A Glass-Ceramic Waste Forms for the Immobilization of Rare Earth Oxides from the Pyroprocessing Waste salt

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1. Introduction

The fission product of rare earth (RE) oxide wastes are generates during the pyroprocess [1,2]. Borosilicate glass or some ceramic materials such as monazite, apatite or sodium zirconium phosphate (NZP) have been a prospective host matrix through lots of experimental results [2]. Silicate glasses have long been the preferred waste form for the immobilization of HLW. In immobilization of the RE oxides, the developed process on an industrial scale involves their incorporation into a glass matrix, by melting under 1200~1300℃. Instead of the melting process, glass powder sintering is lower temperature (~900℃) required for the process which implies less demanding conditions for the equipment and a less evaporation of volatile radionuclides. This study reports the behaviors, direct vitrification of RE oxides with glass frit, glass powder sintering of RE-ceramic with glass frit, formation of RE-apatite (or RE-monazite) ceramic according to reaction temperature, and the leach resistance of the solidified waste forms.

2. Methods and Results

All powders were prepared by thoroughly mixed and grinded in a zircon mortar. The vitrified waste form of CeO_2 (20wt%) as a RE oxides was obtained by mixed with R7T7 glass frit and vitrified under 1200℃ and 4 hours using a electrically heated resistance furnace. From the XRD analysis of the vitrified product, a typical oxy-apatite is present. And the SEM image of the product shows the hexagonal shaped crystal. The hexagonal crystals, found to be Ce-rich from EDS analysis. Therefore, the main components (Si and Ca) of the glass binder were exhausted to make crystal phase and weakened the binding effects in crystal interface. The defects would be increase according to increase waste loading.

To prevent of the phenomena, we find a new solidification routes as shown in figure 1. The method is composed of 2-steps reaction. First, conversion of RE oxide into ceramic materials by solid powder reaction, and then the products solidify with glass frit. The synthesis of the monazite ceramic, the behavior of reaction of the cerium oxide with ammonium di-hydrogen phosphate, the oxides was formed the monazite ceramic at above 700 ℃ and 4 hours reaction from the XRD analysis (Figure 2).

Figure 3 shows the solidified waste forms of the Ce and mixed RE-monazite ceramic with R7T7 and VG 98 glass frit at 950℃ and 4hours heat treatment. From the XRD analysis of the waste forms, the crystalline and glass phase were composed of the monazite and glass binder matrix.

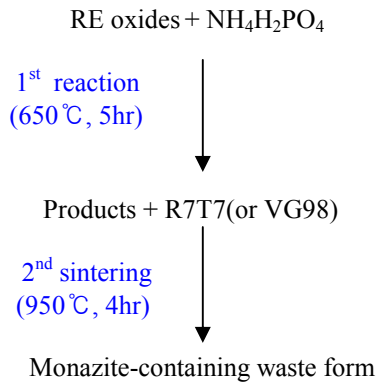

Fig. 1. Manufacturing scheme of glass-ceramic waste form.

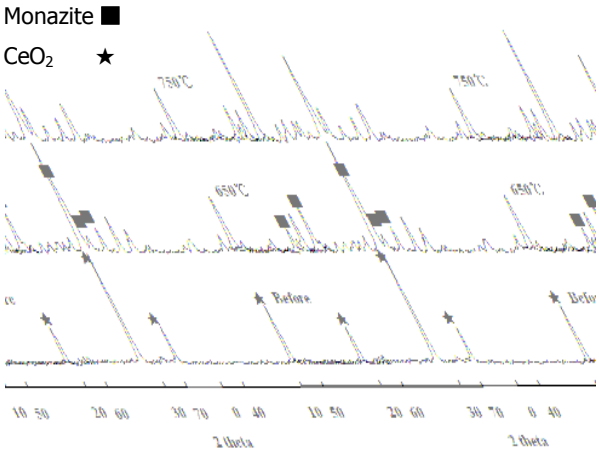

Fig. 2. XRD spectrum obtained from of the reaction product of CeO_2 with ammonium di- hydrogen phosphate according to reaction temperature.

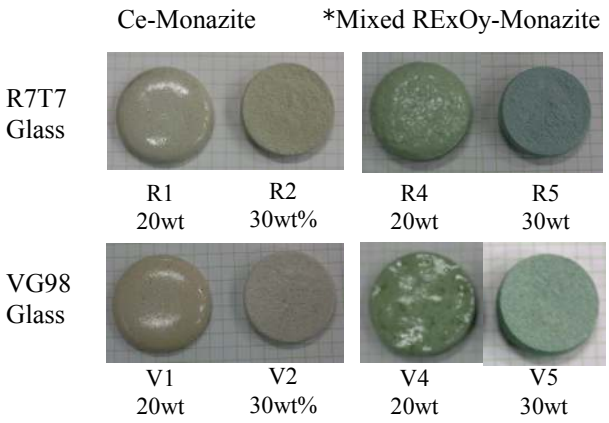

Fig. 3. Photographs of solidified products of the Ce and mixed RE-monazite ceramic with R7T7 and VG 98 glass frit at 950℃ and 4 hours heat treatment. * Mixed RExOy ; CeO_2/ Y_2O_3/ Nd_2O_3/ Pr_2O_3

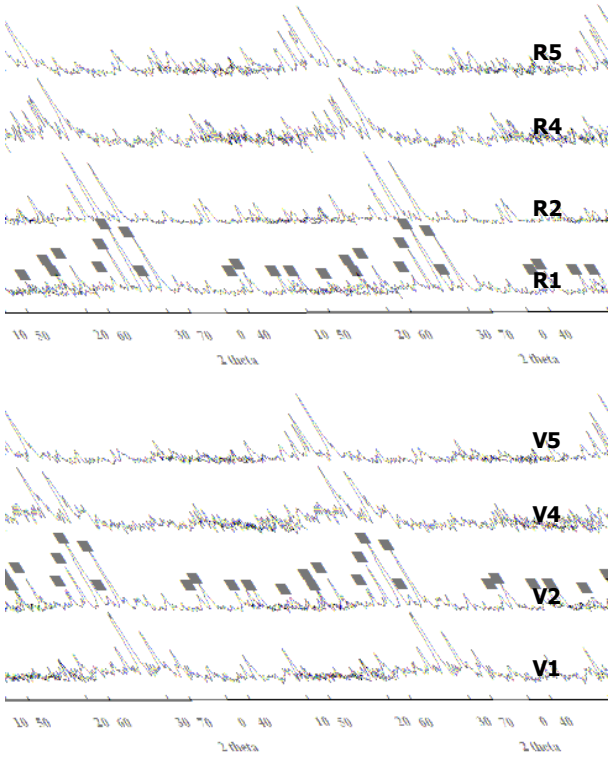

Fig. 4. XRD patterns of the product of the Ce and mixed RE-monazite ceramic with R7T7 and VG 98 glass frit at 950℃ and 4 hours heat treatment. * Mixed RExOy ; CeO_2/ Y_2O_3/ Nd_2O_3/ Pr_2O_3

Conclusion

In order to fabricate a monolithic waste form containing RE oxides, a vitrification at high temperature or ceramization by HIP method is required. In this study, series of monolithic waste form with high waste loadings were successfully produces at mild condition, where the chemical structure was almost equivalent to the product by high temperature process or a monolithic waste forms.

References

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2. Inoue, T. *Prog. Nuc. Energy*, **2002**, 41, 547-554.