Fabrication of Monolithic Wasteform Containing Molten Salt Waste and its Chemical Durability

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1. Introduction

In the management of radioactive wastes, the molten salt waste from a pyroprocess to recover uranium and transuranic elements, which mainly consist of metal chlorides, is one of the problematic wastes not applicable directly to a conventional solidification process due to its physicochemical properties such as its volatility and low comparability with silicate glass**[1]**. The problems of metal chlorides for final disposal are attributed mainly to the Cl-induced physico-chemical properties. Therefore, dechlorination by a reasonable method applicable to the radioactive waste management could remove the limitations on the host matrix and process temperature. As one of dechlorination methods, our research group developed a new reaction system that guarantee the conversion of radionuclides in a salt waste into stable compounds and its solidified product by using glass as a chemical binder showed good leach-resistance. This named GRSS (Gel-Route Stabilization/ Solidification) method for radioactive molten salt wastes**[2]**. Different from the GRSS method using a solution system, our research group developed a solid reaction system; a synthesized inorganic material was reacted with salt waste at 650°C and manageable products for high temperature process could be obtained.

This study investigated the fabrication condition for monolithic wasteform containing molten salt waste and analyzed its physico-chemical properties such as phase distribution, chemical durability and etc.

2. Methods and Results

The inorganic material, SiO_2-Al_2O_3-P_2O_5(**SAP**), was prepared by a sol-gel process. Tetraethyl orthosilicate (TEOS, Aldrich, 98%), aluminum chlorides ($AlCl_3 \cdot 6H_2O$, Junsei, 98%) and phosphoric acid (H_3PO_4, Junsei, 85%) were used as precursors of Si, Al and P, respectively. The molar ratio of Si/Al/P was adjusted to 1/1/1.25. The SAP was mixed with simulated salt waste (90wt% LiCl, 6.8wt% CsCl, 3.2wt% SrCl2) with a mixing ratio, SAP/salt=2, and the mixture was reacted at 650°C for 16hrs. As a chemical binder, borosilicate glass was mixed with the reaction product and heat-treated at 1100~1200°C for 4hrs. The wasteform was characterized by XRD, SEM, TMA analysis. Its chemical durability was tested by PCT-A and ISO method. The borosilicate glass used in this study consist of 7.14wt% Na_2O, 1.6wt% CaO, 22.84wt% B_2O_3, 8.84wt% Al_2O_3 and 58.98wt% SiO_2.

Fig. 1 Photograph of prepared wasteforms at different conditions.

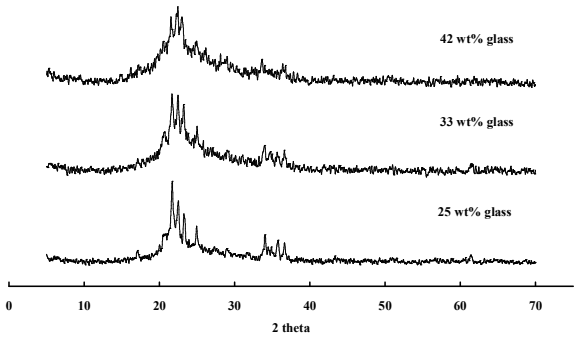

Fig. 2 XRD patterns of wasteforms with different borosilicate glass

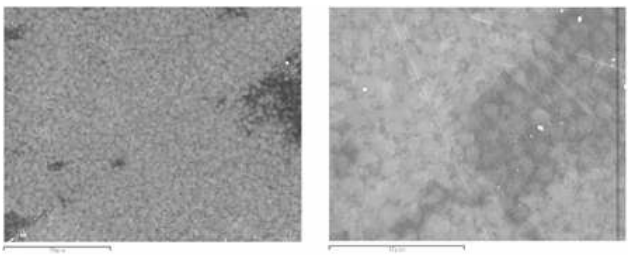

Fig. 3 SEM image of wasteform with 25wt% glass. (scale: 20μm-left, 10μm-right)

As shown in Fig. 1, the main crystalline phase in wasteform is SiO_2 and Li_3PO_4. Under given condition, a monolithic wasteform was successfully fabricated; SEM image shows the highly uniform of wasteform. The density of wasteform is about 2.4g/cm^3 and its thermal expansion coefficient has about 10^{-6}/°C.

The PCT-A test result revealed that the leach rates of Cs and Sr were about 10^{-3}g/m^{2}day. As a dynamic leaching test method, the ISO test is going on.

3. Conclusion

This study reported the fabrication of monolithic wasteform containing molten salt waste and its physico-chemical properties. The wasteform suggested in our research group should be characterized and qualified if it meets the requirements for final disposal. For this purpose, many experiments related with performance of wasteform are going on and planed.

Acknowledgments

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