

Glass-Ceramic Waste Forms for Immobilization of Cs Trapping Filters

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1. Introduction

Cesium gas is evolved from spent nuclear fuel during the head-end process of Pyroprocess. Capture of Cs has been performed with aluminosilicate-based ceramic filters at high temperatures [1]. This study presents a simple and novel scheme to immobilize the Cs trapping filters in glass-ceramic waste forms.

2. Materials and methods

2.1 Cs trapping experiment using the SA filter

The silica-alumina (SA) filter composed with aluminosilicates was prepared with kaolinite powder. The synthesis of SA filter was described elsewhere [2]. Typically, a polyvinyl alcohol (PVA) solution and the kaolinite powder were mixed to make a slurry. Then, a polyurethane (PU) sponge was coated with the slurry. The slurry-filled sponge was sufficiently dried and the coating process was repeated several times. The sponge was thermally elevated at 1200°C to remove the PVA and the PU sponge.

Cesium hydroxide (CsOH·H₂O) placed in a boat crucible and the SA filters were loaded in an alumina tube. The filters were thermally treated at 800°C and then cesium hydroxide was volatilized at 900°C in flowing 4% H₂/Ar gas.

2.2 Synthesis and characterization of waste forms

A glass frit was prepared and the compositions were presented in Table 1. The glass frit and the Cs-trapping filter (Cs-SA filter) were mixed and heat-treated at 1000°C in the air. Characterization of the specimens was performed by X-ray diffraction (XRD) and scanning electron microscope (SEM) equipped

with energy dispersive spectra (EDS). Leaching of the specimens was examined by MCC-1 test at 90 °C.

Table 1. Compositions of the glass frit prepared in this research

Oxide	Reagent	Mass fraction (wt%)
SiO ₂	SiO ₂	47.4
B ₂ O ₃	B ₂ O ₃	30.1
Na ₂ O	Na ₂ CO ₃	22.5

3. Results & discussion

Fig. 1 shows the XRD patterns of the Cs-SA filters after heat-treatment with different temperatures for 3 h. It is seen that the structure of CsAlSiO₄ in the Cs-SA filter is present during the heat treatment at 800°C. CsAlSiO₄ peaks are decreased and pollucite peaks are developed from 900°C. In particular, CsAlSiO₄ peaks are significantly lowered and the intensity of pollucite peaks becomes very prominent at 1200°C. It suggests that the formation of pollucite is possible by the simple heat process of the Cs-SA filter.

The XRD patterns of the waste forms are presented in Fig. 2. Peaks were not found at 10–20 wt% of waste fraction, suggesting that a glassy phase was formed during the thermal elevation. However, peaks appear at 30–50wt% of waste fraction, which were all identified as pollucite. In addition, the intensity of pollucite peaks became higher as waste fraction grows, suggesting that the growth of waste fraction led to the increase of the crystallinity of pollucite.

Fig. 3 shows morphology of the waste form surfaces investigated by SEM. It is seen that there are many round particles with diameters of 100–600 nm (Fig. 3(a)). The irregular and agglomerated particles were also observed (Fig. 3(b)). EDS analysis reveals

that the particles are pollucite.

Leaching rates of the specimens were presented with respect to major elements at 3, 7, 14, and 28 d of duration (Fig. 4). The leaching rates of most elements are gradually declined with the increase in the leaching period. In addition, the increase in the waste fraction generally lowers the leaching rates of all elements. In particular, the Cs leaching rate at 3 d was $8.3 \times 10^{-3} \text{ g}\cdot\text{m}^{-2}/\text{d}$ for the 10wt%-waste specimen (Fig. 4(a)), which was two orders of magnitude higher than that for the 50wt%-waste specimen ($7.21 \times 10^{-5} \text{ g}\cdot\text{m}^{-2}/\text{d}$). This result originated from the difference of main Cs-incorporated phases in the waste forms. Most Cs was contained in an amorphous Cs-aluminosilicate phase at low waste fraction; however, the major Cs phase became pollucite at higher waste fractions.

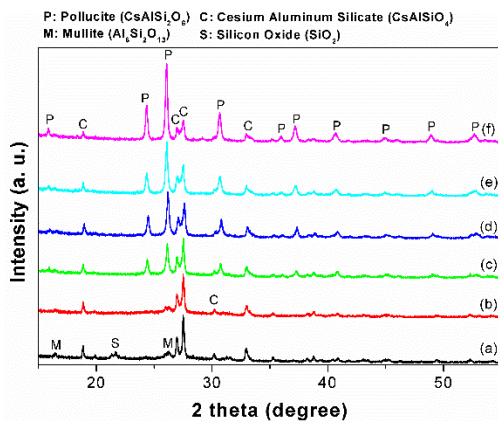


Fig. 1. XRD patterns of the Cs-SA filters after heat treatment for 3 h at different temperatures: (a) no heat treatment, (b) 800°C, (c) 900°C, (d) 1000°C, (e) 1100°C, and (f) 1200°C.

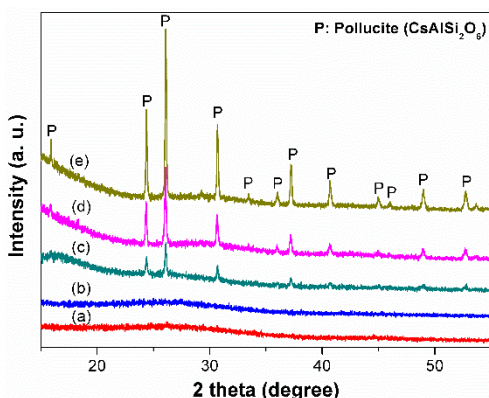


Fig. 2. XRD patterns of the monolithic waste forms at different waste fractions: (a) 10wt%, (b) 20wt%, (c) 30wt%, (d) 40wt%, and (e) 50wt%.

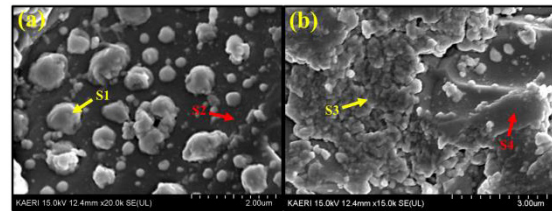


Fig. 3. SEM images of the specimen at 40wt% of waste fraction with different magnifications ((a) 20,000 times and (b) 15,000 times).

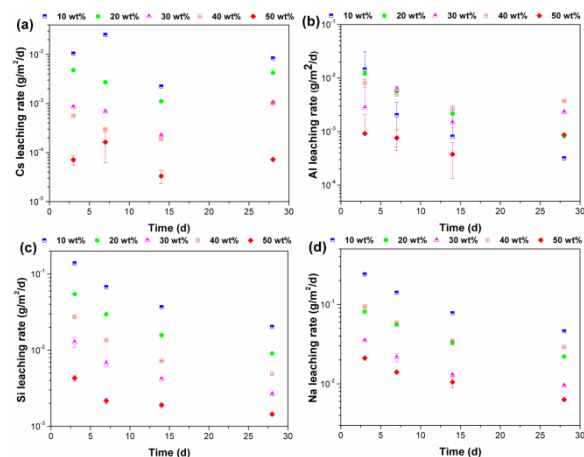


Fig. 4. Normalized leaching rates of (a) Cs, (b) Al, (c) Si, and (d) Na for different specimens obtained from an MCC-1 leaching test performed for 28 d.

4. CONCLUSIONS

The Cs-SA filter was transformed to monolithic waste forms for the final storage of Cs. A glass frit was synthesized and mixed with the Cs-SA filter, which was heat-treated at 1000°C for 3 h to for the synthesis of waste forms. Characterization revealed that pollucite was the major Cs phase when the waste loading was not less than 30wt%.

REFERENCES

[1] Lee, H.S.; Park, G.I.; Kang, K.-H.; Hur, J.-M.; Kim, J.-G.; Ahn, D.-H.; Cho, Y.-Z.; Kim, E.H. (2011) Pyroprocessing technology development at KAERI. Nucl. Eng. Technol., 43 (4): 317.
[2] Yang, J.H.; Yoon, J.Y.; Lee, J.H.; Cho, Y.-Z. (2017) A kaolinite-based filter to capture gaseous cesium compounds in off-gas released during the pyroprocessing head-end process. Ann. Nucl. Energy, 103:29.