

Synthesis of LaYO₃ Crucible Material by Solid-state Reaction at Elevated Temperature

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1. Introduction

U-TRU-Zr-RE metal fuels generally have a low centerline temperature and a fuel cycle economy [1]. RE is composed of rare-earth elements consisting of 53wt.% Nd, 25wt.% Ce, 16wt.% Pr, and 6wt.% La. Metal fuel slugs have been fabricated with an injection casting process operating under atmospheric pressure [2]. The metal fuel is melted in a graphite crucible slurry-coated or plasma-spray coated with Y₂O₃ to prevent melt/material interactions [3]. Since highly reactive RE is included during the pyro-processing process, even the plasma-spray coated Y₂O₃ 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. Therefore, 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.

In this study, the perovskite LaYO₃ was introduced as an alternative reaction-preventing crucible material in this study, referring to a fabrication feasibility and a phase stability. LaYO₃ pellets for the melting crucible of metal fuel were prepared and characterized by a sintering method according to pseudo-phase diagram of LaYO₃ and Y₂O₃.

2. Methods and Results

Mixed powder slurries of La₂O₃ and Y₂O₃ powder were obtained through wet ball milling of La₂O₃ and Y₂O₃ powders for 24 hrs. La₂O₃ powder with a purity of 99.999% and a particle size of approximately 2.5 μm and Y₂O₃ powder with a purity of 99.9%. The powder slurry was then annealed by heating at 1100 °C 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. Spherical mixed powders were prepared from the slurry of mixed powders through a spray drying and a sieving process. Green compacts using the mixed powders of La₂O₃ and Y₂O₃ were made by cold isostatic pressing (CIP). LaYO₃ pellets were synthesized at elevated temperatures. The density of the sintered LaYO₃ pellets was determined using an Archimedeian immersion method. The microstructure of the sintered LaYO₃

synthetics was investigated by scanning electron microscopy combined with energy dispersive spectroscopy (SEM/EDS). The phase structure of the sintered LaYO₃ synthetics was examined by X-ray diffraction (XRD).

The X-ray diffraction patterns of the LaYO₃ synthetic according to sintering temperature and molar ratio of 1:1.1 (La₂O₃ and Y₂O₃) are shown in Fig. 1. The LaYO₃ sintered bodies had a similar X-ray diffraction pattern at sintering temperatures ranging from 1450°C to 1550°C for 10 hrs as shown in Fig. 1. The X-ray diffraction patterns of the sintered bodies indicated the formation of a crystallized orthorhombic LaYO₃ phase with a perovskite structure (Pnma) at a sintering temperature of below 1550°C irrespectively of the molar ratio of La₂O₃ and Y₂O₃. The perovskite orthorhombic LaYO₃ phase is a thermodynamically stable phase from room temperature to 1585°C based on the pseudo-phase diagram between La₂O₃ and Y₂O₃. The perovskite structure comprised an orthorhombic atom arrangement of the oxide ions and alternating layers of transition metal ions in the monoclinic coordination. However, it was confirmed that an ordered monoclinic phase (B phase) was formed partially in the LaYO₃ pellet at a sintering temperature of 1600°C, regardless of the molar ratio of La₂O₃ and Y₂O₃. The LaYO₃ formed a stable monoclinic phase (B phase) in the range of 1585°C to 1730°C based on the pseudo-phase diagram between La₂O₃ and Y₂O₃.

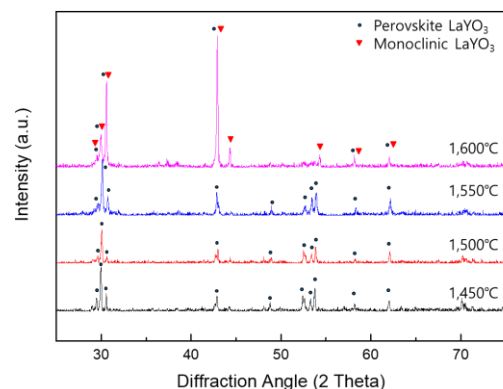


Fig. 1. The X-ray diffraction patterns of the LaYO₃ synthetics according to sintering temperature.

The scanning electron micrographs of the LaYO₃ pellet according to sintering temperature are shown in Fig. 2. The cross-sectioned micrographs of the LaYO₃

pellet showed a similar densification at a constant sintering temperature, independently of the molar ratio. The LaYO_3 pellets sintered at 1450°C and 1500°C showed many pores and an insufficient densification due to a lack of consolidation. The LaYO_3 pellets sintered at 1550°C and 1600°C indicated a considerable densification and small pores, resulting in a high relative density of 89.8% and 97.1% with the progress of consolidation.

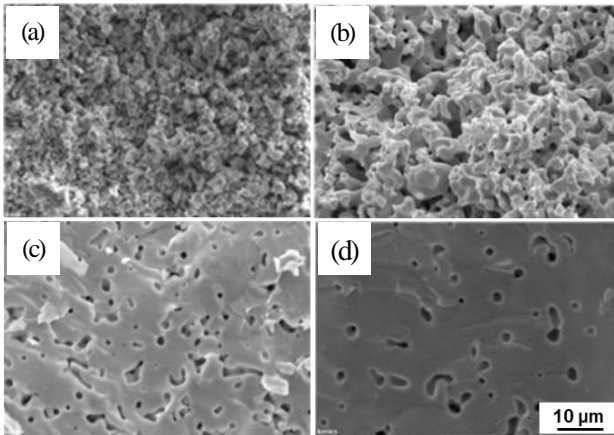


Fig. 2. The scanning electron micrographs of the LaYO_3 pellets according to sintering temperature: (a) 1450°C , (b) 1500°C , (c) 1550°C , (d) 1600°C .

3. Conclusions

LaYO_3 material as an alternative reaction-preventing crucible material was synthesized by a sintering method. The LaYO_3 pellets were composed of a perovskite orthorhombic LaYO_3 phase with phase stability by sintering ranging from 1450°C to 1550°C , irrespectively of the molar ratio of La_2O_3 and Y_2O_3 . The LaYO_3 pellets sintered at 1550°C and 1600°C , independently of the molar ratio. It is thought that the LaYO_3 synthetic is a promising candidate for reaction-preventing crucible material for the injection casting of U-TRU-Zr-RE metal fuels.

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