Long-term Leaching Test of Cs Pollucite Waste Form for Radioactive Cs immobilization

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1. Introduction

Radioactive cesium in spent nuclear fuel is high heat generated and semi-volatile nuclides. It is occurred as the gas phase in the pretreatment process of pyroprocessing. [1,2] A one of the methods of capturing gaseous radioactive cesium is inorganic ion exchange using zeolite.[3] In this study, we are fabricated the Cs pollucite ceramic waste form by calcining at a high temperature for high thermal and chemical stability and evaluated the chemical stability through long-term leaching test using monolithic samples.

2. Experimental

After fabricated Cs pollucite powder using a raw material, the optimal condition for forming Cs pollucite ceramic waste form was found. The monolithic sample prepared using pollucite powder of the optimal condition is performing a leaching test for up to 28 days.

2.1 Fabricated Cs pollucite powder

As the precursors of the matrix material, mullite and SiO_2 mixture, denoted the Adsorbent product (AP), obtained by heat treatment of kaolinite were mixed with Cs_2CO_3, which is a precursor of Cs_2O, as mass ratio of Cs_2O/AP = 0.2, 0.3, 0.4, 0.5, 0.6 and 0.8. the mixture of precursors was calcined at 1000 and 1200°C for each 2 hr to prepare Cs pollucite. The sample was denoted P-1200-(mass ratio of precursor).

2.2 Palletization of pollucite ceramic waste form

A pellet-type waste form was fabricated by cold pressing using the produced Cs pollucite powder of Cs_2O/AP = 0.2. The optimal condition was to apply pressure at a pressure of 50 bar for 1 hour when the diameter of the pellet was 25 mm.

2.2 Characterization

Cs pollucite powder was confirmed the formation of a crystal phase by X-ray diffraction (XRD) and the volatilization of Cs by Thermogravimetric analysis (TGA). Morphology was analyzed field emission scanning electron microscope (FE-SEM) with energy dispersion.

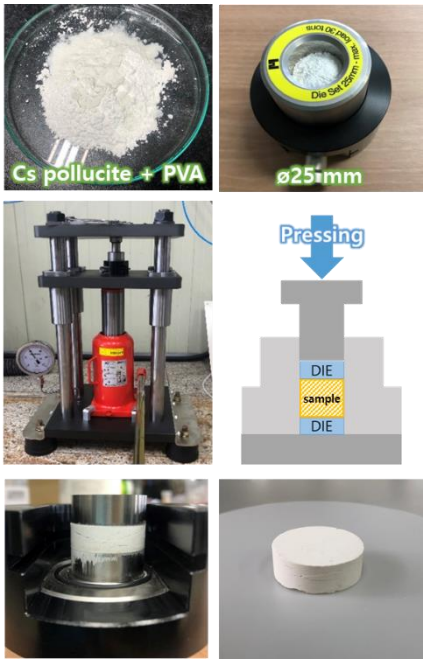

Fig. 1. The process of cold pressing and fabricated pollucite pellet before sintering

2.3 long-term leaching test

long-term leaching test was performed using Cs pollucite waste form prepared under optimal conditions by MCC-1 method. The sample size of leaching test was 10×10×5 mm. The test was conducted allard groundwater (GW) and Deionized Water (DI) as leachate at room temperature (RT) and 60°C (60), respectively. The leachate replacement interval was 5 contact period, which is 3 hours, 3 days, 7 days, 14 days, and 28 days.

3. Results and Discussion

3.1 General Properties

It was shown that there are pollucite phase at all samples of P-1200 as shown in Fig. 1. It was confirmed that the release of Cs was low by TGA and EDS. Thus, pellets were fabricated using the sample including only pollucite phase. However, all samples were not fabricated well with pellets. The pellet was prepared using P-1200-0.2 with a high matrix ratio owing to break sample in layers when the Cs-loading is high.

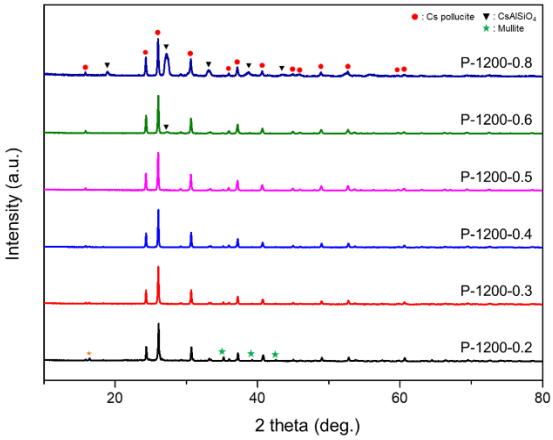

Fig. 2. The XRD pattern of fabricated Cs pollucite powder

3.2 Chemical Stability

It is estimated that the long-term leaching rate of the fabricated waste form will follow the known leaching trend. There are two stages, a rapid leaching occurs in the early stage, and the second stage will decrease due to ion equilibrium between the leachate and samples. In Fig.3 is the variation of mass fractions of elements over time. The leached ion fraction (f) up to the 28th is decreased as shown in Fig. 4.

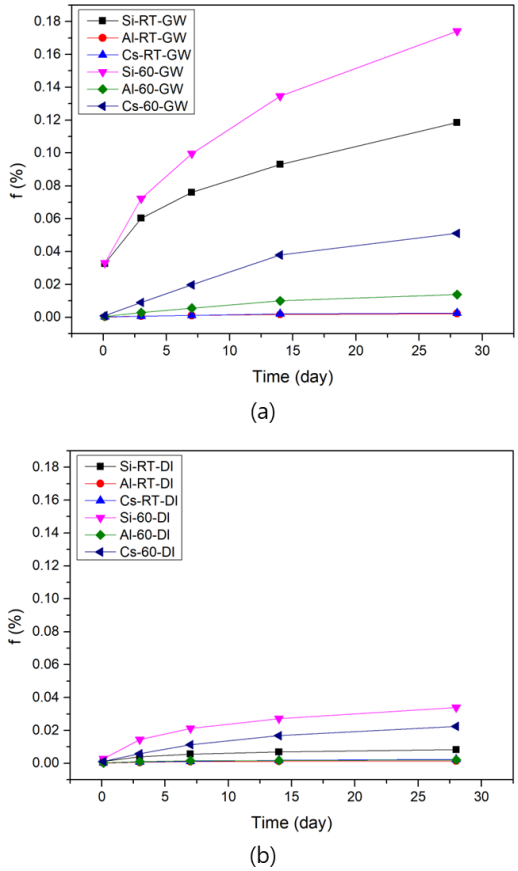

Fig.3. Time-dependent leached element fractions (f) of Cs, Al and Si for 28 days under each leachate (GW and DI) at RT and 60℃

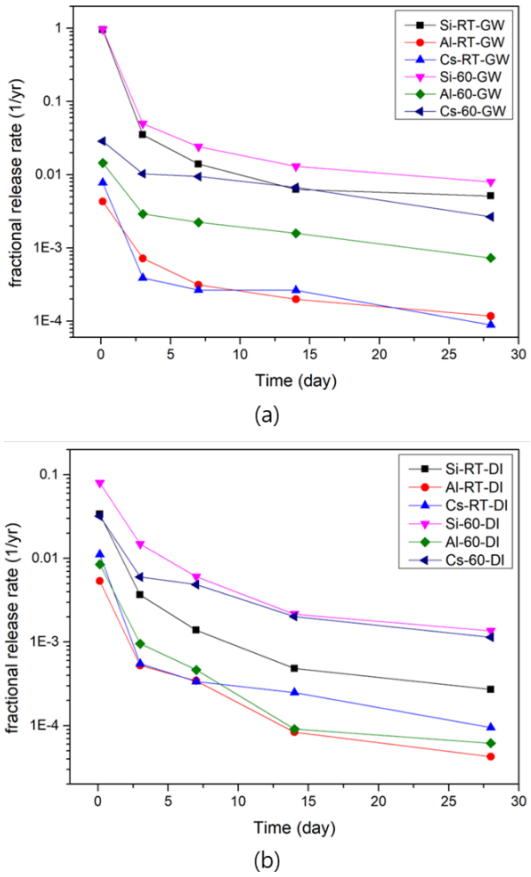

Fig.4. Time-dependent fractional release rate of Cs, Al and Si for 28 days under each leachate (GW and DI) at RT and 60

4. Conclusion

For the stable immobilization of radioactive cesium, Cs pollucite was fabricated and pelletized by the optimal condition. The chemical stability was evaluated several conditions by the fabricated monolithic sample which was well formed. As a result, the mass fraction of element gradually increased and the leaching rate tend to decrease with time.

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