Nuclear Engineering and Technology 54 (2022) 2418–2426

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Verification of the adequacy of domestic low-level radioactive waste grouping analysis using statistical methods

Dong-Ju Lee [a, b], Hyunjong Woo [c], Dae-Seok Hong [a], Gi Yong Kim [d], Sang-Hee Oh [a], Wonjun Seong [e], Junhyuck Im [e, **], Jae Hwan Yang [b, *]

[a] *Radwaste Management Center, Korea Atomic Energy Research Institute (KAERI), 989-111 Daedeok-daero, Yuseong-gu, Daejeon, 34057, South Korea*
[b] *Department of Environmental & IT Engineering, Chungnam National University, 99 Daehak-ro, Yuseong-gu, Daejeon, 34134, South Korea*
[c] *Euclidsoft, 317-20 Daedeok-daero, Seo-gu, Daejeon, 35214, South Korea*
[d] *Radioactive Waste Chemical Analysis Center, Korea Atomic Energy Research Institute (KAERI), 989-111 Daedeok-daero, Yuseong, Daejeon, 34057, South Korea*
[e] *Decommissioning Technology Research Division, Korea Atomic Energy Research Institute (KAERI), 989-111 Daedeok-daero, Yuseong, Daejeon, 34057, South Korea*

ARTICLE INFO

Article history:
Received 5 November 2021
Received in revised form
7 January 2022
Accepted 7 January 2022
Available online 10 January 2022

Keywords:
Low-level radioactive waste disposal
Grouping analysis
Soil
Concrete
Dry active waste
Composite sample

ABSTRACT

The grouping analysis is a method guided by the Korea Radioactive Waste Agency for efficient analysis of radioactive waste for disposal. In this study, experiments to verify the adequacy of grouping analysis were conducted with radioactive soil, concrete, and dry active waste in similar environments. First, analysis results of the major radionuclide concentrations in individual waste samples were reviewed to evaluate whether wastes from similar environments correspond to a single waste stream. As a result, the soil and concrete waste were identified as a single waste stream because the distribution range of radionuclide concentrations was "within a factor of 10", the range that meet the criterion of the U.S. Nuclear Regulatory Commission for a single waste stream. On the other hand, the dry active waste was judged to correspond to distinct waste streams. Second, after analyzing the composite samples prepared by grouping the individual samples, the population means of the values of "composite sample analysis results/individual sample analysis results" were estimated at a 95% confidence level. The results showed that all evaluation values for soil and concrete waste were within the set reference values (0.1–10) when five-package and ten-package grouping analyses were conducted, verifying the adequacy of the grouping analysis.

1. Introduction

Various kinds of the low-level radioactive waste are generated from nuclear facilities such as nuclear power plants, research institutes, nuclear fuel manufacturers, etc. in South Korea. Although the waste generators have continuously carried out the final disposal of radioactive waste after the operation of six underground silos for the low- and intermediate- level radioactive waste disposal at Gyeong-ju [1], large quantities of radioactive waste that must be disposed of are still temporarily stored in individual facilities (Fig. 1) [2]. In addition, as decommissioning of the nuclear power plants are scheduled after the permanent shutdown of Kori Unit 1 in 2017, measures to dispose of the decommissioning waste, are also becoming a national issue on the radioactive waste management [3]. In order to permanently dispose of radioactive waste, the physical, chemical, and radiological characteristics of radioactive waste should be identified [4,5]. In particular, among the radiological characteristics, it is essential and required to identify the concentrations of radionuclides in radioactive waste [6]. The Korea Atomic Energy Research Institute (KAERI) mainly performs the destructive analysis, which entails analyzing the concentrations of radionuclides using the representative samples taken from waste packages to determine the concentrations of radionuclides [7]. However, in the case of the destructive analysis, the capacity of analysis that can be conducted within a predetermined time is limited because the time required for the analysis is long due to the preparation and pre-treatment of analysis samples [8]. Therefore, in order to expand the quantities of

---

[*] Corresponding author.
[**] Corresponding author.
E-mail addresses: jhim@kaeri.re.kr (J. Im), yjh98@cnu.ac.kr (J.H. Yang).

https://doi.org/10.1016/j.net.2022.01.016

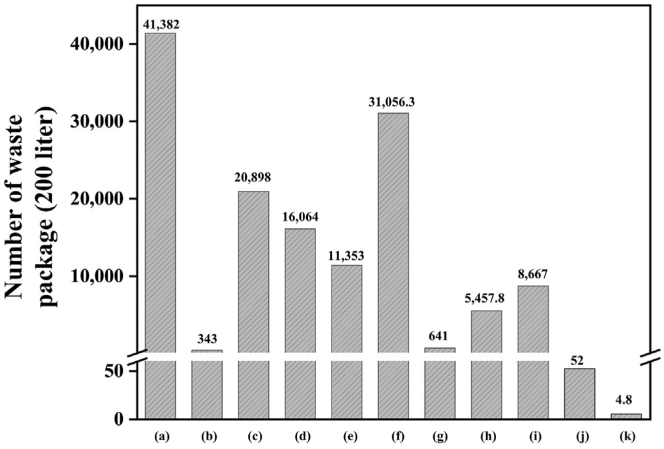


Fig. 1. The status of the radioactive waste storage at the 4th quarter of 2020 [2]. ((a) Kori nuclear site, (b) Saewool nuclear site, (c) Hanbit nuclear site, (d) Hanul nuclear site, (e) Wolsong nuclear site, (f) Deokjin-dong site, Daejeon, (g) Gongneung-dong site, Seoul, (h) Wolsong disposal facility site, (i)Taekwang industrial Co., Ltd., (j)TaeguTec, Ltd., (k) Atomic Creative Technology (ACT) Co., Ltd.).

radioactive waste analyses, the methods that enable the analysis of more radioactive waste analyses within a predetermined time by reducing the quantity of analysis samples are necessary.

The Waste Acceptance Criteria [9] by the Korea Radioactive Waste Agency (KORAD), a domestic intermediate- and low-level radioactive waste disposal facility operator, presented grouping analysis as a plausible method to reduce the quantity of analysis samples. Grouping analysis is a method in which waste packages from similar generation processes are grouped. Then, the composite samples are prepared and analyzed by mixing certain quantities of individual samples collected from individual waste packages, and the resultant value is given equally to individual packages (Fig. 2). Conducting grouping analysis has a great advantage in that the number of samples used for analysis can be reduced. But the problems of overestimation and underestimation of the concentrations of radionuclides can be expected when the radionuclide concentrations of composite samples are equally given to individual waste packages.

In the case of underestimation of radionuclide concentrations, the results of analysis of composite samples indicate concentrations lower than the actual radionuclide concentrations in individual samples, and this can cause problems in terms of safety approach [10] when the disposal facility is operated because the concentrations of radionuclides in the radioactive waste that must be disposed of are “considered” to be lower than actual concentrations. On the other hand, in the case of overestimation of radionuclide concentrations, the results of analysis of composite samples indicate radionuclide concentrations higher than the actual concentrations in individual samples. This can cause the problem that the total inventory of radionuclides in the disposal facility is indicated to be higher than the actual inventory because of the overestimation of radionuclide concentrations given to the radioactive waste for disposal. This eventually leads to the result that the tolerance limit of total radioactivity is reached earlier despite that the total amount of radioactivity allowed for the disposal facility has not been actually reached. That is, the disposal facility may be closed with less radioactive waste compared to the designed disposal capacity, causing problems in terms of the efficiency and economic feasibility of its operation [11–13].

Therefore, the results of grouping analysis should not be underestimated or overestimated. And for this purpose, similarity between the results of analysis of the composite sample and the actual concentrations of radionuclides in individual samples should be secured. The waste acceptance criteria of KORAD [9] present the prerequisites for similarity as the grouping should be limited to “waste from similar environments” and limited to five packages at the maximum. However, since the distributions of the concentrations of radionuclides may not be similar even in the case of waste from similar environments, to apply the grouping analysis method to the analysis of actual radioactive waste, it needs prior studies to verify the similarity of the waste grouping and to check the adequacy of grouping analysis.

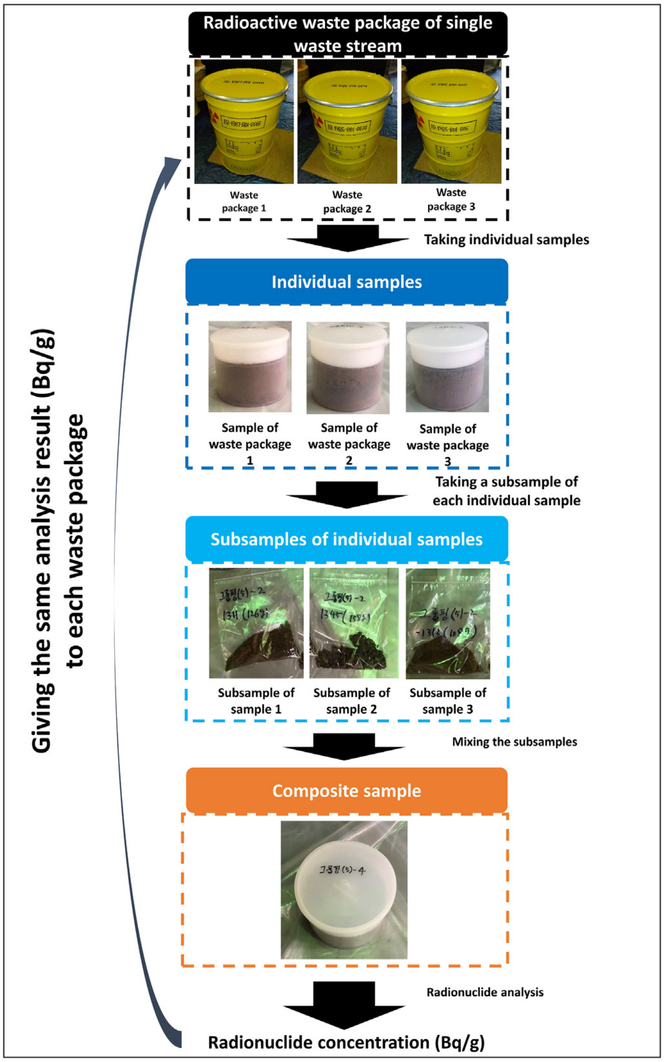


Fig. 2. Schematic diagram of grouping analysis.

In this study, therefore, experiments were conducted using radioactive soil and concrete waste, which are representative decommissioning waste [14], and dry active waste generated during the operation of nuclear facilities [15]. Through the experiments, we evaluated whether the waste from similar environments would be identified to be a single waste stream, and the uniformity of the radionuclide concentrations in individual samples taken from the waste packages. In addition, the adequacy of the grouping analysis of the waste, the subject of experiment, was verified using statistical experiments, and the possibility of expanding the range of grouping analysis, which is five packages, presented in the waste acceptance criteria was reviewed by conducting the same statistical experiments with larger amounts of waste.

2. Materials and experimental methods

2.1. Selection of experimental waste and measurement of radionuclide concentrations

To verify the adequacy of grouping analysis, the dry active waste (vinyl) and the radioactivity contaminated soil waste generated from the nuclear facilities of KAERI, and the concrete waste from dismantling of the research reactor (Triga Mark) in Gongneung-dong, Seoul that satisfied the conditions for similar environments were selected as experimental wastes. The detailed characteristics of the experimental waste are described in Table 1. A high-purity germanium (HPGe) detector (GC2018, Canberra) was used for the analysis of gamma-ray-emitting nuclides, which are major radionuclides in the experimental waste, and certified reference material (CRM) consisting of gamma-ray-emitting mixed nuclides (10 types) produced by the Korea Research Institute of Standards and Science (KRISS) was used to calibrate the energy and efficiency of the equipment. Detailed information on the CRM used for calibration is given in Table 2. In addition, to conservatively measure the concentrations of radionuclides, in the case of dry active waste and soil waste, individual samples were taken from parts with high levels of surface contamination of the waste packages and four samples were collected per waste package. In the case of concrete waste, samples already collected as part of a waste disposal task were used in this study in consideration of the schedule of the Seoul Research Reactor Decommissioning Project and field situations, and two samples per waste package were collected. Individual samples were collected in 1000 mL Marinelli beakers, and the measurement was carried out for 1 h using HPGe.

2.2. Determination of single waste stream from similar environments

The U.S. Nuclear Regulatory Commission (NRC) defines the waste with relatively uniform radiological and physical properties as the waste corresponding to a single waste stream, and judges that waste streams are considered distinct if the concentrations of major radionuclides typically differ by more than a factor of 10 [16]. Since the waste from similar environments selected in this study has the same physical characteristics, it can be judged to correspond to a single waste stream if it shows radiologically uniform characteristics. To identify the foregoing, it was evaluated whether the concentrations of major radionuclides in individual samples of waste from similar environments were distributed within 10 times, which is the criterion for judgment of a single waste stream.

2.3. Review of the uniformity of radionuclide concentrations in individual samples

Before preparing the composite sample, the uniformity of the radionuclide concentrations in individual samples should be checked. Therefore, one individual sample of soil waste among soil and concrete waste, which is particulate matter with relatively uniform characteristics, and one individual sample of dry active waste were selected to carry out a uniformity review. To that end, one individual sample was divided into 10 subsamples, and each of them was collected in a 100 mL cylindrical plastic beaker and analyzed to check whether the distribution of radionuclide concentrations was uniform. The measurement of the radionuclide concentrations was carried out for 1 h using HPGe, and the U.S. NRC's criterion to judge single waste streams was used to judge the uniformity.

2.4. Experiment to verify the adequacy of grouping analysis

2.4.1. Reference value setting

In this experiment, reference value setting is important to confirm that the results of analysis of composite samples are not underestimated or overestimated compared to the results of analysis of individual samples. A report [17] published by the Korea Institute of Nuclear Safety (KINS) presents the criterion for the accuracy of prediction when indirectly evaluating the concentrations of radionuclides as “predicted values do not deviate from the range of 0.1–10 times the actual values”. Since grouping analysis corresponds to an indirect evaluation method for the concentrations of radionuclides in individual samples, the results of analysis of individual samples become actual values and the results of analysis of composite samples become predicted values. Referring to the foregoing, the judgment criterion was set to “[The results of analysis of composite samples/the result of analysis of individual samples] is within 0.1–10”.

2.4.2. Selection of combinations for grouping analysis

To select the combinations of individual samples for grouping analysis, 30 random sample combinations were selected from among all possible combinations (${}_{20}C_r$) that can be made according to the grouping quantities (r). The sample combinations were selected with a sampling method using Monte Carlo probability distribution [18], and the number of sample combinations was selected as the minimum number of samples ($n = 30$) for which the central limit theorem [19] is statistically established for statistical analysis.

2.4.3. Composite sample preparation and analysis

Composite samples were prepared in the 30 combinations selected as sample combinations and collected in 450 mL Marinelli beakers. At this time, the final weights of the composite samples based on the average specific gravity of the experimental waste were calculated according to Eq. (1) and the quantities of samples that must be taken from individual samples were set using Eq. (2).

Table 1
Characteristics of the experimental radioactive waste.

Waste type	Number of waste package (200L)	History of the radioactive waste generation	Major radionuclides	Waste classification	Single waste stream conditions	Methods of pre-treatment
Soil	20	Contaminated radioactive soil	Cs-137	Very low-level	Similar contaminated area	Particle size classification (<20 mm)
Concrete	20	Radioactive concrete waste generated by dismantling research reactor “Triga Mark”	Co-60	Very low-level	Same generation place (shielding concrete)	Crushing
Dry active waste (Vinyl)	20	Dry active waste generated by operating nuclear facilities at KAERI	Co-60, Cs-137	Low-level	Same generation place and date	Cutting

Table 2
Specification of the certified reference material (CRM).

Source type and size	Serial No.	Radionuclides										
			Am-241	Cd-109	Co-57	Ce-139	Cr-51	Sn-113	Sr-85	Cs-137	Co-60	Y-88
Cylindrical plastic beaker (100 mL)	192 PB 100-1	Certified Values (Bq)	1,747	8,651	356	492	36,089	1,120	1,394	780	1,014	2,139
		Uncertainties (%)	4.0	4.0	4.2	4.1	4.2	4.0	4.0	4.1	4.0	4.0
Marinelli beaker (450 mL)	202MIX0303	Certified Values (Bq)	1,044	5,620	272	319	33,581	876	1,093	483	693	1,657
		Uncertainties (%)	4.0	4.1	4.0	4.1	4.2	4.0	4.0	4.1	4.0	4.0
Marinelli beaker (1,000 mL)	202MIX0128	Certified Values (Bq)	1,091	5,875	284	334	35,104	915	1,143	505	725	1,732
		Uncertainties (%)	4.0	4.1	4.2	4.2	4.0	4.0	4.0	4.2	4.0	4.0

$$X(g) = 450(mL) \times \frac{\frac{\sum_{i=1}^{n} Y_i}{n}}{1,000(mL)} \qquad (1)$$

$$K_i(g) = X \times \left(\frac{Y'_i}{\sum_{i=1}^{r} Y'_i}\right) \qquad (2)$$

where X is the final weight of the composite sample (g), Y_i is the weight of the i-th individual sample (g), and n is the total number of individual samples. In addition, K_i is the weight that must be taken from the i-th individual sample, r is the number of individual samples being grouped, and Y'_i refers to the weight of the i-th individual sample in the grouping combination of r pieces of individual samples. The composite sample collected in a 450 mL Marinelli beaker was measured for 1 h using HPGe. The detailed procedures related to the preparation and analysis of composite samples are shown in Fig. 3.

2.4.4. Sample data collection

Sample data for statistical verification were collected using the results of analysis of 30 composite samples. For conservative verification, 30 each of the minimum and maximum values among the values of "composite sample analysis results/individual sample analysis results" in the combinations for grouping analysis were selected as sample data. At this time, the minimum value samples were used for statistical analysis of underestimation and the maximum value samples were used for statistical analysis of overestimation. An example of sample data collection is shown in Table 3.

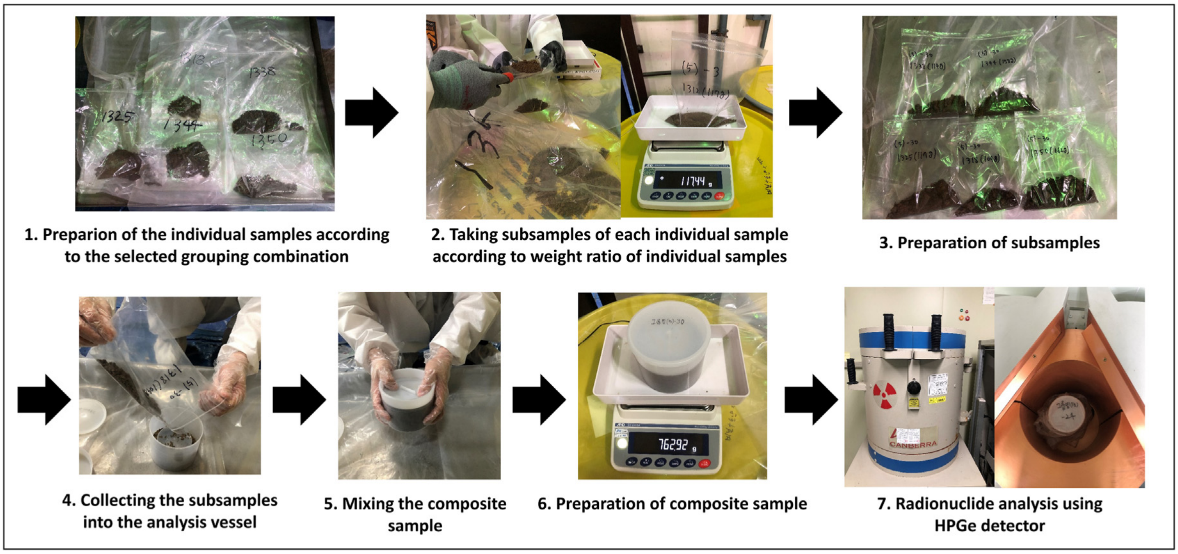
2.4.5. Verification of the adequacy of grouping analysis

The population mean confidence interval was estimated at a 95% confidence level [20] using the collected sample data. Here, the population mean is the mean value of the population, and the population is the group of minimum and maximum values among the values of "composite sample analysis results/individual sample analysis results" within the group for all cases ($_{20}C_r$) where r pieces each of packages are grouped and analyzed. The estimated values (μ) of the population mean interval calculated through the experiment were obtained for the minimum values and the maximum values. In the case of the minimum values, the radionuclide concentration was judged to have been underestimated when $\mu < 0.1$, and in the case of the maximum values, the radionuclide concentration was judged to have been overestimated when $\mu > 10$. In addition, when $0.1 < \mu < 10$, the grouping analysis was judged to be adequate. A diagram of the overall procedure of the verification of the adequacy of the grouping analysis is shown in Fig. 4.

3. Results and discussion

3.1. Judgment of single waste stream from similar environments

Using the results of analysis of individual samples collected from the experimental waste packages, the distribution of radionuclides concentrations in individual waste packages is shown as a schematic box plot [21,22] in Fig. 5. At this time, the whiskers of the boxplot extend to the most extreme data within the inner fences set using the distributions of radionuclide concentrations, and values outside the inner fences were regarded as outliers [23].



1. Preparion of the individual samples according to the selected grouping combination
2. Taking subsamples of each individual sample according to weight ratio of individual samples
3. Preparation of subsamples
4. Collecting the subsamples into the analysis vessel
5. Mixing the composite sample
6. Preparation of composite sample
7. Radionuclide analysis using HPGe detector

Fig. 3. Composite sample preparation and analysis procedure.

Table 3
Example of the sample data collection (5-package grouping case).

Analysis result (Bq/g)						Sample data ($X_{1,min}$)	Sample data ($X_{1,max}$)
Individual sample 1	Individual sample 2	Individual sample 3	Individual sample 4	Individual sample 5	Composite sample 1		
a	b	c	d	e	A	Min (A/a, A/b, A/c, A/d, A/e)	Max (A/a, A/b, A/c, A/d, A/e)

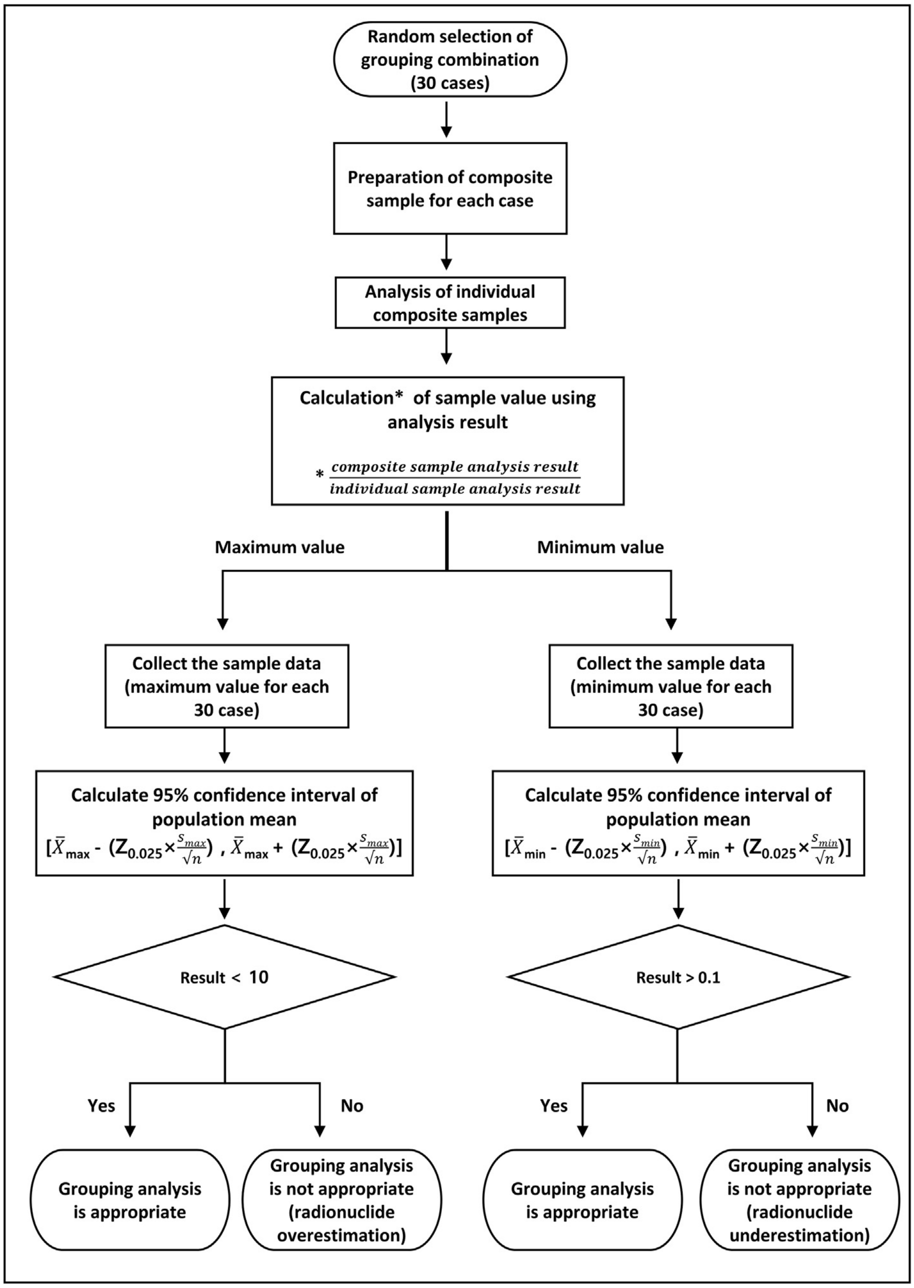


Fig. 4. Process for the verification of grouping analysis.

Fig. 5a and b shows the distributions of radionuclide concentrations in the soil waste individual sample group (Soil Individual, SI) and the concrete waste individual sample group (Concrete Individual, CI), respectively. One outlier was identified in each of SI-1–4 and CI-1, and two outliers were identified in CI-2. However, it was confirmed that the distributions of radionuclide concentrations were included within 10 times even when outliers were included for all sample groups, and that they are values that satisfy the criterion for a single waste stream presented by the U.S. NRC. Through this, it can be judged that all the experimental soil waste and concrete waste correspond to a single waste stream under the condition of similar environments.

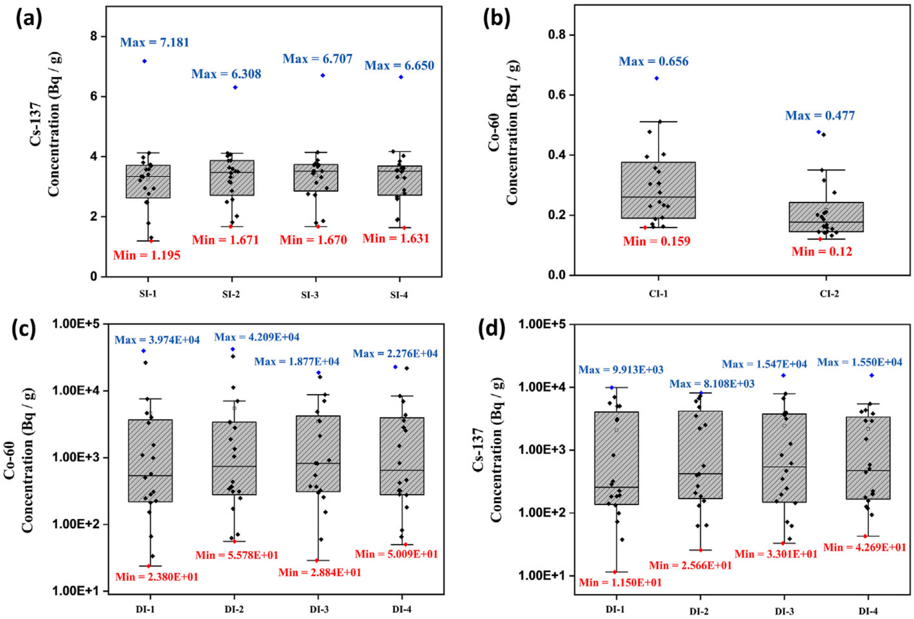


Fig. 5. Result of major radionuclide concentration in individual sample groups of (a) soil waste, (b) concrete waste, (c) dry active waste (Co-60) and (d) dry active waste (Cs-137).

Fig. 5c and d shows the distributions of radionuclide concentrations (Co-60, Cs-137) of the groups of dry active waste individual sample (Dry active waste Individuals, DI), respectively. In the case of the distribution of Co-60 concentrations, two outliers were found in each of DI-1,3,4, and three outliers were found in DI-2, and in the case of the distribution Cs-137 concentrations, one outlier was found in each of DI-3,4. According to the results of the review, it could be seen that the distributions of the concentrations of both of the radionuclides were out of 10 times even when outliers were excluded. Unlike soil and concrete, these values deviate from the criterion for judgment of a single waste stream of the U.S. NRC, and the experimental dry active waste is judged to correspond to distinct waste streams based on the U.S. NRC criterion, even if they are from similarly occurring environmental conditions. The reason is considered to be the fact that for dry active waste, even if the time of occurrence and properties (vinyl) are the same, the radioactive contamination routes cannot be completely the same due to differences in the working time and working environment where detailed contents (vinyl gloves, vinyl masks, etc.) were used, and there are limitations in controlling them to be identical or homogenizing them during collection so that the distributions of radionuclide concentrations are not equal among individual samples. Table 4 shows the box plot data for the distributions of nuclide concentrations in individual samples of soil waste, dry active waste, and concrete waste packages in detail.

3.2. Review of uniformity of radionuclide concentrations in individual samples

Table 5 shows the results of examination of the distributions of radionuclide concentrations in individual subsamples prepared using individual samples of soil waste and dry active waste. On reviewing Table 5, it can be seen that the distribution of radionuclide concentrations in the subsamples (Soil Homogeneity, SH) of the individual soil sample is 2.818E+00–3.471E+00 Bq/g, and the value of "maximum value/minimum value" is 1.232. In addition, it can be seen that the distributions of radionuclide concentrations in the subsamples (Dry active waste Homogeneity, DH) of the individual dry active waste sample were 1.554E+02–2.630E+02 Bq/g in the case of Co-60, and 1.511E+03–3.104E+03 Bq/g in the case of Cs-137 and that the value of "maximum value/minimum value" is 1.693 for Co-60 and 2.054 for Cs-137. According to the results of examination of the distributions of the radionuclide concentrations, all the distributions were included within 10 times, thereby satisfying the U.S. NRC criterion for a single waste stream so that the distributions of radionuclide concentrations in individual samples can be judged to be uniform. In particular, in the case of dry active waste, when the distributions of radionuclide concentrations among the individual samples were reviewed, they were judged to be in distinct waste streams (Fig. 5c and d), but the distributions of radionuclide concentrations in one individual sample were judged to correspond to a single waste stream. Therefore, it is considered that the radionuclide concentrations in individual samples are distributed relatively uniformly because the nuclide concentrations in subsamples are generally similar since individual samples were selectively collected from parts with high surface contamination levels.

3.3. Verification of the adequacy of grouping analysis

Fig. 6 shows the results of estimation of the population means at the 95% confidence level using the sample data collected after conducting grouping analyses of soil waste, concrete waste, and dry active waste, respectively. Fig. 6a is the results of verification of individual sample groups (SI-1, CI-1, DI-1) of soil waste, concrete waste, and dry active waste by conducting grouping analyses of five waste packages. In the case of soil waste, the population mean was estimated to be distributed between 0.746 and 0.86 when the minimum values were evaluated and between 1.767 and 2.07 when the maximum values were evaluated, and in the case of concrete waste, the population mean was estimated to be distributed between 0.474 and 0.595 when the minimum values were evaluated and between 1.268 and 1.539 when the maximum values were evaluated. All of these results satisfy the reference values for verification of the adequacy of grouping analysis, so that it can be considered that the composite samples would not be underestimated or overestimated at the 95% confidence level in any case of grouping five waste packages of the individual samples of soil

Table 4
Box plot data for the distribution of radionuclides concentrations in individual samples of each waste type.

Waste type (major radionuclides)	Sample group ID[a]	Quartiles of the distribution of radionuclide concentrations			Inner fences (Q_1 -1.5 × IQR[b], Q_3 +1.5 × IQR)		Whiskers of box plot[c]		outlier
		first quartile (Q_1)	second quartile (Q_2)	third quartile (Q_3)	Lower fence	Upper fence	Lower whisker	Upper whisker	
Soil (Cs-137)	SI-1	2.624E+00	3.338E+00	3.709E+00	9.959E-01	5.338E+00	1.195E+00	4.123E+00	7.181E+00
	SI-2	2.715E+00	3.476E+00	3.868E+00	9.855E-01	5.598E+00	1.671E+00	4.112E+00	6.308E+00
	SI-3	2.854E+00	3.514E+00	3.737E+00	1.529E+00	5.062E+00	1.670E+00	4.143E+00	6.707E+00
	SI-4	2.717E+00	3.515E+00	3.688E+00	1.260E+00	5.145E+00	1.631E+00	4.171E+00	6.650E+00
Concrete (Co-60)	CI-1	1.898E-01	2.601E-01	3.759E-01	−8.926E-02	6.550E-01	1.589E-01	5.114E-01	6.561E-01
	CI-2	1.450E-01	1.769E-01	2.428E-01	−1.707E-03	3.895E-01	1.205E-01	3.502E-01	4.772E-01, 4.677E-01
Dry active waste (Co-60)	DI-1	2.186E+02	5.358E+02	3.664E+03	−4.949E+03	8.832E+03	2.380E+01	7.548E+03	2.640E+04, 3.974E+04
	DI-2	2.790E+02	7.375E+02	3.394E+03	−4.394E+03	8.067E+03	5.578E+01	7.025E+03	4.209E+04, 3.282E+04, 1.122E+04
	DI-3	3.103E+02	8.210E+02	4.204E+03	−5.531E+03	1.005E+04	2.884E+01	8.755E+03	1.610E+04, 1.877E+04
	DI-4	2.789E+02	6.453E+02	3.964E+03	−5.249E+03	9.492E+03	5.009E+01	8.377E+03	2.276E+04, 2.177E+04
Dry active waste (Cs-137)	DI-1	1.367E+02	2.558E+02	4.057E+03	−5.744E+03	9.938E+03	1.150E+01	9.913E+03	–
	DI-2	1.703E+02	4.194E+02	4.172E+03	−5.832E+03	1.017E+04	2.566E+01	8.108E+03	–
	DI-3	1.491E+02	5.437E+02	3.755E+03	−5.261E+03	9.165E+03	3.301E+01	7.907E+03	1.547E+04
	DI-4	1.666E+02	4.737E+02	3.367E+03	−4.633E+03	8.166E+03	4.269E+01	5.458E+03	1.550E+04

[a] SI: Soil Individual, CI: Concrete Individual, DI: Dry active waste Individual.
[b] IQR (Interquartile Range): $Q_3 - Q_1$.
[c] Lower whisker: minimum value within inner fence, Upper whisker: maximum value within inner fence.

Table 5
Results of radionuclide concentration uniformity in individual waste samples.

Waste type	Sample group ID[a]	Major radionuclides	Analysis result (Bq/g)	X±SD[b] (Bq/g)	Minimum value (Bq/g)	Maximum value (Bq/g)	Max/Min
Soil	SH-1	Cs-137	3.294E+00	3.09E+00 ± 2.019E-01	2.818E+00	3.471E+00	1.232
	SH-2		3.075E+00				
	SH-3		3.136E+00				
	SH-4		3.471E+00				
	SH-5		3.159E+00				
	SH-6		3.082E+00				
	SH-7		2.836E+00				
	SH-8		2.818E+00				
	SH-9		3.123E+00				
	SH-10		2.906E+00				
Dry active waste	DH-1	Co-60	2.466E+02	2.102E+02 ± 4.398E+01	1.554E+02	2.630E+02	1.693
	DH-2		1.844E+02				
	DH-3		2.057E+02				
	DH-4		1.554E+02				
	DH-5		2.619E+02				
	DH-6		2.630E+02				
	DH-7		2.013E+02				
	DH-8		1.642E+02				
	DH-9		1.612E+02				
	DH-10		2.585E+02				
	DH-1	Cs-137	3.093E+03	2.226E+03 ± 5.798E+02	1.511E+03	3.104E+03	2.054
	DH-2		2.624E+03				
	DH-3		2.143E+03				
	DH-4		1.660E+03				
	DH-5		3.104E+03				
	DH-6		2.506E+03				
	DH-7		2.047E+03				
	DH-8		1.715E+03				
	DH-9		1.858E+03				
	DH-10		1.511E+03				

[a] SH: Soil Homogeneity, DH: Dry active waste Homogeneity.
[b] X±SD: mean of sample analysis results ± standard deviation.

waste and concrete waste to be verified. Therefore, the grouping analysis was judged to be adequate. On the other hand, with regard to dry active waste, when the minimum values were evaluated, the population mean was estimated to be distributed between 0.201 and 0.278 for Co-60 and 0.292 to 0.405 for Cs-137, so that it was confirmed that all distributions were included within the reference values. However, when the maximum values were evaluated, the population mean was estimated to be distributed between 27.047 and 71.466 for Co-60 and between 37.375 and 101.365 for Cs-137, so that it was identified that all the distributions deviated from the reference values. Therefore, in the case of the experimental dry active waste, the concentrations of radionuclides were found to be overestimated during the 5-package grouping analysis and so it was judged that it is somewhat difficult to apply the grouping analysis. The reason for this is thought to be the fact that unlike soil and concrete waste, the distributions of radionuclide concentrations in dry active waste are not uniform among individual samples (Fig. 5c and d) so that the radionuclide concentration in the

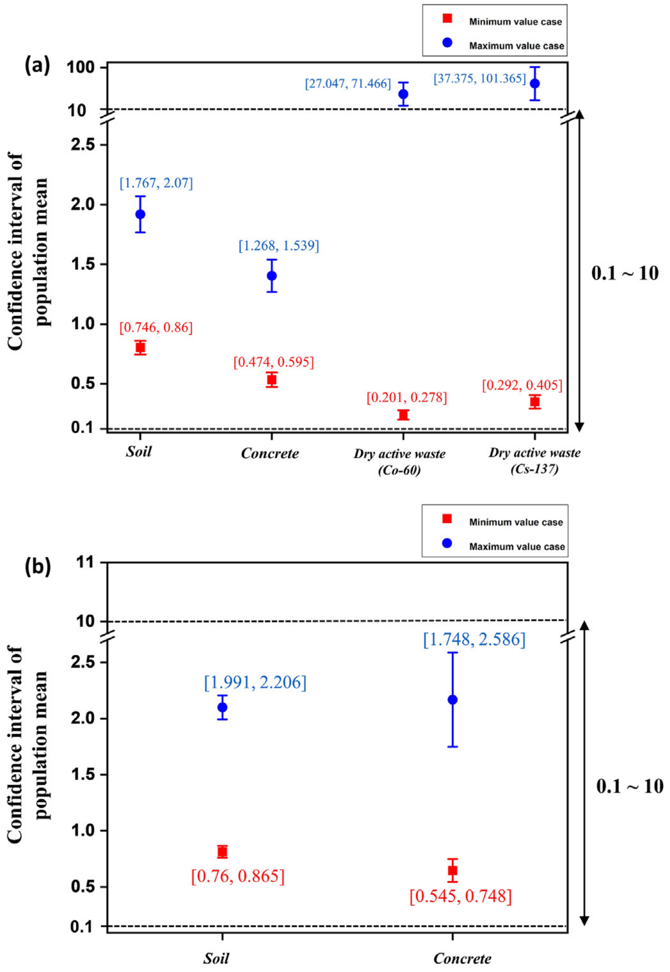


Fig. 6. Results of estimation of population mean interval of each waste type in (a) 5-package grouping analysis case and (b) 10-package grouping analysis case.

composite samples will be relatively high when individual samples with high nuclide concentrations are included in the group, and cases where the nuclide concentrations in composite samples are overestimated compared to some individual samples in the group occur.

Furthermore, 10-package grouping analyses were additionally conducted with individual sample groups (SI-2, CI-2) of soil waste and concrete waste for which 5-package grouping analysis was judged adequate, and the population mean interval was estimated using the collected sample data. (Fig. 6b). According to the results of the 10-package grouping analyses, with regard to soil waste, the population mean was estimated to be distributed between 0.76 and 0.865 when the minimum values were evaluated and between 1.991 and 2.206 when the maximum values were evaluated, and in the case of concrete waste, the population mean was estimated to be distributed between 0.545 and 0.748 when the minimum values were evaluated and between 1.748 and 2.586 when the maximum values were evaluated. These results satisfied all of the reference values for the verification of the adequacy of grouping analysis. Therefore, in the case of soil and concrete waste, grouping analysis was judged to be adequate even when the number of waste packages for grouping was expanded to 10. Table 6 describes the resultant values of estimation of population mean intervals using the results of grouping analysis in detail.

4. Conclusion

In this study, to verify the adequacy of grouping analysis, experiments were conducted using radioactive waste generated in similar environments. The distributions of radionuclide concentrations in individual samples of each waste were examined, and according to the results, soil waste and concrete waste were evaluated to correspond to a single waste stream, and dry active waste was evaluated to correspond to distinct waste streams. In addition, the distributions of radionuclide concentrations in subsamples obtained by dividing individual samples of soil waste and dry active waste were examined, and it was verified that the distributions of radionuclide concentrations of individual samples were uniform in both wastes. 5-package grouping analysis was verified, and as a result, it was identified that with regard to soil and concrete waste, all the results of estimation of population means of the values of “composite sample analysis results/individual sample analysis results” were included in the reference values (0.1–10) set in this study and that for dry active waste, the values of “composite sample analysis results/individual sample analysis results” were estimated to be larger than 10, indicating that the radionuclide concentrations in composite samples were overestimated. Furthermore, the number of waste packages for grouping of soil waste and concrete waste was expanded to 10 and the grouping analysis was statistically verified according to the results, and it was identified that all the resultant values were included in the reference values. Therefore, it can be concluded that the 5-package grouping analysis presented in the waste acceptance criteria is adequate for the experimental soil waste and concrete waste, and that grouping analysis is adequate even when the range of analysis is expanded to 10-package. On the other hand, it was found that it was somewhat difficult to apply the 5-package grouping analysis using the statistical criteria and verification method presented in this paper in the case of the experimental dry active waste because the distributions of radionuclide concentrations appeared to not be uniform among individual samples even under similar environmental conditions. In order to apply the results of this study to practice, it is judged

Table 6
Resultant of population mean intervals in individual waste samples.

Number of grouping	Waste type	Minimum value case				Maximum value case			
		Sample mean	Sample standard deviation	Standard error	95% confidence interval of population mean	Sample mean	Sample standard deviation	Standard error	95% confidence interval of population mean
5	Soil	0.803	0.16	0.057	[0.746, 0.86]	1.918	0.423	0.151	[1.767, 2.07]
	Concrete	0.535	0.169	0.061	[0.474, 0.595]	1.403	0.378	0.135	[1.268, 1.539]
	Dry active waste (Co-60)	0.239	0.108	0.039	[0.201, 0.278]	49.256	62.064	22.209	[27.047, 71.466]
	Dry active waste (Cs-137)	0.348	0.158	0.057	[0.292, 0.405]	69.37	89.41	31.995	[37.375, 101.365]
10	Soil	0.812	0.147	0.053	[0.76, 0.865]	2.099	0.3	0.107	[1.991, 2.206]
	Concrete	0.646	0.283	0.101	[0.545, 0.748]	2.167	1.171	0.419	[1.748, 2.586]

necessary to conduct additional studies with experiments using diversified experimental waste, and methods to control similar environments for dry active waste and homogenize them. Based on the study, it can be expected that if grouping analysis is established in practice, the time required for analysis and worker exposure can be reduced through increases in the efficiency of waste analysis, and consequently the final disposal of radioactive waste, which is expected to be generated in large quantities, will be actively promoted.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by a research grant from the Korea Atomic Energy Research Institute (KAERI) [Grant No. 521220-21, South Korea] and [Grant No. 521240-21, South Korea] and Nuclear Research & Development Program (Grant No. 2019M2C9A1059067) of the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (MIST).

References

[1] Y.-J. Choi, S.-C. Lee, C.-L. Kim, Proposal for effective disposal options of very low level decommissioning waste, Prog. Nucl. Energy 94 (2017) 36–45, https://doi.org/10.1016/j.pnucene.2016.10.001.

[2] Korea Institute of Nuclear Safety, Status of Radioactive Waste Storage at the 4th Quarter of 2020, Waste Comprehensive Information Database, Korea Institute of Nuclear Safety, 2021. http://www.kins.re.kr/wacid.

[3] S.-C. Bae, J. Park, Status and research progress of nuclear decommissioning concrete waste disposal, J. Korean Recycl. Constr. Resour. Inst. 15 (2020) 45–51, https://doi.org/10.14190/MRCR.2020.15.2.045.

[4] International Atomic Energy Agency, Characteristics of Radioactive Waste Forms Conditioned for Storage and Disposal: Guidance for the Development of Waste Acceptance Criteria, IAEA-TECDOC-285, International Atomic Energy Agency, Vienna, 1983.

[5] M.H. Ahn, S.C. Lee, K.J. Lee, Disposal concept for LILW in Korea: characterization methodology and the disposal priority, Prog. Nucl. Energy 51 (2009) 327–333, https://doi.org/10.1016/j.pnucene.2008.07.001.

[6] Nuclear Safety and Security Commission, Regulations for the Waste Acceptance of Low and Intermediate Level Radioactive Waste, 2021. Notice No. 2021-26.

[7] J.W. Yeon, B.M. Kang, N.U. Kim, Chemical Analysis of Radioactive Materials, No. KAERI/RR-4323/2017, Korea Atomic Energy Research Institute, 2018.

[8] K. Jee, H. Ahn, A Strategy on the Disposal of below Intermediate Level Radwastes (No. KAERI/RR-4041/2015), Korea Atomic Energy Research Institute, 2015.

[9] Korea Radioactive Waste Agency, Waste Acceptance Criteria for the 1st Phase Disposal Facility of the Wolsong Low- and Intermediate-Level Waste Disposal Center (No. WAC-SIL-2020-1), 2020.

[10] J.B. Park, J.T. Jeong, J.-W. Park, Development of the safety case program for the Wolsong low- and intermediate-level radioactive waste disposal facility in Korea, J. Nucl. Fuel Cycle Waste Technol. 12 (2014) 335–344, https://doi.org/10.7733/jnfcwt.2014.12.4.335.

[11] Nuclear Safety and Security Commission, Criteria for Structure and Equipment of the Low- and Intermediate-Level Radioactive Waste Repository, 2017. Notice No. 2017-59.

[12] J.B. Park, H.R. Jung, E.Y. Lee, C.L. Kim, G.Y. Kim, K.S. Kim, Y.K. Koh, K.W. Park, J.H. Cheong, C.W. Jeong, J.S. Choi, K.D. Kim, Wolsong low-and intermediate-level radioactive waste disposal center: progress and challenges, Nucl. Eng. Technol. 41 (2009) 477–492, https://doi.org/10.5516/NET.2009.41.4.477.

[13] Y. Shin, J. Lee, The status and experiences of LILW disposal facility construction, J. Korean Soc. Miner. Energy Resour. Eng. 54 (2017) 389–396, https://doi.org/10.12972/ksmer.2017.54.4.389.

[14] International Atomic Energy Agency, Disposal Aspects of Low and Intermediate Level Decommissioning Waste: Results of a Coordinated Research Project 2002-2006, IAEA-TECDOC-1572, International Atomic Energy Agency, Vienna, 2007, http://www-pub.iaea.org/MTCD/publications/PDF/TE_1572_web.pdf.

[15] International Atomic Energy Agency, Classification of Radioactive Waste, IAEA Safety Standards Series No. GSG-1, International Atomic Energy Agency, Vienna, 2009.

[16] US Nuclear Regulatory Commission, Concentration Averaging and Encapsulation Branch Technical Position, Revision 1, vol. 1, Federal Register 80, US Nuclear Regulatory Commission, Washington, DC, 2015.

[17] K.J. Lee, M.C. Song, G.H. Hwang, C.M. Lee, D.S. Yuk, S.C. Lee, Research on the Assessment Technology of the Radionuclide Inventory for the Radioactive Waste Disposal (No. KINS/HR-590), Korea Institute of Nuclear Safety, 2004.

[18] W.K. Hastings, Monte Carlo sampling methods using Markov chains and their applications, Biometrika 57 (1970) 97–109, https://doi.org/10.1093/biomet/57.1.97.

[19] H. Kim, J. Bae, M. Lee, S. Lee, E. Lee, J. Lee, Introduction to Statistics, third ed., Jungik, Korea, 2019.

[20] S.-H. Kim, J. Yeom, I.-S. Baek, J.-S. Kim, S.-I. Sung, Determining the statistical sample size for reliability testing, J. Appl. Reliab. 20 (2020) 84–93, https://doi.org/10.33162/jar.2020.3.20.1.84.

[21] J.W. Tukey, Exploratory Data Analysis, Addison-Wesley, United States, 1977.

[22] K. Potter, H. Hagen, A. Kerren, P. Dannenmann, Methods for Presenting Statistical Information: the Box Plot, Vis. Large Unstructured Data Sets, vol. 4, 2006, pp. 97–106.

[23] R. Dawson, How significant is a boxplot outlier? J. Stat. Educ. 19 (2) (2011) https://doi.org/10.1080/10691898.2011.11889610.