containing the concentrate is selected. In Ulchin site, concentrate bottom is solidified using the cement without the process of drying and stored in concrete drum type II until 1998. Since then, CWDS (concentrate waste dying system) is operated. In this case, cement-solidified concentrate bottom is selected as a target of case study and summarized in Table III. Drum modeling and related input data are briefly introduced in Figure 4.

Table III. Summary of Input Data for Radwaste and Drum

Related Site	Ulchin
Drum type	Concrete drum type II (Inner volume : 350L)
Radwaste	Concentrate bottoms
Solidification	Cement
Generation time	88'~98'

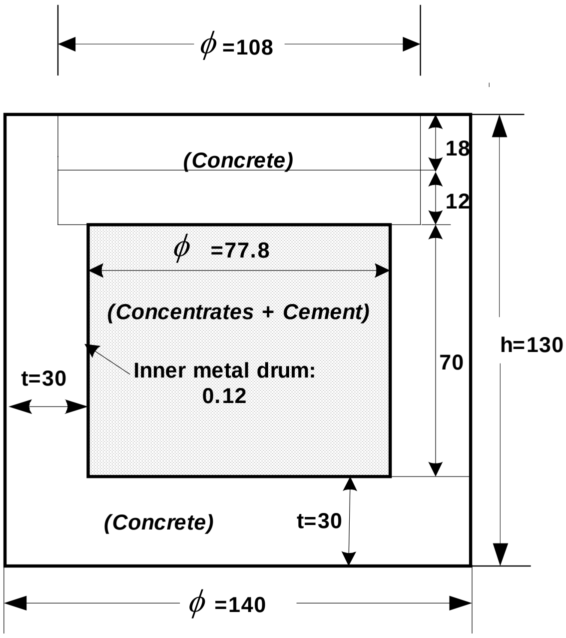

Fig. 1. Modeling of Concrete Drum Type II in Ulchin NPP

Results and Discussion

Prior to analysis of the impact of decay correction in DTC method, selection of gamma radionuclides and determination of fractional abundance are conducted for concentrate bottom in the data set. It is restricted to the gamma radionuclide, which its average fractional abundance is higher than 0.01(1%). Fractional abundance and CF values are summarized in Table IV.

Table IV. Fractional Abundance and CF

Gamma Radionuclide	Initial Fractional Abundance (%)	Conversion Factor (mR/hr/mCi)
Co-60	48.2	1.01E-02
Cs-137	19.7	5.44E-04

Ag-110m	16.6	6.12E-03
Cs-134	6.4	1.82E-03
Mn-54	3.5	1.37E-03
Nb-95	3.8	8.73E-04
Co-58	1.8	1.21E-03

Decay correction is conducted as a function of assumed decay correction time (=Measured time-generation time). Decay correction time is assumed from 16 to 6 yr, which is corresponding to the generation time of radwaste from 1988 to 1998. Fractional abundance is corrected to a current one, which is based on the generation time of radwaste and shown in figure 2. On the contrary to long-lived radionuclides, short-lived radionuclides, such as Co-58 and Nb-95, are decreased rapidly, a highly short-lived radionuclides, such as Mn-54 and Ag-110m, are decay out in a short time. Therefore, activities of these short-lived radionuclides decrease rapidly and can be negligible in a short period of time.

Based on the corrected fractional abundance, corrected activity of each gamma radionuclide is drawn in Figure 3. In this figure, it is assumed that average surface dose rate is 100 mR/hr. Corrected activity as a function of generation year is shown in figure 3. Activity in long-term stored radwaste is composed of mainly Co-60 and Cs-137. Especially, contribution of Cs-137 to the total activity is increased as the stored time goes on. Other radionuclides except the Co-60 can be ignorable in the long-term stored drum. In figure 4, activity ratio is shown between the corrected and the uncorrected activity. Long-lived Cs-137 is predominant in long-term stored radwaste drum, and its activity ratio is increased as the decay correction time is increased. However, activity ratio of Co-60 is not changed significantly. Other relatively short-lived radionuclides are decreased significantly as decay correction time increased.

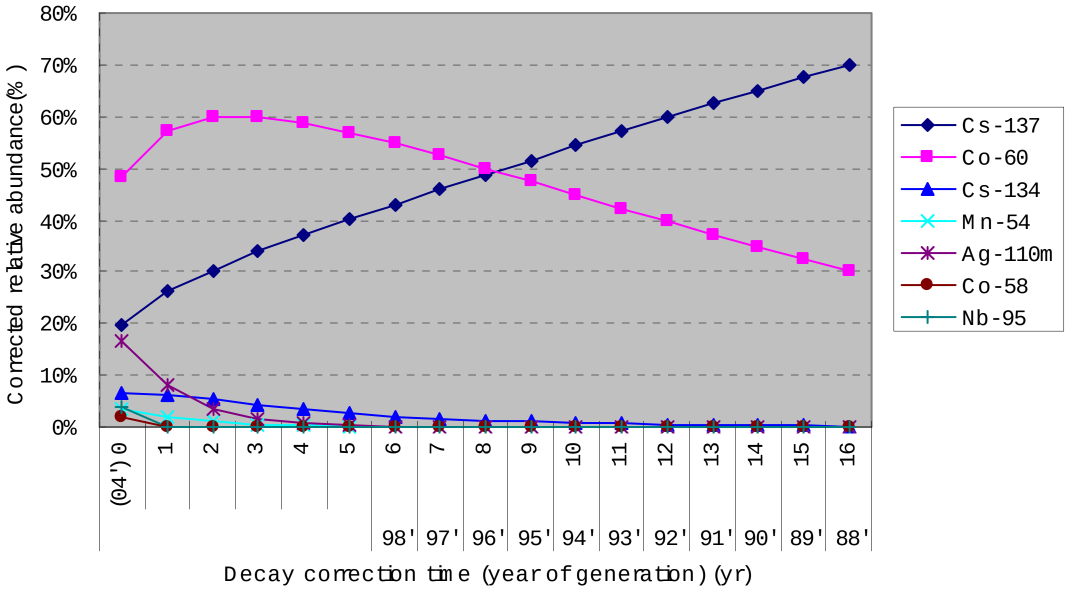

Fig. 2. Corrected fractional abundance as a function of generation year

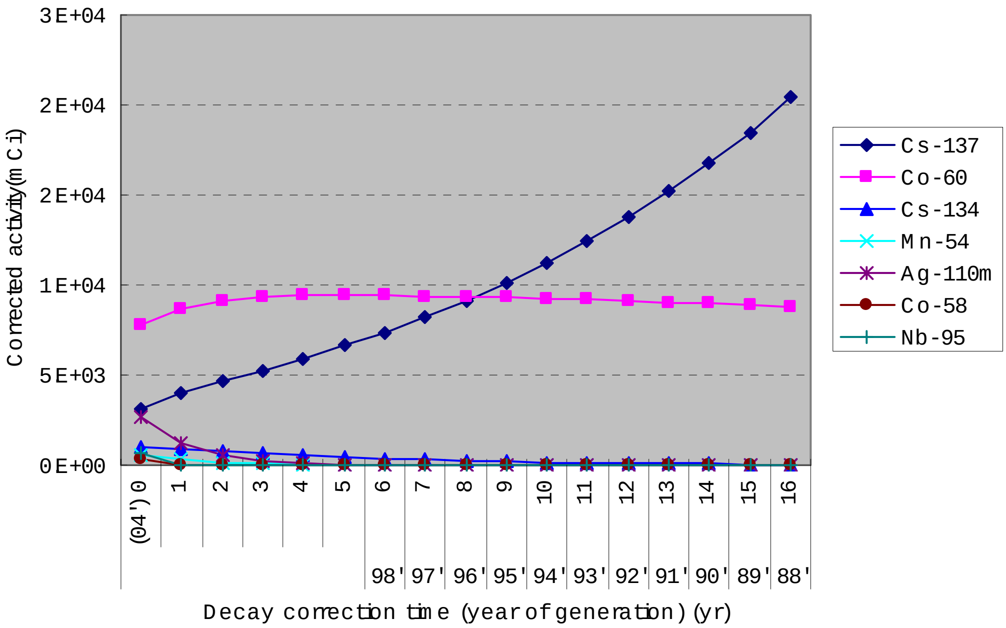

Fig. 3. Corrected activity as a function of generation year

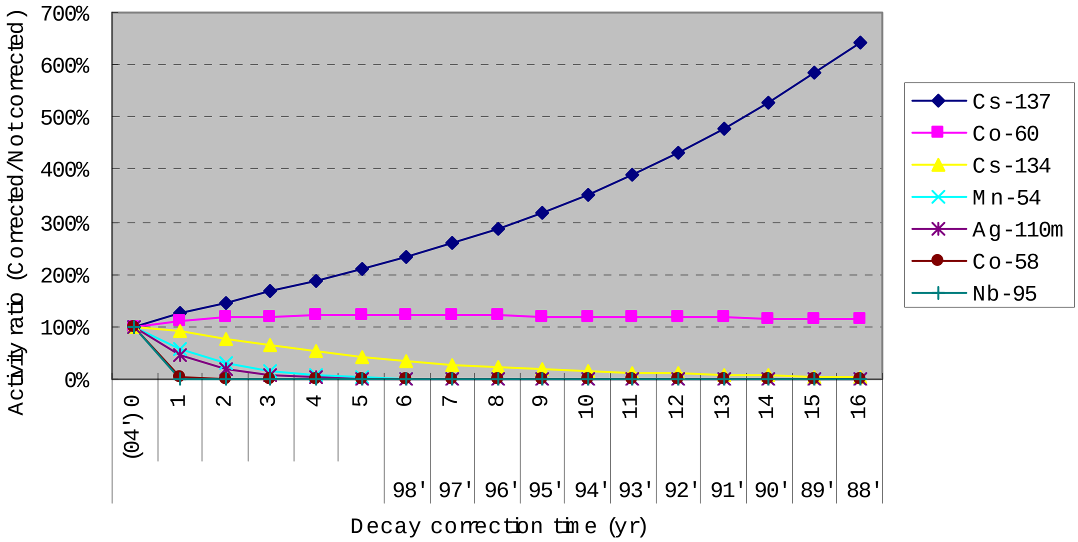

Fig. 4. Activity ratio between the corrected and uncorrected activity

CONCLUSIONS

In this paper, the impact of decay correction is assessed from a viewpoint of the variation of fractional abundance and activity for each gamma emitting radionuclide using a typical radwaste drum. First, the radioactive waste assay system in KORI site and new system are briefly introduced and compared to each other. In the new system and program, target NPPs are expanded from KORI unit to other PWRs and PHWRs. The numbers of assay target radionuclides are also increased from 10 to 22. From the 1st accepted sampling data set, sampling data set is assessed and CF is evaluated using the MCNP-4C code for the typical long-term stored radwaste drum in Ulchin NPPs. Fractional abundance and activity of dominant gamma emitting radionuclides are evaluated. From the case study, it is concluded that decay correction process is essentially for the accurate activity evaluation and should be used for the reliable application of DTC method. Decay correction is also necessary for the prevention of an inordinate under-estimation of key radionuclide activity in a long-term stored radwaste drum.

An additional and frequent sampling procedure is in progress to update sampling data set. As this study goes on, conformation of correlation pairs based on the Korean analyzed data will be provided. Accuracy and representativeness of related parameters, such as SF, fractional abundance and so on, will be improved. Through these progresses, more accurate and reliable prediction of radionuclide inventory in radioactive waste based upon Korean sample-analyzed data set will be possible.

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