Synthesis of Alternative Y_2O_3-based High-temperature Material for Melting Crucible of Metal Fuel

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1. Introduction

The transuranic(TRU) element is used to fabricate metal fuels for Sodium-cooled Fast Reactors(SFRs) [1-3]. U-TRU-Zr-RE metal fuel slugs have been fabricated with an injection casting process operating under atmospheric pressure [4-5]. RE is composed of rare-earth elements consisting of 53wt.% Nd, 25wt.% Ce, 16wt.% Pr, and 6wt.% La. Since highly reactive RE is included during the pyro-processing process, even the plasma-spray coated Y_2O_3 layer on the melting crucible is reacted with RE in the metal fuel and forms the reaction products of RE-Y-O system, which produces a considerable amount of fuel loss and a large amount of radioactive crucible waste. It is necessary to develop an alternative reaction-resistant crucible material that prevents high reactivity with fuel melt to control the fuel loss and reduce the radioactive waste. Alternative Y_2O_3-based crucible material, is of interest as a potential candidate for thermal barrier coating in highly refractory materials due to its chemical and thermal stability [6].

In this study, Y_2O_3-based high-temperature ceramic material was introduced as an alternative reaction-preventing crucible material candidate, referring to a fabrication feasibility and a phase stability. 5 kinds of Y_2O_3-based ceramic pellet were prepared and characterized as a sintering method according to the pseudo binary phase and pseudo ternary phase diagrams.

2. Methods and Results

High temperature ceramic powders of Al_2O_3, Y_2O_3, La_2O_3, Nd_2O_3, ZrO_2 and CeO_2 were used with a purity higher than 99.9% and a particle size smaller than 2.5 ㎛ as raw materials in this experiment. Mixed powder slurry of the ceramic material with was obtained through wet ball milling for 24 hrs. The powder slurry was then annealed by heating at 1000 ℃ for 10 hrs to remove foreign adhering substances including moisture.

Wet ball milling was again performed for 24 hrs to obtain a powder slurry with a uniform composition of the calcined powder. At this time, when the powder slurry was put into the spray-drying apparatus, the rotation speed of the disk was in the range of 6000 rpm to 7000 rpm. Spherical mixed powder ranging from 10 ㎛ to 60 ㎛ in particle size was prepared from the slurry of mixed powders through a spray drying and a sieving process. Green compact was made using the mixed powders by cold isostatic pressing(CIP). Finally, Y_2O_3-based high-temperature ceramic pellets of Y_2O_3-50mol.% Al_2O_3(YAG), Y_2O_3-25mol.%Nd_2O_3-15mol.% La_2O_3, Y_2O_3-30mol% Nd_2O_3-5mol%La_2O_3, Y_2O_3-50mol.% Nd_2O_3-40mol.% ZrO_2, and Y_2O_3-25mol.%Nd_2O_3-15mol.%CeO_2 were synthesized by holding the temperature ranging from 1550ºC to 1620ºC upto 10 hrs. The density of the sintered pellets was determined using an Archimedean immersion method. The microstructure of the sintered synthetics was investigated by scanning electron microscopy combined with energy dispersive spectroscopy(SEM/EDS). The phase structure of the sintered synthetics was examined by X-ray diffraction (XRD).

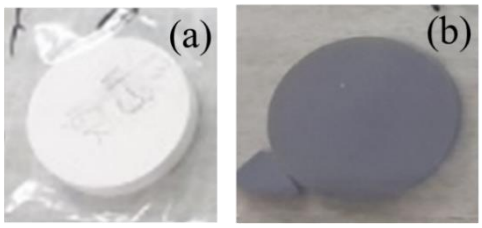


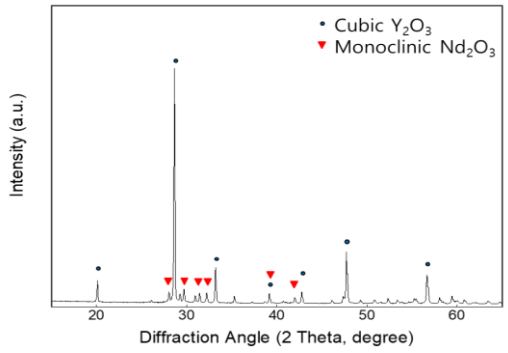
Fig. 1. Typical Y_2O_3-based ceramic pellets synthesized at high temperature; (a) Y_2O_3-50mol.%Al_2O_3(YAG) and (b) Y_2O_3-Nd_2O_3-ZrO_2

Y_2O_3-based ceramic pellets were generally soundly fabricated as showed in Fig. 1. Density of the alternative crucible material according to molar composition was showed in Table 1. As the sintering temperature and the sintering time increased, the density of Y_2O_3-based ceramic pellets prominently increased. The density generally shows an indirect indication of the internal defects such as pores in the sintered pellet. The Y_2O_3-based ceramic pellets sintered below 1550ºC generally showed insufficient densification with a low relative density of 78.8% in average density due to the lack of consolidation. When the sintering temperature was raised to 1620°C, the relative density increased greatly up to 91.1% of theoretical density with sufficient consolidation. Especially, bulk density and relative density of Y_2O_3-25mol.%Nd_2O_3-15mol.%La_2O_3 and

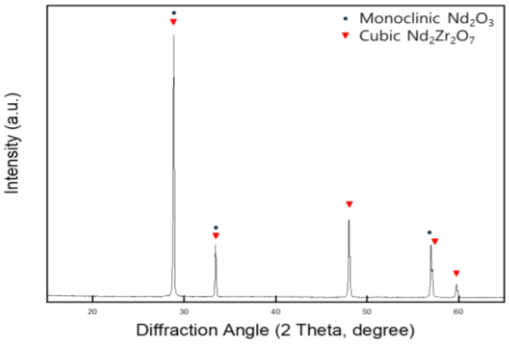
Y_2O_3-30mol% Nd_2O_3-5mol%La_2O_3 pellet showed an excellent densification, with a high relative density of above 99% and low porosity of below 1%.

Table 1. Measured bulk density and relative density of sintered pellets according to molar composition of the Y_2O_3-based ceramic pellets.

Y_2O_3-based crucible material	Measured density (g/cm^3)	Relative density (%)	Porosity (%)
Y_2O_3-50Al_2O_3(YAG)	3.58	79.4	16.5
Y_2O_3-25Nd_2O_3-15La_2O_3	5.81	99.9	0.1
Y_2O_3-30Nd_2O_3-5La_2O_3	5.76	99.8	0.8
Y_2O_3-50Nd_2O_3-40ZrO_2	5.62	89.4	3.6
Y_2O_3-25Nd_2O_3-15CeO_2	5.07	87.2	7.4



(a)



(b)

Fig. 2. Typical X-ray diffraction patterns according to molar composition of the Y_2O_3-based ceramic synthetics.

Typical X-ray diffraction patterns of the sintered synthetic according to alternative material composition are shown in Fig. 2. The major phases of Y_2O_3-30mol.%Nd_2O_3-5mol.%La_2O_3 and Y_2O_3-25mol.% Nd_2O_3-15mol.%La_2O_3 sintered body revealed as a cubic Y_2O_3 and a monoclinic Nd_2O_3 phase as shown in Fig. 2-(a). The major phases of Y_2O_3-50mol.%Nd_2O_3-40mol.% ZrO_2 sintered body revealed as a cubic $Nd_2Zr_2O_7$ and a monoclinic Nd_2O_3 phase as shown in Fig. 2-(b). The XRD analysis result was found to be in good agreement with the previously reported results of the Y_2O_3-Nd_2O_3 and the Y_2O_3-Nd_2O_3 pseudo phase diagram [7].

3. Conclusions

Y_2O_3-based ceramic material as an alternative reaction-preventing crucible material was synthesized by a sintering method at elevated temperature. The bulk density and the relative density of Y_2O_3-25mol.%Nd_2O_3-15mol.%La_2O_3 and Y_2O_3-30mol% Nd_2O_3-5mol%La_2O_3 synthetics showed an excellent densification, with a high relative density of above 99% and low porosity of below 1%. The major phases of Y_2O_3-30mol.%Nd_2O_3-5mol.%La_2O_3 and Y_2O_3-25mol.% Nd_2O_3-15mol.%La_2O_3 sintered body revealed as a cubic Y_2O_3 and a monoclinic Nd_2O_3 phase with phase stability by sintering ranging from 1550°C to 1620°C, irrespectively of the molar ratio of La_2O_3. It is thought that the Y_2O_3-Nd_2O_3-La_2O_3 synthetic is the most promising candidates of the reaction-preventing crucible materials for the injection casting of U-TRU-Zr-RE metal fuels.

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