Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 18-19, 2023

Evaluation of Leaching Test Method for Gyeongju Low- and Intermediate Level Waste Repository

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1. Introduction

Korea is operating a silo-type low- and intermediate-level radioactive waste repository, a concrete structure with a thickness of 1 to 1.6 meters and a height of 50 meters, 130 meters underground in Gyeongju. In addition, the vault-type repository is under construction that dispose of radioactive waste under ~20 m depth. To dispose of radioactive waste in the repository, the waste acceptance criteria should be satisfied, and the test requirements for solidified waste are compressive strength, irradiation, leaching, immersion, and thermal cycling test. Among the tests, leachability index should be at least 6 or higher for Cs, Sr, and Co nuclides [1].

ANS 16.1, a leaching test method of waste acceptance criteria for Gyeongju LILW repository, applies distilled water as leachant. Since Gyeongju LILW repository structures are mainly composed of concrete, the actual leachant inside the repository is expected to be highly alkaline due to reaction between groundwater and concrete structures. Therefore, the current leaching test method of Gyeongju LILW repository may not sufficiently reflect the actual disposal environment. This study compares the disposal environment and leaching test methods of foreign countries operating radioactive waste repository, and discusses enhancement direction of leaching test methods reflecting the disposal condition of Gyeongju LILW repository.

2. Radioactive waste repository and leaching test methods of foreign countries

2.1. Radioactive repositories

France is currently operating a low- and intermediate-level waste disposal facility called the Aube disposal facility. The type of repository is a near-surface disposal, and a vault-type disposal warehouse is operated on the protrusion of sedimentary rock composed of a semi-permeable layer (white sand) covering the impermeable layer (clay) [2].

The United States is operating four low- and intermediate-level waste disposal facilities. Among them, the Barnwell disposal facility is a near-surface repository type, and it is operated in the form of loading waste into a trench that excavated a depth of 9 m, filling it with sand or soil, and installing a multi-layer cap of sand, clay, and high-density polyethylene [3].

Figure 1. Aube disposal facility (ANDRA)

Figure 2. Barnwell disposal facility (Energysolutions)

2.2. Leaching test methods

According to the repository, different leaching test methods are considered where most of them reflects the actual disposal condition of their own repository. [4].

- · **IAEA**: Leaching test Methods Proposed by the International Atomic Energy Agency
- · **ANS 16.1**: Leaching test method proposed by the American Nuclear Society (Leaching test method of Gyeongju disposal facility)
- · **FT-04-020**: Leaching test Method Proposed in France
- · **ASTM C1308**: Leaching test method proposed by the American Society for Testing and Materials [5]
- · **EPA 1315**: Leach test method proposed by the United States Environmental Protection Agency [6]

3. Comparison of leaching test methods

3.1. Dimensions and types of test specimen

Table I shows the size and shape criteria of the specimen required in each leaching test method.

Table I: Size and shape criteria of the specimen

Categories	Size	Shape
IAEA	L=D=5 cm (> 1 Ci/L) L=D=2.5 cm (< 1 Ci/L)	Cylinder
ANS 16.1	L=D > 1 cm	Cylinder, Cube, Sphere
FT-04-020	L=D=8 cm L=75, D=60 cm	Cylinder, Cube
ASTM C1308	L=D=2.5 cm	Cylinder
EPA 1315	L=D > 5 cm	Cylinder, Cube, Rectangular, Wafer

From table I, it can be confirmed that all leaching test methods require cylindrical test specimens and the size of the specimen that derives the sufficient leaching rate.

Each leaching test method determines the range of applicable solidification material. Table II shows the range of solidification materials.

Table II: Range of solidification materials

IAEA	Cement, asphalt, glass mixtures and melt
ANS 16.1	Solidified of waste that didn't generate heat
FT-04-020	Cement, asphalt, plastic
ASTM C1308	Any solid that does not deform during the test
EPA 1315	Monolith, compacted granular materials which behave as a monolith

From Table II, it can be confirmed all leaching test methods require solidification, and these solids should prevent deformation during the leaching test. From those materials, cement solidification is the most common solidification material which is shown in all leaching test method.

3.2. Leachant

The type of leachant differs according to the leaching test method. Table III shows the type of leachant in each leaching test methods.

Table III: Type of leachant

IAEA	Distilled water
ANS 16.1	Distilled water, synthetic seawater
FT-04-020	Standard water that matches groundwater composition at disposal site
ASTM C1308	ASTM Type I water, synthetic or actual groundwater, chemical solution capable of evaluating leaching
EPA 1315	Reagent water, solutions that can evaluate leaching behavior under specific environmental conditions

3.3. Test methods

Table IV shows the number and frequency of leachant exchange in each leaching test method.

Table IV: Number and frequency of leachant exchange

IAEA	21 times / 240 days (every day (7 times), once a week (8 times), once a month (6 times))
ANS 16.1	5 times / 5 days (once a day)
	10 times / 90 days (2, 7, 24, 48, 72, 96, 120, 456, 1128, 2160 h accumulated)
FT-04-020	6 times / 275 days (15, 30, 60, 90, 180, 275 d accumulated)
ASTM C1308	11 times / 11 days (once a day)
EPA 1315	9 times (additional exchange is possible every 14 days to check long-term behavior)

In each leaching test method, there is a slight difference in the surface area of specimen exposed to leachant. ANS 16.1, FT-04-020, ASTM C1308 of the leaching test methods specify that all sides of the specimen are exposed, the IAEA specifies that only the upper section is exposed, and EPA 1315 specifies that more than 98% of the specimen's surface is exposed to leachant.

4. Discussion

In Gyeongju LILW repository, the silo-type repository is in operation and vault-type repository is under construction. Among the test methods for Gyeongju LILW repository, leaching test methods follows ANS 16.1 which applies distilled waste as leachant. However, the expected leachant inside the repository is alkaline groundwater from the reaction between groundwater and concrete structures inside the repository. Thus, the compatibility of leaching test method is discussed in this study by investigating foreign repositories and leaching test methods.

The major type of investigated radioactive waste repository is near-surface type where the disposal depth is ~20 m. The leaching test methods introduced in this study are adopted for those near-surface type radioactive waste repositories.

The leaching test methods commonly require a specimen of cylindrical shape and size that can sufficiently measure the leaching rate. Most test methods recommend the use of leachant reflecting the actual disposal environment, and FT-04-020 and ASTM C1308 recommend the use of actual groundwater. However, the test period and the leachant exchange frequency in each leaching test method are different.

ANS 16.1, a leaching test method within the waste acceptance criteria for the silo and vault type repositories in Gyeongju, uses distilled water as leachant unlike foreign repositories. Although distilled water is generally used in leaching tests, nuclides in radioactive waste have different leaching behavior depending on the chemical conditions of leachant. As shown in figure 3 and figure 4, the solubility of Ce, Co, etc. decreases as the pH increases in the groundwater environment, while the solubility of Ni, Fe, etc. decreases to pH 9, and then the solubility increases when the pH increases larger than 9 [7, 8].

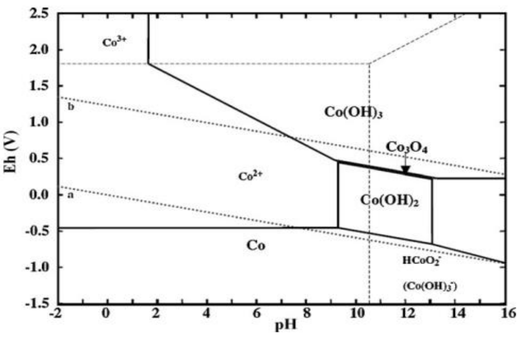

Figure 3. Pourbaix diagram of cobalt in H_2O system

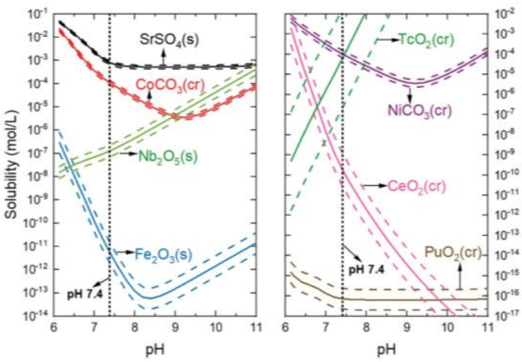

Figure 4. Solubility of nuclides according to the pH

Since leaching behavior of nuclides in radioactive waste differs depending on the chemical environment of leachant, the identification of the leaching behavior of major nuclides according to the actual leachant environment seems important. If sufficient data on leaching behavior of major nuclear species in the actual disposal environment is secured and reflected in the waste acceptance criteria, more reasonable and efficient radioactive waste disposal is expected. In addition, based on the improved waste acceptance criteria, the radioactivity concentration limit of the current disposal facility can be enhanced, which is expected to improve the economic feasibility of the disposal facility. Therefore, the leaching test reflecting actual disposal condition should be performed as future work to accumulate the leaching behavior database of several radionuclides.

5. Conclusion

In this study, the disposal environment and the leaching test method of foreign countries were investigated and compared. As a result, it was confirmed that most leaching test methods in countries that operate disposal facilities similar to domestic disposal facility recommend the use of leachant reflecting the disposal environment. Therefore, it seems necessary to secure leaching behavior data for each major radionuclides reflecting disposal condition of repository in the future work.

ACKNOWLEDGEMENT

This work was supported by the Nuclear Safety Research Program through the Korea Foundation of Nuclear Safety (KoFONS), granted financial resources from the Nuclear Safety and Security Commission (NSSC), Republic of Korea (No. 2203028).

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