Radionuclides Leaching Characteristics in Different Sized Geopolymer Waste Forms with Simulated Spent Ion-exchange Resin

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1. Introduction

Radioactive liquid waste generated during the operation of nuclear facilities and decontamination processes has to remove the radionuclides before being reused or released into the environment. Purification of radioactive liquid waste using ion-exchange resin (IER) is commonly used [1]. However, the spent IER is contaminated by radionuclides such as Cs-134/137, Co-58/60, Mn-54, Fe-59, and Sr-90 [2], and it is known that a total of 13,799 drums (200 liter) of waste IERs have been generated in Korean nuclear power plants as of September 2021 [3]. Hundreds of drums are additionally generated every year in nuclear industries, but they are simply stored inside plants due to no proper treatment methods [2].

According to Korean standards, spent IERs should be solidified and proved for the safety of mechanical and chemical stabilities. However, it is difficult to solidify the spent IER due to its characteristics, and cement which is a commonly used solidifying material has several disadvantages such as crack, swelling, low leaching resistance, and low waste loading (about 10 wt.%) [4].

The alternative solidifying method, geopolymer can be hardened by the reaction of amorphous aluminosilicate materials (e.g., fly ash and metakaolin) and an alkali activator (e.g., NaOH and KOH). The geopolymer has good properties such as early-age compressive strength, acidic resistance, thermal stability, and capacity to immobilize radionuclides compared to cement. Recently, studies of solidifying spent IERs using geopolymers have been conducted and tested for the waste form acceptance criteria [5, 6].

The leaching characteristics of radionuclides such as Cs, Co, and Sr are very important for the long-term safety evaluation of a repository. Considering the leachable mass of radionuclides from the solidification material and the size of the waste form are expected to correlate and affect the leaching characteristics. However, no studies were carried out on the leaching characteristics of the waste form with different sizes of waste forms. In this study, the simulated spent IERs were solidified using geopolymer with several different sizes, and the leaching test was conducted according to ANS-16.1-2019 for comparison the leaching results of the different sized waste forms.

2. Materials and Methods

2.1 Materials

For geopolymer synthesis, metakaolin (MetaMax®, BASF) was used as an aluminosilicate material, and potassium silicate solution (PS-2228, R.S CHEM) was used as an alkaline activator. The geopolymer was synthesized with a solid-to-liquid ratio of 1.0 for efficient solidification of simulated spent IERs.

For making the simulated spent IERs, pure IER (AmberLite's IRN-150) was stirred for 1 week in a simulated waste solution dissolved with Cs, Fe, Cr, Mn, Ni, Co, and Sr in concentrations of 300, 6500, 5000, 5000, 5000, 500, and 700 mg/L, respectively, and then dried for 1 week at room temperature. The amount of nuclide adsorbed to the simulated spent IERs are shown in Table 1.

Table I: Adsorbed radionuclide to simulated spent IERs

Nuclide	Cs	Fe	Cr	Mn	Ni	Co	Sr
Adsorbed radionuclides per gram of IER (mg/g)	0.44	8.03	6.22	4.21	4.66	0.48	0.9

2.2 Experimental Methods

For comparison by different sizes and loadings, geopolymer waste forms were prepared with loading of 10 wt.% and 15 wt.%, and sizes of 2.1 cm × 4 cm, 2.9 cm × 5.8 cm, 5 cm × 10 cm, and 15 cm × 30 cm. Waste form with a size of 15 cm × 30 cm was prepared only with waste loading of 15wt.% to investigate the leachability and compressive strength.

Concentration of each radionuclide in the leaching solution was measured by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). The leaching test was performed with reference to ANS-16.1-2019. ANS-16.1-2019 requires leaching data to be obtained through sampling at least 4 times with an interval of 24 ± 0.5 hours. The leachability index (L_i) can be calculated using the formula below:

$$L_i = log\left(\frac{\beta}{D_{e,i}}\right) \quad (1)$$

where, L_i is the leachability index of radionuclide i, β is a defined constant as 1.0 cm^2/s, and $D_{e,i}$ is the effective diffusivity of radionuclide i calculated from the test data (cm^2/s).

In this study, leaching sampling was carried out at 24-hour intervals for 5 days.

3. Results

The compressive strength of the geopolymer waste form containing the highest spent IER loading (15 wt.%) was 10 MPa in both 2.9 cm × 5.8 cm and 15 cm × 30 cm specimens.

As shown in Figs. 1 and 2, the L_i values exceeded 9 in all sizes, irrespective of the loadings and type of radionuclides. In addition, most L_i values increased as the size of the specimen increased, regardless of the loading or type of radionuclides.

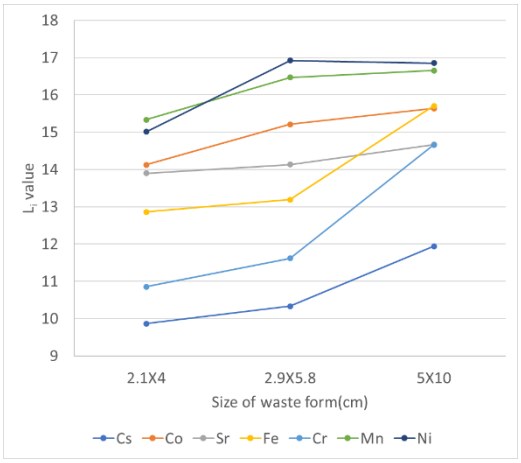

Fig. 1. L_i values of geopolymer loaded 10 wt.% of simulated spent IERs.

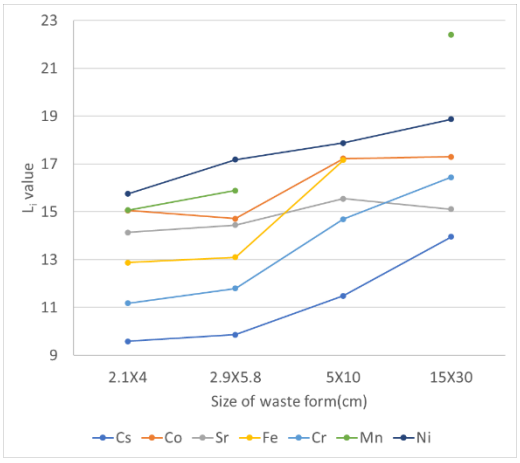

Fig. 2. L_i values of geopolymer loaded 15 wt.% of simulated spent IERs (Mn and Fe was not detected in the leachate of 5 cm × 10 cm specimen and 15 cm × 30 cm specimen, respectively).

4. Conclusion

The compressive strength of both 2.9 cm × 5.8 cm and 15 cm × 30 cm waste forms was 10 MPa, and the mechanical strength of the geopolymer waste form loaded by the IER wastes was not affected by the size.

As a result of the leaching test, the L_i value exceeded 9 for all radionuclides even in different sized waste forms, which satisfies the waste acceptance criteria (> 6.0). Overall, for all radionuclides, the L_i value increased as the size of the geopolymer waste form increased. The radionuclide with the lowest L_i value was cesium, and the lowest value was measured to be 9.6 in 2.1 cm × 4 cm waste form loaded with 15 wt.% of simulated spent IERs.

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