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Stability Evaluation of Particle Generated from Scrubbed Concrete Waste under Simulated Concrete Pore Water Conditions of Underground Disposal

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1. Introduction

In South Korea, the Low- and Intermediate-Level Waste (LILW) disposal facility in Gyeongju has been operating since 2015. As of March 2025, 32,729.6 drums were stored in the current silo-type facility, which has a disposal capacity of 100,000 drums. An additional near-surface disposal facility, which has a disposal capacity of 125,000 drums, has been under construction. However, nuclear power plants temporarily stored about 100,000 drums of LILW, and 3,000 - 4,000 drums were generated annually. At this situation, the disposal facility could reach capacity within 20 years, even with the constructed site. If a nuclear power plant that has been in operation for more than 40 years is decommissioned, the disposal facility will not be able to accommodate all the decommissioning waste.

A significant portion of LILW from decommissioning was concrete waste with the form of particulate waste. According to disposal criteria, particulate-containing waste must be immobilized with cement before being taken over by the disposal facility. One of the immobilization techniques, cementation, generally had 20 - 30% waste loading, following the standard of KS F 2403. Immobilization ensured the stability of waste, but at the same time increased the volume of waste, reducing the efficiency of waste disposal. However, there was no study about evaluating the stability of particulate-containing concrete waste without additional cement immobilization. [1]

Specifically, the particulate waste might generate colloids (1 nm to 1 μm) through dissolution and aggregation interacting with rainwater or groundwater in the disposal site. The presence of colloids accelerated the migration of radionuclides in the disposal site. In general, the natural rock at the disposal site was negatively charged, causing cationic radionuclides to be readily adsorbed on the rock surface. However, when radionuclides interacted with colloids, the cationic species transitioned to negatively charged forms, making them difficult to be retained by the rock. Thus, the migration of positively charged radionuclides was accelerated within the disposal sites due to the presence of colloids. [2]

To address these gaps, this study evaluated colloid formation in particulate-containing concrete LILW in Gyeongju and its stability without additional immobilization. All experiments were conducted in a simulated environment of LILW disposal site in Gyeongju. These findings aimed to evaluate the stability of particulate-containing concrete waste without additional immobilization in radioactive disposal facilities.

2. Methodology

2.1 Representative Particulate-containing Concrete Waste

Concrete was chosen as representative LILW in this study because it was generated in large quantities during nuclear power plant decommissioning. During the waste drum packaging process, concrete was crushed to meet the waste acceptance criteria requiring at least 85% of drum capacity, which generates particulate waste. The concrete waste used in the experiment was sourced from the Gyeongju LILW disposal facility. To meet the particulate material criteria (≤ 0.2 mm constitutes at least 15 wt%), the concrete was crushed and sieved to a particle size of 173-185 μm. [1]

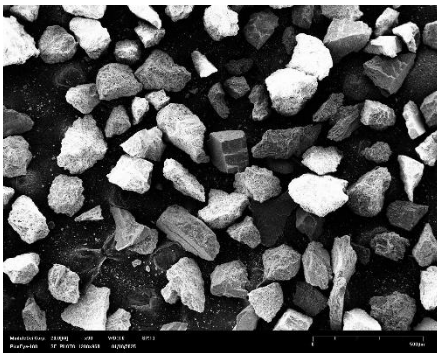

Figure 1. SEM image of crushed concrete particle

2.2 Experimental Setup

Since colloids are highly influenced by their surrounding environment, it is crucial to maintain experimental conditions similar to real conditions. For this study, the experimental environment was designed to simulate the conditions of the LILW disposal facility in Gyeongju, which was currently in operation. In the case of concrete waste, it was more likely to be disposed of in a near-surface disposal facility rather than a silo-type repository. Therefore, the composition of the leachate was shown in **Table 1**. [3]

Table 1. Composition of concrete pore water (pH 11.54)

Concrete Pore Water			
Ions	M (mol/L)	Ions	M (mol/L)
Na^+	3.47E-04	Si	1.69E-04
Ca^{2+}	1.32E-03	Cl^-	2.57E-04
K^+	1.11E-04	SO_4^{2-}	6.09E-05
Al^{3+}	5.15E-05	NO_3^-	1.69E-05

2.3 Experimental Conditions

The experiment was conducted under two different conditions: short-term experiments and accelerated experiments. Short-term experiments were designed to investigate the colloid formation mechanisms and stability at 20 °C over a period of 90 days. Accelerated experiments were performed by increasing the temperature to 60 °C to accelerate the reaction. The summarized experimental conditions were shown in **Table 2**. [4]

Table 2. Experimental conditions of the study

Test type	Temp	Waste	Period	Leachate
Short-term experiment	20 ℃	170 μm Concrete Powder	0-90 days	Concrete pore water
Accelerated experiment	60 ℃			

2.4 Experimental Procedure

The overall experimental setup was illustrated in **Figure 2**. The main experimental procedure was designed following ASTM C1220-21 and ISO 18772:2008. [4]

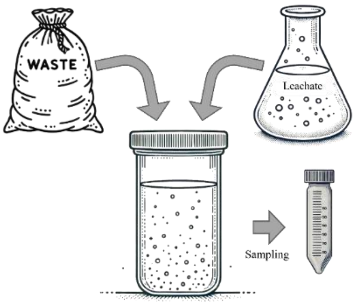

Figure 2. Overall experimental setup: the leachate and particulate concrete waste in a PP container for 90 days at different temperature

2.4.1 Experiment Preparation

A leachate solution was synthesized according to **Table 1** and stirred for 30 days to ensure the complete dissolution of all ions. Before the experiment, the leachate was filtered using a 0.45 μm of PTFE filter to remove remaining impurities. 11.25 g of crushed concrete particles were mixed with 200 mL of leachate in a PP container and maintained at a monitored temperature for 90 days.

2.4.2 Experiment Execution

Throughout the 90-day experiment, samples were periodically collected (0, 15, 30, 45, 60, 75, 90 days) for analysis. A total of 50 mL of the sample was collected at each sampling time. The samples were then centrifuged to remove particles larger than 1 μm, which was the range of colloids before analysis. The processed samples were analyzed using Dynamic Light Scattering (DLS) and Electrophoretic Light Scattering (ELS) techniques to measure particle size and zeta potential. In this study, Day 45 and Day 90 data were used for result analysis due to the highest measurement reliability.

Particle size was measured using DLS. DLS determines the hydrodynamic diameter of particles in an aqueous solution by analyzing their random motion due to Brownian movement and applying the Stokes-Einstein equation (1), which describes the relationship between particle size and movement. This method was used to measure the size of colloids.

$$D = \frac{k_B T}{3\pi\eta d_H} \tag{1}$$

Where D is the diffusion coefficient, k_B is the Boltzmann constant, T is the temperature, η is the viscosity, and d is the hydraulic diameter.

The stability of colloids is evaluated based on the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory, which explains the interaction between van der Waals attraction and electrostatic repulsion. Zeta potential is a key concept in DLVO theory, as it indirectly represents the surface potential of colloids. Zeta potential was measured using ELS, which precisely detects the Doppler effect. In general, colloids are considered stable when the absolute value of the zeta potential exceeds 30 mV. However, this value can vary depending on environmental factors such as pH and ionic strength.

At the end of the 90-day experiment, the concrete waste and leachate were separated using a vacuum filtration system with a 1 μm of PES membrane filter. The separated concrete waste was dried in a vacuum oven, sealed, and stored. The leachate was transferred to a thoroughly sterilized PP container, sealed, and stored. [5]

3. Results and Discussions

3.1 Short-term Experiments

In the short-term experiment, a 90-day leaching test was conducted at 20 ℃. The results from the concrete waste showed that the average particle diameter increased from 100 nm to 400 nm between Day 0 and Day 90, as shown in **Figure 3**. On Day 90, the PDI value in **Table 3** was very high at 1, indicating low measurement reliability. This is suspected to be due to low concentration. To address this, additional measurements were taken after concentrating the Day 90 sample fivefold. After concentration, the PDI value decreased from 1 to 0.21, which falls within the generally accepted reliable range of 0.1 to 0.3. [5]

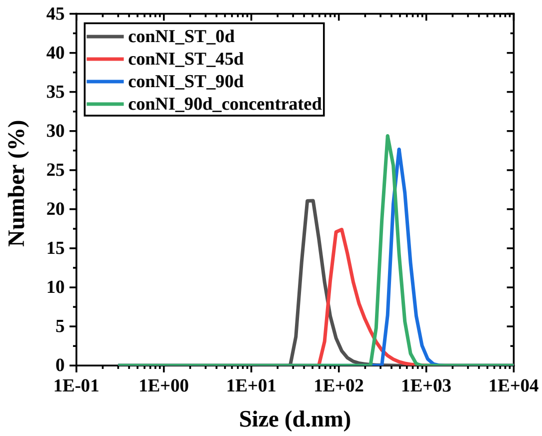

Figure 3. Number distribution of generated colloids in the short-term concrete experiment

The average zeta potential changed from -7 mV to -3 mV, indicating a decreasing trend in surface charge stability as shown in **Table 3**. Additionally, as Day 90, the Quality Factor value, which indicates the reliability of the zeta potential measurement, significantly decreased to 0.28. Even after concentration, it remained low at 0.40. This is considerably lower than 1, which is generally considered the threshold for reliable data. Since the particle diameter did not remain stable throughout the 90-day period, it is necessary to compare these findings with the results from the accelerated experiment.

Based solely on the short-term experiment results, it was difficult to conclude that the colloids generated from concrete waste were stable over a short period. This is because the particle diameter did not show any tendency to remain constant, the absolute value of the zeta potential was not sufficiently large, and the concentration was too low to ensure measurement reliability. [5]

Table 3. Zeta Potential and Quality Factor in the short-term concrete experiment

	Day 0	Day 45	Day 90	Day 90 (Concentrated)
Zeta Potential (mV)	-7.60	-7.12	-3.03	-2.90
Quality Factor	0.97	1.02	0.28	0.40
PDI	0.39	0.57	1	0.21

3.2 Accelerated Experiments

In the results of the accelerated experiment, a higher concentration of colloids was generated compared to the short-term experiment, increasing the reliability of the measurements. In the accelerated experiment, colloids were observed to maintain a consistent particle size of approximately 400 nm from Day 45 to Day 90. This result aligned with the findings from the short-term experiment, confirming that colloids originating from concrete waste were likely to remain stable at a diameter of around 400 nm. However, since the particle size distribution on Day 45 and Day 90 was 200 - 800 nm, it cannot be ruled out that the particles are slowly aggregating. [5]

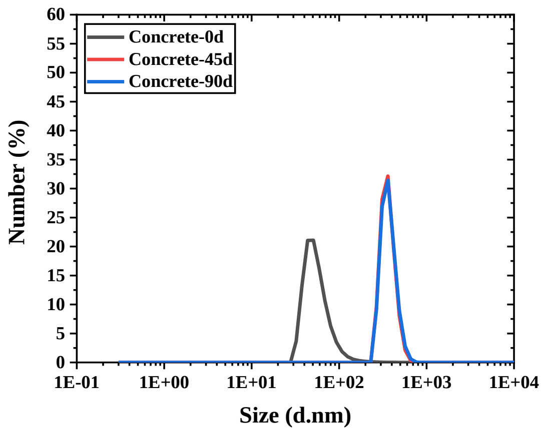

Figure 4. Number distribution of generated colloids in the accelerated concrete experiment

The analysis of the zeta potential results showed that the absolute value of the zeta potential of colloids generated from concrete waste drastically decreased to -0.64 mV over 90 days. Additionally, while a Quality Factor of 1.08 was obtained by Day 45, indicating reliable measurements, by Day 90, the Quality Factor had significantly dropped to 0.20, similar to the short-term experiment. The most common reason for a low Quality Factor was irregular particle size due to particle aggregation. Therefore, it was difficult to conclude that the colloids generated in the concrete experiment remained stable over time. [5]

Table 4. Zeta Potential and Quality Factor in the accelerated concrete experiment

	Day 0	Day 45	Day 90
Zeta Potential (mV)	-7.60	-8.53	-0.64
Quality Factor	0.97	1.08	0.20
PDI	0.39	0.16	0.10

4. Conclusions

This was the first study for understanding the generated colloids from particulate-containing concrete waste. The generation of colloids and its stability from particulate-containing concrete waste was evaluated by simulating the environment of the Gyeongju LILW disposal site. The experiment was conducted through short-term experiment (25 °C, 0-90 days) and an accelerated experiment (60 °C, 0-90 days).

In the short-term experiment, 400 nm colloids were predominantly observed on Day 90. In the accelerated experiment, 400 nm particles were already dominant from Day 45 and remained stable in size until Day 90.

However, the zeta potential analysis showed a decreasing trend in both conditions as the experiment progressed, with absolute values of -3 mV in the short-term experiment and -0.64 mV in the accelerated experiment, indicating relatively low surface charge stability. Additionally, the Quality Factor, which represents the reliability of the zeta potential measurement, was very low (around 0.2–0.4 in both conditions), suggesting the aggregation of unstable particles. This implied that while unstable colloidal particles appear within a certain size range, they may be slowly aggregating and settling over time. Further analysis of the colloidal particle concentration would provide more definitive evidence.

Based on the results of this study, particulate-containing concrete waste in the LILW disposal site environment in Gyeongju was expected to generate 400 nm colloidal particles without additional cementation. However, due to their low stability, the colloidal particle concentration was likely to decrease over time.

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