Immobilization of Rare Earth oxides into Ceramic Wasteform: Synthesis of Some Candidates for Host Matrix

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1. Introduction

The LiCl-KCl electrolytes used in an electro-refining process of pyroprocess contains a fraction of α-emitting radionuclides such as rare earth elements. In KAERI, selective reaction by using oxygen gas has been under investigation for reducing final waste volume. From this separation process, a series of oxides is expected to be generated, which should be immobilized into a reliable matrix for long time. To our knowledge, such wastes containing only rare earth radionuclides have never been treated for final disposal and still under investigation on the possible host matrix for α-emitting radionuclides.

Borosilicate glass or single ceramic materials have been proposed for the immobilization of long-lived radionuclides. In order to guarantee the performance of wasteform, it requires test time as long as half-life of α-emitting radionuclides at least. However, some natural rocks survived weathering, erosion and other environmental changes for millions year. This might answer the question, "what matrix is reliable for α-emitting radionuclides".

Candidates for these radionuclides were reported to be monazite($CePO_4$), apatite($Ca_{4-x}REE_{6+x}(SiO_4)_{6-y}(PO_4)_y(O,F)_2$ and zircon($ZrSiO_4$)**[1,2]**. Different SYNROCK phase that contains perovskite, hollandite, titanium oxide and zirconolite, this study considered monazite and apatite as possible host matrix and tried to find an in-situ synthetic route to the target mineral phase.

2. Methods and Results

In order to synthesize monazite or apatite in solid-solid reaction system, it is necessary oxide-based reagent. In this study, CaO-SiO_2-P_2O_5 (CaPS) material as a base reagent for rare earth oxides was considered. This material was synthesized by a conventional sol-gel process. The molar ratio of Ca/Si/P was adjusted to 0.8/1/1.25. The prepared hydrogel was dried and heat-treated at 600°C for 2hrs. This product (CaPS) was mixed with rare earth oxides (Y, La, Ce and Nd) at different ratios and the mixture was reacted at 600~1100°C for 1 hrs. The reaction products were analyzed by X-ray diffractometer.

Fig. 1 shows the XRD patterns of reaction products for CeO_2. This material contained mainly monazite as crystalline phase. Below 900°C, CeO_2 was not react with CaPS and the reaction start at 900°C.

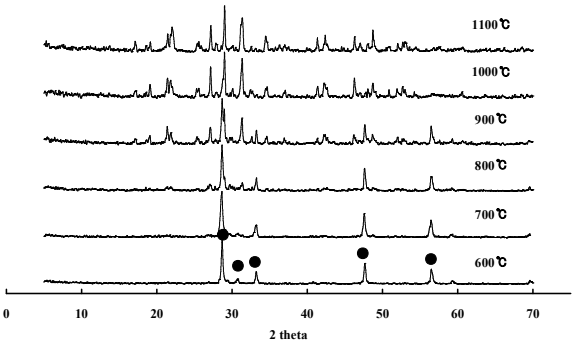

Fig. 1 XRD patterns of reaction products for CeO_2

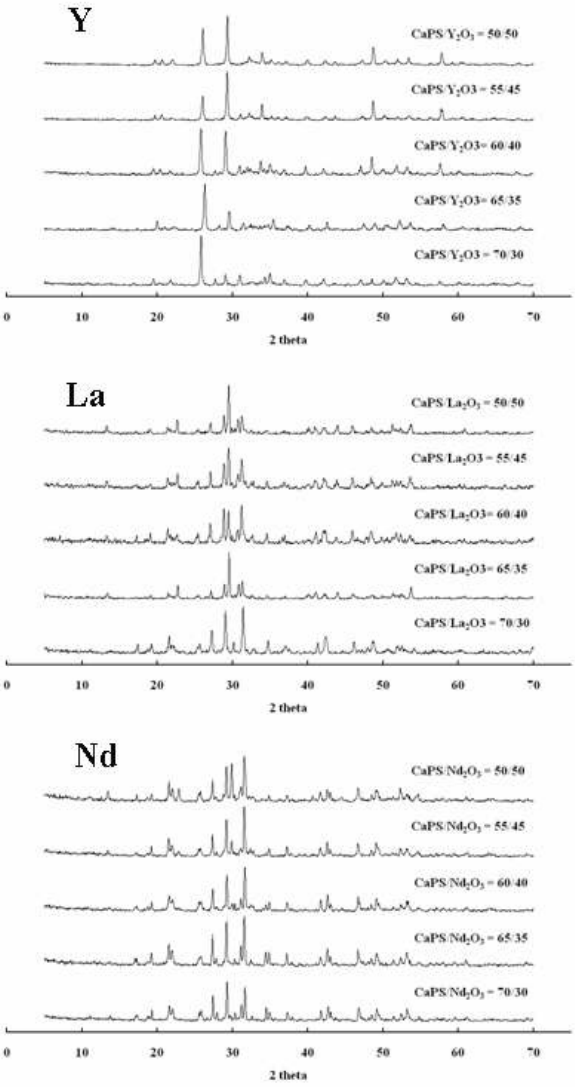

Fig. 2 XRD patterns of each reaction product for Y_2O_3, La_2O_3 and Nd_2O_3.

Fig. 3 Photograph of wasteform containing rare earth (Y, La, Ce and Nd) oxides

Fig. 2 indicated the XRD patterns of each reaction product for Y, La and Nd oxide. The crystalline phases were monazite(main) and oxy-apatite or britholite (minor).

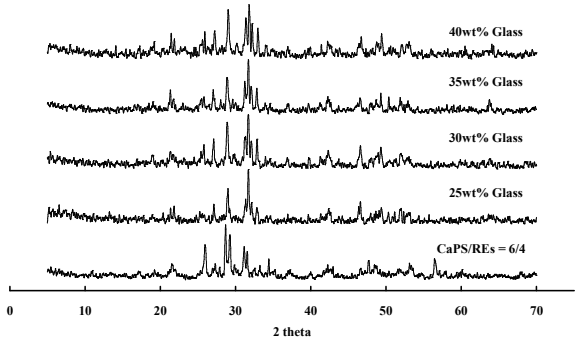

Fig. 4 XRD patterns of wasteforms with different glass

Fig. 3 shows the XRD patterns of prepared wasteforms with different glass ratios. Before addition of glass, the reaction products are mainly monazite. After the addition of glass, the phases were britholite as well as monazite. By using CaPS as a reactive reagent, the target host matrix was successfully synthesized. The chemical durability of the target matrix is under investigation.

3. Conclusion

In the immobilization of α-emitting radionuclides, it is very important to find reliable host matrix. Lanthanide-bearing phase in nature such as monazite, apatite or zircon is verified host matrix which survives the natural environment shock for millions year. By using CaPS reagent, this study intended to synthesize these matrices and the result indicated that monazite and apatite phase containing rare earth element could be obtained.

Acknowledgments

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REFERENCES

[1] R.C. Ewing, Nuclear Wasteform for Actinides, Proceedings of National Academic Science, Vol. 99, p.3432, 1999.
[2] R.C. Ewing, Ceramic Matrices for Plutonium Dispostion, Progress in Nuclear Energy, Vol.49, p.635, 2007.