Direct Radioactive Waste Treatment with Amorphous Aluminosilicate Sorbents

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1. Introduction

During operating nuclear power plants, radioactive liquid wastes are generated because of activated corrosion products [1]. Radioactive nuclides must be removed and there are several techniques such as ion exchange, filtration, precipitation, membrane separation and adsorption [2]. Among them, adsorption is an easy and economical way to treat liquid radioactive wastes.

After removing radioactive nuclides, the used material is also one of the radioactive solid wastes. For disposal, it is required to be immobilized and consolidated because leaching happens during long-term storage. There are several techniques for immobilization such as vitrification, cementation and consolidation using ceramics [3–7]. However, they require high-temperature processing (> 600 °C) which induces volatile ions to evaporate, longer time (hours to days), and additional materials as a binder which brought low loading of radioactive wastes.

However, the cold sintering technique is outstanding for applying to immobilization of radioactive wastes because it operates under 300 °C within a few minutes [8,9]. Thus, the technique is an energy-saving (temperature, time and material) and safe way. Moreover, the relative density obtained by cold sintering was reported as higher than that by conventional sintering methods.

In this study, we show that the cold sintering technique is applied to direct immobilization with as-spent amorphous aluminosilicates. Furthermore, we analyzed whether the cold sintered matrix is suitable for the use as a resistant matrix by standard leaching tests.

2. Methods/Experimental

2.1. Batch Adsorption Test

Synthesized amorphous aluminosilicate powder used for batch adsorption tests with nuclides (Cs^+, Co^{2+}, Ni^{2+}) followed by OECD guidelines. Both kinetics and isotherms experiments were performed at specific times (t = 0, 5, 15, 30, 60, 120, 1440 min) with 0.2 g for 100 mL of specific concentrations (C_i = 12.5, 25, 50, 75, 100, 150 ppm). The used materials were separated using a centrifuge at 3000 rpm for 5 min and a PTFE syringe filter of 0.45 ㎛. Next, the separated adsorbents were dried at 90 °C for 24 h.

2.2. Direct Cold-Immobilization and Leaching Tests

The as-spent dried adsorbents were consolidated by cold sintering at 200 °C under 500MPa for 10 min in an open environment.

The chemical durability and leaching resistance were performed by following ASTM C1285 (7-days PCT) and ANSI/ANS 16.1 (90-days leaching test) [10,11].

2.3. Characterization

The crystal structures of pre- and post-cold sintered adsorbents were analyzed by X-ray diffractometry (XRD, RIGAKU). The adsorption capacity of amorphous aluminosilicates was calculated with ion concentrations measured by the inductively coupled plasma optical emission spectroscopy (Agilent ICP-OES 720).

The microstructure of pre and post-cold sintered adsorbents and post-leaching matrix was analyzed by the scanning electron microscopy (SEM) and the transmission electron microscopy (TEM).

The chemical durability of cold sintered amorphous aluminosilicates adsorbing nuclides was investigated under static leaching conditions. The ASTM standard C1285 and ANSI/ANS 16.1 were used for measuring the resistance of the cold sintered matrix.

3. Results and Discussion

3.1. Synthesized Amorphous Aluminosilicate

Amorphous aluminosilicate was synthesized by the co-precipitation method [12]. For analyzing the phase of aluminosilicate, XRD was done as shown in Fig. 1. The result indicated that synthesized aluminosilicate was fully amorphous and there was no dominant identified peak.

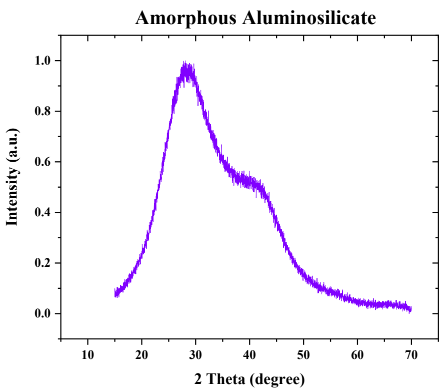

Fig. 1 The XRD pattern of synthesized amorphous aluminosilicate

3.2. Kinetics and Isotherms of Amorphous Aluminosilicate

For interpreting the mechanism of adsorption, we used the theoretical models such as pseudo-second order and Langmuir to fit the measured ion concentration data by ICP-OES as shown in Fig. 2. These results suggested that the chemisorption process was dominated and the adsorption site on amorphous aluminosilicates was a monolayer.

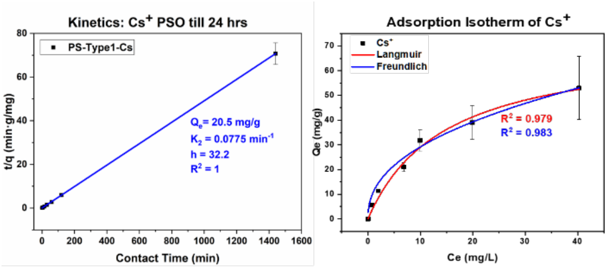

Fig. 2 (Left) Kinetics and (Right) isotherms of amorphous aluminosilicate

3.2. Cold Sintered Amorphous Aluminosilicate

Cold sintered amorphous aluminosilicate reached full densification of 2 g/cm^3 and the microstructure with free of pores was observed by TEM as shown in Fig. 3. Before cold sintering, there were pores and scratches in the synthesized particles, but after cold sintering, the matrix was densified without any scratches and pores. Thus, the cold sintering technique provided a well-densified matrix.

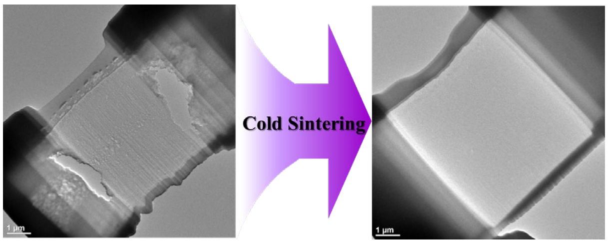

Fig. 3 TEM images (low magnification) of pre and post-cold sintered amorphous aluminosilicate.

3.3. Resistance of the cold sintered matrix (ASTM C1285 and ANSI/ANS 16.1)

For measuring the chemical stability and leaching resistance of the cold sintered waste matrix, ASTM C1285-14 and ANSI/ANS16.1 were done.

Compared with other materials which were immobilized by conventional sintering such as vitrification and hot isostatic pressing (HIP), cold sintered amorphous aluminosilicate showed not only the highest chemical durability but also the long-term leaching resistance as shown in Fig. 4. These results indicated that the cold sintered matrix can be one of the promising immobilized systems.

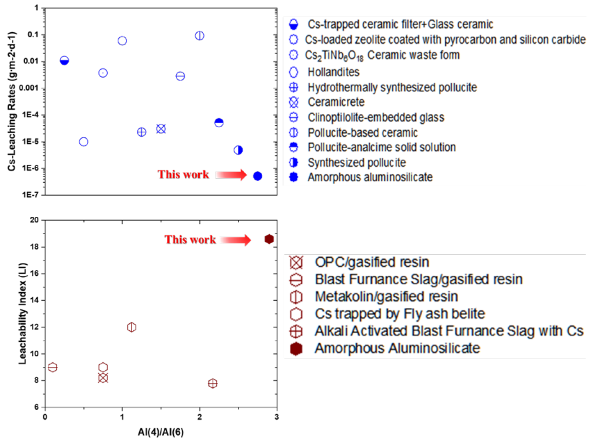

Fig. 4 (Top) PCT results and (Bottom) ANSI results

4. Conclusions

The present study demonstrated a one-through treatment of radioactive nuclides from adsorption to immobilization by cold sintering without any binders. Moreover, the cold sintered matrix resulted in the highest chemical durability and leaching resistance. Thus, this concept may be considered as enhancing the safety and economical treatment of radioactive wastes.

Acknowledgments

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