Validation of Radiochemical Analysis Results for NPP Dismantling Radwastes : Using Radioactive Metal, Concrete and Soil Reference Materials

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1. Introduction

As the Kori-1 Nuclear Power Plant (NPP) was permanently shut down in 2017, the various researches related to its decommissioning are performing in depth. The amounts of low- and intermediate-level radioactive wastes generated in the dismantling of nuclear power plant is expected to be about 14,500 drums, these wastes are mainly contaminated metal scraps, concrete and soil fragments discharged through dismantling and decontamination process of various equipment and structures related to operation of the NPP. However, they have complex chemical matrices and extensive radioactivity, making it difficult to differentiate between deregulated and regulated waste unless managed systematically through radiochemical analysis. Therefore, a procedure for ensuring or monitoring the validity of the results must be established after confirming and verifying the radioactivity analysis method for such radioactive waste samples. In this study, the comparative analysis sample are SUS 304 liquid reference material (RM) and Portland concrete and soil certification reference material (CRM), respectively. And gamma-emitting nuclides (^{60}Co, ^{134}Cs, ^{137}Cs) and beta-emitting radionuclides (^{90}Sr) were selected as the analyte. Finally, En-score was applied to a statistical treatment of the results obtained from three institutes participated in the interlaboratory comparisons. En-score indicates at what multiple of the value of the extended uncertainty between the certified value and the measured value analyzed by the analysis agency, and was judged to be suitable when -1< En <1.

2. Materials and Methods

2.1 Characteristics of Rad-wastes from dismantling.

As for the amount of the waste generated from unit 1 of the light-water reactor predicted by the IAEA, contaminated metals are most frequently generated and followed by concrete as shown in Table 1. Therefore, SUS 304 (metal), Portland concrete (matrix: SiO_2, CaO, Al_2O_3) and soil (matrix: SiO_2 and Al_2O_3) were selected.

Table 1. The amount of Radwaste from PWR dismantling.

Waste Forms	Amount (Drum)	Proportion (%)
Metal Waste	650	10.5
Concrete Waste	300	4.8
Contaminated Metal	3,500	56.5
Contaminated Concrete	600	9.7
Contaminated Metal Piece	150	2.4
Contaminated Solid Piece	1,000	16.1
Total	6,200	100

2.2 Preparation of the radioactive Reference Materials

The radioactive acidic solutions based on SUS-304 used as the matrix for the RM were prepared. In order to carry out this study, the Korea Research Institute of Standards and Science (KRISS) was commissioned to manufacture this RMs with the same chemical composition as shown in Table 2 and Fig. 1. The CRMs of the concrete and soil in bead type with the same chemical compositions as shown in Table 3 and 4 along with Fig. 1 were purchased from KRISS.

Table 2. Characteristics of a metal RM.

Species	Concentration (g/kg)	Radioactivity (Bq/kg)
Fe	13.72	-
Ni	1.86	-
Cr	3.8	-
Mn	0.4	-
^{60}Co	-	267
^{134}Cs	-	206
^{137}Cs	-	255
^{90}Sr	-	248

Table 3. Characteristics of a concrete CRM.

Species	Concentration (g/kg)	Radioactivity (Bq/kg)
SiO_2	740	-
CaO	189	-
Al_2O_3	34.6	-
^{60}Co	-	0.1 ~ 1.0
^{137}Cs	-	0.1 ~ 1.0

Table 4. Characteristics of a soil CRM.

Species	Concentration (g/kg)	Radioactivity (Bq/kg)
SiO_2	670	-
Al_2O_3	199	-
K_2O	23.5	-
Fe_2O_3	14.8	-
Na_2O	14.5	-
CaO	7.3	-
MgO	3.5	-
^{60}Co	-	0.1 ~ 1.0
^{137}Cs	-	0.1 ~ 1.0

Fig 1. RM and CRMs for the analysis of ^{90}Sr, ^{134}Cs, ^{137}Cs, ^{60}Co

3. Result and Discussion

3.1 Statistical review for the analysis results

$$(E_n)_i = \frac{x_i - x_{pt}}{\sqrt{U^2(x_i) + u^2(x_{pt})}}$$

In the equation [2],

- *x_i: Reported value of A, B and C test institutes*
- *x_{pt}: Assigned value determined by reference laboratory*
- *$U(x_i)$: Expanded uncertainty of A, B and C institute*
- *$U(x_{pt})$: Expanded uncertainty of certified value*
- *Satisfactory range: -1.0 < En < 1.0*

x_i is the average value of A, B, and C institutions, x_{pt} is the certification value of the cross-analysis sample, $U(x_i)$ is the extended uncertainty of A, B, and C institutions, and $u(x_{pt})$ is the extended uncertainty of the certification value.

3.2 Evaluation of the analysis results

To ensure the validity of the radionuclide analysis results, A, B and C test institutes were participated in the comparative test and the results were shown in Table 5-7.

Table 5. Analysis results using RM of SUS 304

Analysis Institute	Radio-nuclide	Assigned value		Reported value		En-score	Result
		Radioactivity (Bq/kg)	Uncertainty (k=2)	Radioactivity (Bq/kg)	Uncertainty (k=2)		
A	^{134}Cs	206	8	179	3	-3.1	US
	^{137}Cs	255	13	254	6	-0.1	S
	^{60}Co	267	10	259	6	-0.7	S
	^{90}Sr	248	12	245	8	-0.2	S
B	^{134}Cs	206	8	182	11	-1.8	US
	^{137}Cs	255	13	245	14	-0.5	S
	^{60}Co	267	10	265	15	-0.1	S
	^{90}Sr	248	12	335	15	4.5	US
C	^{134}Cs	206	8	185	12	-1.5	US
	^{137}Cs	255	13	246	22	-0.4	S
	^{60}Co	267	10	265	24	-0.1	S
	^{90}Sr	248	12	347	21	4.1	US

* US: Unsatisfactory, S: Satisfactory

Table 6. Analysis results using CRM of concrete

Analysis	Num	Radio-nuclide	Assigned Value		Reported Value		E_n-Score	Result
			Radioactivity (Bq/kg)	Uncertatinty (k=2)	Radioactivity (Bq/kg)	Uncertatinty (k=2)		
A	I	Co-60	80.3	3.1	123.9	10.9	3.8	US
		Cs-137	75.4	4.5	117.7	17.3	2.4	US
	II	Co-60	1030	70	1004.0	86.1	-0.2	S
		Cs-137	1080	67	991	146	-0.6	S
B	I	Co-60	80.3	3.1	84.0	5.5	0.6	S
		Cs-137	75.4	4.5	77.0	5.0	0.2	S
	II	Co-60	1030	70	1121	75	0.9	S
		Cs-137	1080	67	1126	75	0.5	S
C	I	Co-60	80.3	3.1	88.8	8.0	1.0	S
		Cs-137	75.4	4.5	83.2	5.1	1.2	US
	II	Co-60	1030	70	1120	102	0.7	S
		Cs-137	1080	67	1121	70	0.4	S

Table 7. Analysis results using CRM of soil

Analysis	Num	Radio-nuclide	Assigned Value		Reported Value		E_n-Score	Result
			Radioactivity (Bq/kg)	Uncertatinty (k=2)	Radioactivity (Bq/kg)	Uncertatinty (k=2)		
A	I	Co-60	124	10	119	11	-0.3	S
		Cs-137	119	10	120	18	0.1	S
	II	Co-60	1176	68	1174	39	-0.03	S
		Cs-137	1115	56	1161	65	-0.5	S
B	I	Co-60	124	10	128	8	0.3	S
		Cs-137	119	10	127	8	0.6	S
	II	Co-60	1176	68	1200	78	0.2	S
		Cs-137	1115	56	1214	79	0.9	S
C	I	Co-60	124	10	128	8	0.3	S
		Cs-137	119	10	123	11.	0.3	S
	II	Co-60	1176	68	1201	109	0.2	S
		Cs-137	1115	56	120	75	0.9	S

In the case of the metal radioactive liquid reference material (RM), all participating institutions provided satisfactory results that were in accordance with the reference values for gamma emitters, ^{60}Co and ^{137}Cs. However, for ^{134}Cs, the results were lower than the reference values, and this is due to inadequate correction of simultaneous synthesis effect of ^{134}Cs, and correction for this was necessary [3].

As for ^{90}Sr, a pure beta emitter, only one of the three institutes provided satisfactory result that matched the reference value, while the other two labs presented significantly high results than the reference value. If the interfering radionuclides gamma-ray emitting beta nuclides such as ^{134}Cs and ^{137}Cs are not completely removed, the result of radioactivity concentration of ^{90}Sr in the sample can be overestimated, more than 100%.

In the case of the soil radioactive certified reference material (CRM), all three institutes reported satisfactory results for ^{60}Co and ^{137}Cs. However, for the concrete CRM, unsatisfactory results for ^{60}Co and ^{137}Cs in the low radioactive ranges were reported by A and C institutes. It was assumed that this is due to the failure to correct the effect of the density of the bead sample in the low-concentration radioactivity range.

4. Conclusions

Interlaboratory comparisons was carried out to monitor the validity of analysis results using the RM of SUS 304 and two CRMs of concrete and soil. If such study is continuously carried out, it will be useful that QA and QC of radioactivity measurements of the radioactive waste generated from NPP dismantling.

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