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Study on Radionuclide Inventory Evaluation in LILW using Material Balance Method

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1. Introduction

It is necessary to assess the inventory of radionuclides in the waste for disposal of Low-and Intermediate-Level Radioactive Waste (LILW). According to the law in South Korea, the evaluation of 14 radionuclides is required (^{3}H, ^{14}C, ^{55}Fe, ^{58}Co, ^{59}Ni, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{137}Cs, ^{144}Ce, Gross alpha). Radionuclides whose concentration is easily evaluated by measuring gamma rays from the surface of the drum are designated as Easy-to-Measure (ETM) nuclides, while those that cannot be directly measured are classified as Difficult-to-Measure (DTM) nuclides.

The scaling factor method is used to indirectly determine the inventory of DTM nuclides when direct, non-destructive measurement methods, such as external gamma-ray spectrometry, are not feasible. This approach utilizes the correlation derived from radiochemical analysis results between ETM and DTM nuclides. The ETM nuclide that strongly correlates with the DTM nuclide is called the Key nuclide, which is usually either ^{137}Cs or ^{60}Co. However, due to recent improvements in nuclear fuel clad integrity and decontamination techniques, DTM nuclides and ^{137}Cs may not be detected in the samples. While an alternative method of deriving the conservative scaling factor [1] for ^{129}I to ^{60}Co may be used when ^{137}Cs is not detected, this approach could lead to increased volume of LILW or greater uncertainty, which requires more suitable alternatives. In this work, we analyze an alternative method to estimate the radionuclide inventory in the LILW using a material balance method utilizing the Pressurized Water Reactor-Gaseous and Liquid Effluents (PWR-GALE) code from sampled waste.

2. Methods and Results

2.1 Measured Data for Waste Drum

The database used for analysis utilized the radiochemical analysis data from the report Updated Scaling Factors in Low Level Radwaste (NP-5077), which was published by the Electric Power Research Institute (EPRI, USA) [2]. In this study, data were used for waste filters and waste resins, and units were converted into Ci/year for ease of comparison. The assumptions for unit conversion include considering the drum size as 208L and estimating the total annual waste drum generation per nuclear power reactor to be 250 drums. The generation ratios of filters and resins are assumed to be 3 percent and 10 percent, respectively. The unit conversion results of some nuclides present in the filters and resins are shown in Table 1.

Table I: Nuclide concentrations of Filter and Resin

Nuclide	Concentration [Ci/year]	
	Filter	Resin
^{14}C	2.81E-03	1.04E-02
^{55}Fe	1.23E+00	9.15E-01
^{60}Co	2.65E-01	1.62E+00
^{63}Ni	1.31E-01	1.29E+00
^{90}Sr	6.86E-04	7.49E-03
^{99}Tc	4.84E-05	1.04E-04
^{129}I	7.18E-06	4.99E-05
^{137}Cs	8.27E-03	8.74E-01
^{144}Ce	2.81E-02	4.16E-02

2.2 Material Balance Method

The Material Balance Method is a method that uses the Law of Conservation of Mass to balance the introduction, discharge, and accumulation of substances. Equation (1) represents the balance equation in the system [3].

$$(1)\quad A = I - O + P - C$$

If balanced quantity is set to the total mass, Production (P) and Consumption(C) are set to 0, which changes as shown in Equation (2) below as only Input(I), Output(O), and Accumulation(A) remain.

$$(2)\quad A = I - O$$

2.3 PWR-GALE code

The PWR-GALE Code is a computerized mathematical model for calculating the releases of radioactive material in gaseous and liquid effluents from pressurized water reactors [4].

When the material balance method mentioned above is applied to the PWR-GALE code, it can be thought of as a concept of comparing before and after Purification. So, the waste generated from a nuclear power plant before purification was set as the Input, after purification as the Output, and the concentration of nuclides remaining in the purification system as Accumulation.

In this case, the PWR-GALE code was used, assuming that the default input values of the PWR-GALE code represent the post-purification state. Meanwhile, the pre-purification state was defined by setting all decontamination-related input values, such as the efficiency of the HEPA filter, to zero. The difference between these two states was then considered as the amount of nuclides retained in the purification system.

2.4 Comparison of Calculated and Measured Data

Since the types of nuclides derived from the PWR-GALE code differ from those in EPRI's measured data, the comparison was conducted only for the overlapping nuclides. Table 2 presents the comparison of nuclide concentrations in filters and resins obtained using the PWR-GALE code and the Material Balance Method with EPRI's measured data.

Table II: Comparison of Calculated and Measured Data

Waste type	Nuclide	Calculated value [Ci/year]	Measured value [Ci/year]	Ratio[a]
Filter	^{60}Co	1.09E-02	2.65E-01	0.04
	^{90}Sr	6.24E-03	6.86E-04	9.09
	^{137}Cs	8.91E-03	8.27E-03	1.08
Resin	^{55}Fe	1.07E-02	9.15E-01	0.01
	^{60}Co	5.00E-03	1.62E+00	0.00[b]
	^{63}Ni	0.00E+00[c]	1.29E+00	0.00[b]
	^{90}Sr	1.20E-04	7.49E-03	0.02
	^{137}Cs	1.70E-01	8.74E-01	0.19
	^{144}Ce	3.58E-02	4.16E-02	0.86

[a](PWR-GALE value) / (EPRI's measured data).
[b]This value is less than 1E-02
[c]This value is less than 1E-05

In the case of filters, ^{60}Co showed a very large difference of 0.04 and ^{90}Sr of 9.09, while ^{137}Cs showed a relatively consistent value of 1.08. In the case of resin, the results derived by the PWR-GALE code for all compared nuclides showed smaller results than the measured values.

3. Conclusions

When assessing the radionuclide inventory of Low- and Intermediate-Level Radioactive Waste, the concentration of ^{137}Cs determined using the PWR-GALE code showed satisfactory agreement for filters. The radionuclide inventory evaluation method based on material balance proposed in this study may serve as a supplementary approach for addressing challenges in measuring ^{137}Cs concentration in waste samples.

However, since the results of this study were primarily compared with the EPRI's measured data from 1987, a reassessment will be conducted in the future using the latest measured data on radioactive waste to reflect recent developments, such as improvements in nuclear fuel clad integrity and extended fuel cycles.

REFERENCES

[1] Cox, B., & Saunders, P. (2015). Development of Generic Scaling Factors for Technetium-99 and Iodine 129 in Low and Intermediate Level Waste. *Electric Power Research Institute Report, Technical Report 3002005564*.

[2] Best, W. T., & Miller, A. D. (1987). *Updated scaling factors in low-level radwaste* (No. EPRI-NP-5077). Impell Corp., Walnut Creek, CA (USA); Advanced Process Technology, Milpitas, CA (USA); Electric Power Research Inst., Palo Alto, CA (USA).

[3] Zeman, F. (2012). Separation of mixtures: fundamentals and technologies. In *Metropolitan Sustainability* (pp. 53-74). Woodhead Publishing.

[4] Chandrasekaran, T., Lee, J. Y., & Willis, C. A. (1985). *Calculation of releases of radioactive materials in gaseous and liquid effluents from pressurized water reactors (PWR-GALE Code). Revision 1* (No. NUREG-0017-Rev. 1). US Nuclear Regulatory Commission (NRC), Washington, DC (USA). Office of Nuclear Reactor Regulation.

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