



Cesium extraction from simulated high-level vitrified wastes

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ABSTRACT

A phase separation technique was applied to a simulated high-level vitrified waste, which consisted of non-radioactive elements, to determine the efficient extraction of cesium from the vitrified material. Experimentally, the simulated vitrified material was melted at 900–1000 °C and subsequently annealed at 700 °C under the reductive atmosphere. The sodium-potassium-rich materials were found to be phase-separated on the glass surface that was in contact with the gas phase, which enhanced the leachability of cesium in the glass. Such phase separation was not observed under other atmospheres (a CO₂-rich or air atmosphere), thus elucidating the effect of the reductive atmosphere. The glass was converted to particles with higher surface area through a dropping test of the molten glass in the melting stage, and the sodium-potassium-rich materials, subsequently, were more effectively phase-separated on the surfaces. During water leaching, 70–90% of the total cesium was extracted from the particles.

1. Introduction

Borosilicate glass has been widely used as a matrix for vitrifying high-level liquid wastes (HLLWs) generated from reprocessing facilities of spent fuels in nuclear power plants. Because of its glass forming ability, chemical durability, thermal stability, and ability to incorporate several different elements (Donald, 2007; Boucetta et al., 2012; McCloy et al., 2012), vitrification of HLLWs into such a durable borosilicate glass has been proven to effectively prevent leakage of radioactive nuclides in HLLWs into the environment during their storage (Lutze and Ewing, 1988). However, these nuclides contain long-lived fission products (LLFPs) with long half-lives (Takibayev et al., 2005) that require long-term storage of the high-level vitrified wastes (HLVWs) after final disposal. In addition, disposal sites for the HLVWs are limited in Japan. From a management and security point of view, a new treatment for the HLVWs is imperative for the future. To treat the existing HLVWs, one possible option is to separate LLFPs from the HLVWs and transform them into short-lived or stable nuclides using nuclear transmutation techniques (Takibayev et al., 2005; Cabinet Office, 2016; Inoue et al., 1991). Among the LLFPs, rare metals are present and these can be recycled if they are transmuted to non-radioactive rare metals (Koyama et al., 2010; Ozawa et al., 2011). Therefore, LLFP recovery from the HLVWs would be important for the reduction of the existing HLVWs and resource conservation in future. In previous studies, separation techniques of nuclides from HLLWs (liquid) have been proposed (Fujita and Akai, 1998; Ozawa et al., 2002, 2005; Zhang et al., 2006;

Dakshinamoorthy et al., 2008; Jayakumar et al., 2009; Uozumi et al., 2011; Ruhela et al., 2012; Alok Rout et al., 2012; Yuezhou et al., 2013; Xu et al., 2013; Nayak et al., 2013; Raut et al., 2013; Kawamura et al., 2013; Chen et al., 2013; Nogami et al., 2015; Jagasia et al., 2016, 2017; Chaudhury et al., 2018), whereas studies of the nuclide separation from HLVWs (solid) are limited (Jensen et al., 1984; Naito et al., 1986; Uruga et al., 2007, 2008; Jena et al., 2011). We focused on an LLFP separation technique from HLVWs under the “Impulsing Paradigm Change through Disruptive Technologies (ImPACT)” Program (Cabinet Office, 2016) in Japan.

In the above-mentioned previous studies for HLVWs (Jensen et al., 1984; Naito et al., 1986; Uruga et al., 2007, 2008; Jena et al., 2011), platinum group metals (PGMs) or molybdenum in LLFPs was extracted in liquid metals such as copper, lead, and tin. In these processes, extraction of cesium, also target LLFPs in the ImPACT program, is difficult, because it is more stably present in the oxide phase than PGMs. Among the LLFPs, the radioactive cesium release radiation at higher doses compared to other radioactive nuclides (Ministry of Education), and it should be also separated from the glass and transmuted to short-lived or stable nuclides together with PGMs. Our target nuclides among the LLFPs were cesium and PGMs, for which we have developed a phase separation technique to enhance cesium leachability from glass materials for efficient cesium extraction (Xu et al., 2016, 2017). In this process, heavy-metal-containing borosilicate glass was melted at 1000 °C under a continuous CO-containing reductive atmosphere generated by the combustion of carbon. During melting, the heavy metal

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oxides in the molten glass were reduced to metallic states by CO and PGMs were extracted in the generated liquid metal. The molten oxide was subsequently annealed at 700 °C and water-soluble sodium-potassium-rich (Na-K-rich) materials were phase-separated on the surface of the treated glass that was in contact with the gas phase (Okada et al., 2015a) (referred to as the “contact surface”). Simultaneously, cesium was transferred to the Na-K-rich materials. Therefore, cesium was effectively extracted in water along with the Na-K-rich materials from the contact surface. As a proof of concept, we applied this phase separation technique to a cesium-containing simulated glass in the SiO₂-B₂O₃-Na₂O-K₂O-Bi₂O₃-CaO system and reported that cesium extraction was promoted by the technique (Xu et al., 2017). If the HLWVs are converted to such heavy-metal-containing glass, cesium and PGMs can possibly be recovered separately using this process. However, as noted in this study, in the practical HLWVs, a higher variety of chemical components are present compared with the above-mentioned simulated glass we used previously. The difference in the chemical composition affects glass viscosity and the mobility of ions in the glass, and it has not been confirmed if the phase separation of the Na-K-rich materials occurs in the HLWVs. To verify the practical application of this phase separation technique, a glass material whose chemical composition is similar to the practical HLWVs should be examined.

To investigate the applicability, we focused on the following three issues. The first is the demonstration that phase separation of the Na-K-rich materials from this type of glass is promoted by CO. Secondly, factors determining the phase separation efficiency in the glass should be clarified. The reaction between CO and the glass may affect phase separation behaviors, and the reactivity depends on the melting temperature and the partial pressure of CO. The effects of these parameters on the phase separation efficiency were not investigated in our previous work (Xu et al., 2016, 2017). Third, it is necessary to develop a technique that produces a heat-treated product with a higher surface area to enable more efficient cesium extraction. This is because the surface area of the product that was in contact with water significantly affected the extraction efficiency of cesium in the leaching process. By dropping the molten glass to produce a granular product with a high surface area, a larger quantity of the Na-K-rich materials can be phase-separated on the contact surface and cesium extraction can be promoted. To the best of our knowledge, the dropping test of molten HLWVs for the preparation of the granular product has not been studied.

In this study, phase separation by a reductive heat treatment was applied to a simulated HLWV whose chemical composition was similar to the practical HLWVs used for cesium extraction. Our purpose is to investigate the following issues: (i) phase separation of the Na-K-rich materials from the simulated HLWV under a CO-containing atmosphere, (ii) melting conditions that enhance the phase separation efficiency in the simulated HLWV, and (iii) cesium extraction efficiency from the granular heat-treated product with increased surface area. The simulated HLWV was heat-treated under different conditions in a batch experiment. A dropping test of the molten oxides was designed for the first time in this study and performed to convert the oxide into particles with higher surface area. Leaching tests were also performed on the resulting products, and the cesium extraction efficiencies were evaluated.

2. Materials and methods

2.1. Simulated HLWV

The simulated HLWVs used in this study were provided by IHI Co. Ltd. (Tokyo, Japan). The practical HLWVs consist of glass forming reagents (SiO₂, B₂O₃, Na₂O, Al₂O₃, ZnO, CaO, and Li₂O) and other materials containing radioactive nuclides. Based on the characteristics, the simulated HLWVs were designed using nonradioactive chemical compounds so that their chemical compositions were similar to those of the above-mentioned practical HLWVs. Table 1 shows the chemical

Table 1
Chemical composition of the simulated HLWV.

| | Concentration (wt.%) |
|---------------------------------|----------------------|
| SeO ₂ | 0.02 |
| Rb ₂ O | 0.11 |
| SrO | 0.27 |
| Y ₂ O ₃ | 0.17 |
| ZrO ₂ | 1.31 |
| MoO ₃ | 1.36 |
| MnO | 0.34 |
| RuO ₂ | 0.7 |
| Rh ₂ O ₃ | 0.12 |
| PdO | 0.37 |
| Ag ₂ O | 0.02 |
| SnO ₂ | 0.03 |
| TeO ₂ | 0.13 |
| Cs ₂ O | 0.71 |
| BaO | 0.54 |
| La ₂ O ₃ | 0.33 |
| CeO ₂ | 1.94 |
| Pr ₆ O ₁₁ | 0.35 |
| Nd ₂ O ₃ | 1.09 |
| Sm ₂ O ₃ | 0.21 |
| Eu ₂ O ₃ | 0.03 |
| Gd ₂ O ₃ | 1.24 |
| P ₂ O ₅ | 0.03 |
| Cr ₂ O ₃ | 0.01 |
| Fe ₂ O ₃ | 0.21 |
| NiO | 0.02 |
| Na ₂ O | 9.24 |
| SiO ₂ | 50.5 |
| B ₂ O ₃ | 13.6 |
| CaO | 3.08 |
| Al ₂ O ₃ | 5.47 |
| ZnO | 3.21 |
| Li ₂ O | 3.06 |

composition of the simulated HLWVs. This information is also provided by IHI Corporation. The cesium concentration of the simulated HLWVs was 0.71 wt%. The corresponding is 45 GWD/t in a pressurized water reactor.

2.2. Batch heat-treatment experiment and leaching test

The heat-treatment experiment was performed following the methodology presented in our previous studies (Xu et al., 2016, 2017; Okada et al., 2015a, 2015b; Okada and Yonezawa, 2013, 2014). A 30 mL alumina crucible with a mixture of the simulated HLWVs, Na₂CO₃, Bi₂O₃, and K₂CO₃ was placed into a 100 mL alumina crucible. Activated carbon or CaCO₃ was added to the 100 mL crucible to perform the heat treatment under different atmospheres. When the activated carbon was added, CO gas was generated by the carbon combustion in the reaction container (CO-rich atmosphere). The generation of CO varied according to the different carbon dosage. When CaCO₃ was added, CO₂ gas was generated by the decomposition of CaCO₃ (CO₂-rich atmosphere). In the experiment, the heat treatment was also performed under an air atmosphere, and in this case, no additive was put in the 100 mL alumina crucible. The reason for changing atmospheres is explained in Section 3.1. An alumina cover was placed on the 100 mL crucible and this reaction container was placed in an electric furnace. The temperature in the furnace was raised to around 900–1000 °C over 30 min, and the mixture in the 30 mL crucible was melted at this temperature for 1 h. The furnace was then allowed to naturally cool to 700 °C, and the molten oxide was annealed for 2 h. Finally, the furnace was allowed to cool naturally to room temperature. The obtained material is referred to as “annealing product.” The experimental conditions are shown in Table 2.

A water leaching test was performed on the obtained products. The 30 mL alumina crucible with the product was placed directly into a 200 mL beaker containing 150 mL of ion exchange water (electrical

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