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[Technical Paper]

Development of Radionuclide Inventory Declaration Methods Using Scaling Factors for the Korean NPPs
- Scope and Activity Determination Method -

국내 원전 대상의 척도인자를 활용한 핵종재고량 규명 방법의 개발
- 범위 및 방사능 결정 방법-

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Abstract

Regulations and guidelines for radioactive waste disposal require detailed information about the characteristics of radioactive waste drums prior to transport to the disposal sites. However, estimation of radionuclide concentrations in the drummed radioactive waste is difficult and unreliable. In order to overcome this difficulty, scaling factor (SF) method has been used to assess the activities of radionuclides, which could not be directly analyzed. A radioactive waste assay system has been operated at Korean nuclear power plant (KORI site) since 1996 and consolidated SF concept has played a dominant role in the determination of radionuclide concentrations. However, SFs are somewhat dispersive and limited in KORI site. Therefore establishment of the assay system using more improved SFs is planned and progressed. In this paper, the scope of research is briefly introduced. For the selection of more reliable activity determination method, the accuracy of predicted SF values for each activity determination method is compared. From the comparison of each activity determination method, it is recommended that SF determination method should be changed from the arithmetic mean to the geometrical mean for more reliable estimation of radionuclide activity. Arithmetic mean method and geometric mean method are compared based on the data set in KORI system. And, this change of SF determination method will prevent an inordinate over-estimation of radionuclide inventory in radwaste drum.

Key words : scaling factor, radioactive waste, activity, inventory, arithmetic mean, geometric means,

요약

원전에서 발생된 중·저준위 방사성 폐기물의 경우 처분장으로 이송되기 이전에 드럼에 대한 세부적인 정보 특히 핵종 재고량에 대한 평가가 수행되어야 한다. 그러나 드럼처리된 방사성폐기물의 경우 평가 대상 핵종 농도에 대한 예측이 어려운 것이 일반적이다. 따라서 이를 극복하고자 직접측정이 어려운 경우 척도인자 방법을 활용하고 있다. 국내의 경우 1996년부터 고리원전에서 척도인자 개념이 적용된 핵종분석장치를 운영해오고 있다. 그러나 고리원전에 적용된 척도인자의 경우 많은 개선의 여지가 남겨져 있다. 따라서 현재 척도인자의 향상을 위한 연구가 진행중에 있다. 본 논문에서는 연구의 범위에 대한 개략적인 소개와 핵종 재고량 평가 방법 중 보다 신뢰할 수 있는 평가 방법을 찾고자 통계적인 척도인자 평가 방법을 비교 평가했으며 이를 통해 고리원전에 사용된 산술평균 방법을 기하평균 방법으로 바꾸는 것이 예측의 정확성을 향상시킬 수 있을 뿐만 아니라 드럼 내 핵종 재고량의 과대평가를 막고 합리적인 보수성을 유지할 수 있음을 알수 있었다.

중심단어 : 척도인자, 방사성 폐기물, 방사능, 재고량, 산술평균, 기하평균

Ⅰ. Introduction

An environmentally sound and safe management of low-level radioactive waste requires knowledge of the characteristics and the inventories of radionuclides in the radioactive waste package. Accordingly, regulations and guides developed in various countries require detailed description of the radioactive waste package and its contents. In Korea, the Enforcement Decree of the Korean Atomic Energy Act (Articles 88) requires 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 NPP.

An established waste characterization program in KORI site measures the concentrations of gamma-emitting nuclides directly and that of other relevant nuclides indirectly by relating Difficult-To-Measure (DTM) radionuclides to other Easy-To- Measure (ETM) radionuclides. SFs are generated by use of sample data that are gathered from the radiochemical analysis of waste samples collected from 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. And other PWRs except NPPs of KORI site and PHWRs are not considered. It also needs to more number of sampling and reliable sampling procedures for the improvement of reliability. Furthermore, it needs to improve the accuracy of derived SF values based on selection of reliable activity calculation method and conformation of correlation pairs using a Korean sample-analyzed data.

For that reason, it is in progress to establish an assay system using more improved SFs for updating the performance of Korean nuclear waste management. Korean Hydro & Nuclear Power Co. Ltd. (KHNP) organizes the overall project with partial cooperation with Korean 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 prediction program]. In this paper, the status of radionuclide activity determination method in KORI site is briefly introduced. And the changes of assay target nuclides, target NPPs and so on are explained. Also, the comparison of applicable activity determination methods is conducted and the more reliable activity determination method is selected.

Ⅱ. Status of Radionuclide Activity Determination Method in Kori Site

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.

1. Radioactive Waste Assay System (SGS: Segmented Gamma Scanning)

Radioactive waste assay system was designed, manufactured, and installed at KORI NPP in order to reduce the exposure of worker and to measure the total activities of homogeneous and non-homogeneous waste. This system also was designed to calculate the total activities of waste drum using SFs by measurement of the representative gamma-emitting radionuclides such as ^{60}Co and ^{137}Cs. Dividing a waste drum into eight vertical segments and eight radial sectors in each vertical segment, the activities of homogeneous and non-homogeneous waste drum were measured by this system.

2 Scaling Factor Method and Assay Target Radionuclides

In the selection of assay target radionuclides in KORI site, the following conditions were considered;

A) Costs for radiochemical analysis
B) Radiological and biological toxicity
C) Radionuclides listed in regulations and safety assessments

Under these considerations, the following radionuclides were selected; ^{3}H, ^{14}C, ^{55}Fe, ^{63}Ni, ^{60}Co, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{137}Cs and gross alpha (TRU). The established waste characterization program measured the concentrations of all relevant nuclides either directly or indirectly by relating DTM radionuclides to other ETM radionuclides (Key nuclides). Pairs of correlated nuclides are introduced in Table I.

III. Scope of New Assay System

The project of new radioactive waste assay system is started in 2003 and will be finished in the end of 2005. Principal change of new system compared to KORI system is assay target DTM radionuclides and NPPs.

1. Assay Target DTM Radionuclides

In the selection of assay target radionuclides, the following conditions are considered;

A) Half life >= 30 yr
B) Physical and chemical reactivity with groundwater
C) Radionuclides listed in regulations and safety assessments (10 CFR 61.55)
D) Radiological and biological toxicity
E) Difficult-to-measurable beta/gamma emitting radionuclides without destructive method

From the above selection criteria, the following radionuclides are selected by KAERI ^{3}H, ^{14}C, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{58}Co, ^{60}Co, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm and gross alpha (TRU).

2. Assay Target NPPs and Waste Types

Assay target NPPs are expanded from KORI site to all Korean NPPs including PHWRs. All PWRs are classified by sharing radwaste treatment system and 4 PHWRs are classified for each unit. Finally, all 20 NPPs are classified into 13 groups. Classified 13 groups are summarized in Table II.

Assay target waste types and its representative samples are the same as the following.

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

Table I. Pairs of Correlated Key & DTM Nuclides

Scaling factor	^{60}Co	^{14}C, ^{55}Fe, ^{63}Ni, ^{94}Nb
	^{60}Co	^{14}C, ^{55}Fe, ^{63}Ni, ^{94}Nb
	^{137}Cs	^{3}H, ^{90}Sr, ^{99}Tc, TRU

Table II. Classified 13 Groups of NPPs

PWR (16 unit)		PHWR (4 unit)
Kori Unit #1 Kori Unit #2 Kori Unit #3,4 Ulchin #1,2 Ulchin #3,4 ***Ulchin#5,6***	Yonggwang #1,2 Yonggwang #3,4 Yonggwang #5,6	Wolsong #1 Wolsong #2 Wolsong #3 Wolsong #4
In Operation: 18 units *In preparation Ulchin #5* *Under construction: Ulchin #6*		

Ⅳ. Activity Determination Methods

Applicable activity determination methods are compared. All methods are summarized in Table Ⅲ. Statistical techniques are used in these methods such as the arithmetical mean, geometrical mean, linear regression, and logarithmic regression [1, 2, and 3]. However, there is not any definition for the most reliable activity determination method. For that reason, each country uses its own preferred scaling factor method. In general, linear regression of logarithm is preferred for the calculation of SF value in the activity determination method. However, arithmetical mean was used for the activity determination methods in KORI site because it has a little sample-analyzed data. In this study, two sets of input data, which are shown in Table IV, were used for the comparison of each method. At first, foreign data set is used for the comparison of four activity determination methods [3]. Next, KORI sample-analyzed data set is used for the comparison of arithmetic mean method and selected most reliable method [4]. For the comparison of each method, proper data set of key/ DTM nuclides in a specific waste type or all waste type was used. Detailed information of data set is summarized in Table IV.

Ⅴ. Results and Discussion

The resulting plots of measured and estimated concentrations for each activity determination methods are shown in Figure 1 and 2 based on the data scale. In arithmetic mean method, each activity and total activity are overestimated. Linear regression method is not proper for activity determination because it shows large disparity between the measured and estimated activities. The linear regression of logarithms

Table Ⅲ. Activity Determination Methods Using Key Nuclides

Method		Mathematical expression	Coefficient	Activity determination
Arithmetic mean	Linear relati	$A_{RN}=a*A_{KN}$	a= Average ratio [SF]	Arithmetic mean of SFs
Linear regression		$A_{RN}=a+b*A_{KN}$	a, b=cons	Linear regression of key & DTM nuclides
Geometric mean	Linear relation of logarithm	$A_{RN}=c*A_{KN}$	c= Average ratio [SF]	Geometric mean of SFs
Linear regression of logarithms		$\mathrm{Log}(A_{RN})=c'+d*\log(A_{KN})$ $A_{RN}=c*(A_{KN})^{d}$	c, c', d =const	Logarithmic linear regression of Key & DTM nuclides

Table IV. Information of The Input Data Set

Data set	(1) EPRI-5077	(2) KORI data set
Waste type (# of data set)	Spent Resin (139) (Excluding 7 extreme data)	A) Spent filter (4) B) Concentrate bottom (4) C) Spent resin (4) D) DAW (12)
Key nuclide	^{60}Co	^{60}Co, ^{137}Cs
DTM nuclide	^{63}Ni	^{3}H, ^{14}C, ^{63}Ni ^{90}Sr, ^{99}Tc, Gross alpha

Table. V. Comparison Results of Each Activity Determination Methods.

Method	Estimated (Y) Vs Measured (X) Y=A*X+B	Ratio of total activity [Estimated/Measured]	Accuracy of estimation
Arithmetic mean	A=1.0101 B=0.5481	5.20	Overestimation of each activity and total activity
Linear regression	A=0.0607 B=0.8343	1.00	Large disparity between measured activity and estimated activity
Geometric mean	A=1.0101 B=0.0081	1.50	Proximity of each activity Conservative estimation of total activity
Linear regression of logarithms	A=0.8648 B=- 0.1080	0.65	In a low activity region: Overestimation In a high activity region: Underestimation Underestimation of total activity (Under-conservative)

has a characteristic to underestimate the total activity. The reason is that it overestimates the activities in low activity region and underestimates the activities in high activity region. Therefore, this method is under-conservative. In the geometric mean method, the estimated activity is very close to the measured one and total activity is conservatively estimated at a reasonable level. These comments could be confirmed through the comparison of measured and estimated concentrations of each activity determination method in Figure 1. More detailed comparison results of each activity determination method are summarized in Table V. From the ideal case (ideal regression = reference line), we can establish the conservative conditions such as followings. (In here, A (slope) and B (y intercept) are coefficients in log-scale regression)

A) Individual activity

(Ideal case: A=1, B=0)

1) Estimated concentration = Measured concentration

2) A ≥ 1 & B≥0

B) Ratio of total radioactivity

(Ideal case: Ratio=1; Estimated total activity = Measured total activity)

1) Ratio ≥ 1 (Estimated total activity ≥ Measured total activity)

From the overall comparison of activity determination methods, it is concluded that geometric mean method is the most reliable activity determination method. Geometric mean and arithmetic mean are evaluated and compared by use of the sample-analyzed data set in KORI system. The ratio values of arithmetic mean and geometric mean for each waste type and radionuclide are illustrated in Fig 3. The ratio values are higher than 1 for all waste types and pairs of DTM/Key radionuclides, which are corresponding to the viewpoint of statistics. This is particularly high in resin and DAW.

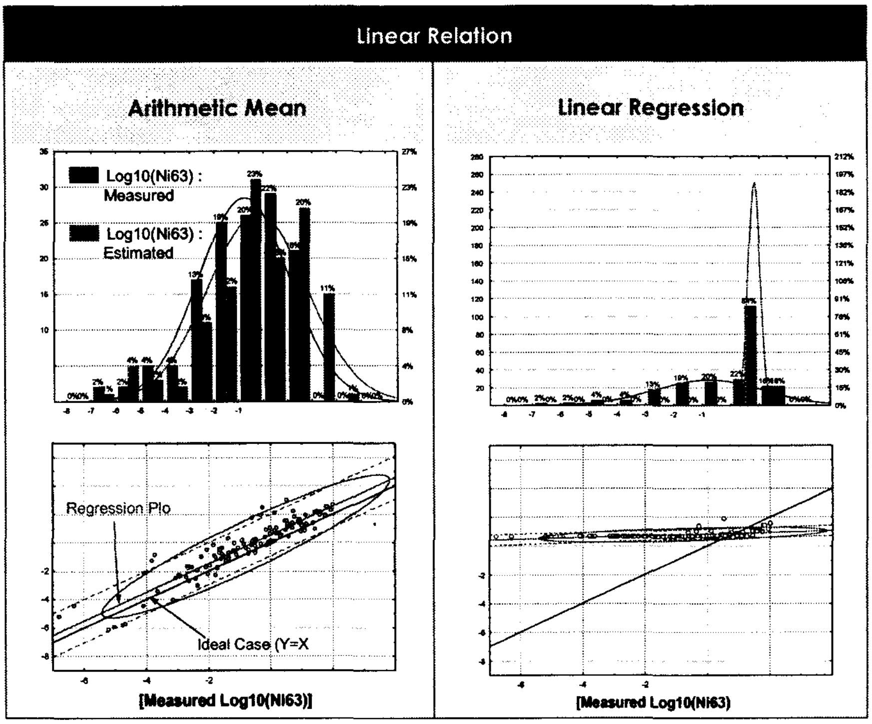

Fig. 1. Comparison of Measured and Estimated Concentrations for Activity Determination Method Using a Linear Relation.

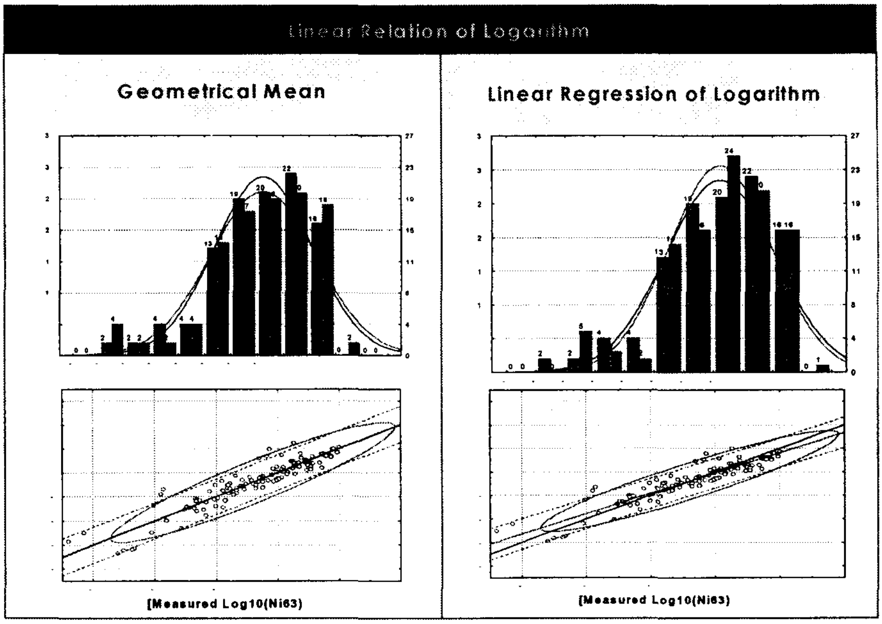

Fig. 2. Comparison of Measured and Estimated Concentrations for Activity Determination Method Using a Linear Relation of Logarithm.

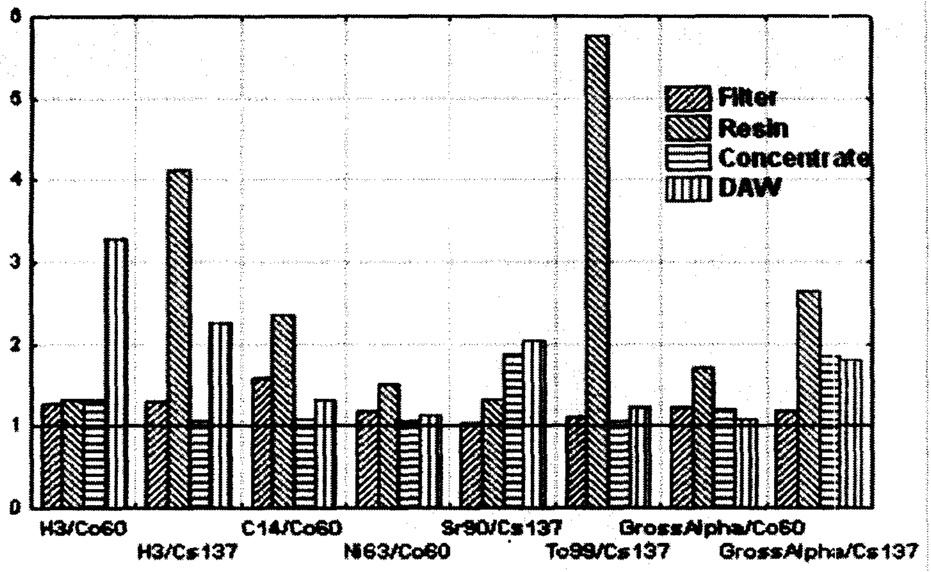

Fig. 3. The Ratios of Arithmetic Mean and Geometric Mean for Each Waste Type And Pairs of Radionuclides.

VI. Conclusion

In this paper, radioactive waste assay system in KORI site is briefly introduced. Also, scope and plan for new radioactive waste assay system are compared with previous installed one in KORI site. In this 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. For the evaluation of accuracy for each activity determination method, foreign and KORI data set were used. Inter-comparison was conducted in a viewpoint of accuracy and conservation of estimation. From the comparison of each activity determination method, it is concluded that geometric mean method is the most reliable activity determination method. Also, it is recommended that SF determination method should be changed from the arithmetic mean to the geometrical mean for the improvement of accuracy and reasonable conservation in activity determination. From the comparison of geometric and arithmetic means based on the sample-analyzed data in KORI system, arithmetic mean is higher than geometric mean for all waste types and pairs of DTM/Key radionuclides. This is corresponded to a viewpoint of statistics. In particular, SF values in resin and DAW is higher than ones in other waste types. This change of SF determination method will prevent an inordinate over-estimation of radionuclide inventory in radwaste drum.

An additional and frequent sampling procedure is in progress to update the performance of Korean nuclear waste management. As this study goes on, conformation of correlation pairs based on the Korean analyzed data will be provided. Also, accuracy and representativeness of derived scaling factor values will be improved. Through these progresses, more accurate and reliable prediction for the radionuclide inventory of radioactive waste based upon Korean sample-analyzed data set will be possible.

Ⅶ. Acknowledgement

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