

SANG-GYU PARK^{1*}, KI-HWAN KIM¹, JUN HWAN KIM¹

DEVELOPMENT OF NdYO₃ POWDER FABRICATION AS A REACTION PREVENTING RAW MATERIAL FOR METAL FUEL CASTING

Metal fuel is a promising candidate for the pyro-processed nuclear fuel, but the problem of loss of nuclear material due to the high reactivity of metal fuel and melting crucible in the metal fuel casting process must be solved for loss control and waste reduction. In this study, fabrication test was conducted to develop a new material NdYO₃ as a new crucible material to improve the degree of anti-reactivity. The NdYO₃ compact was manufactured by the CIP (Cold isostatic pressing) method with changing fraction of Nd₂O₃ and Y₂O₃ powders. Sintering process was performed at 1550°C for 10 hours. The systematic trends of XRD patterns shows that phase transformations from cubic structure to monoclinic structures occurred with the addition of Y₂O₃. The rate of pore were discussed with change of fraction of Nd₂O₃ and Y₂O₃.

Keywords: Metal fuel; Reaction prevention; Y₂O₃; NdYO₃; Sintering

1. Introduction

Fast reactor in nuclear systems is an important attractive energy source [1,2]. Metallic fuel of U-Zr system for the fast reactor has been developed with the pyro-processing of spent fuel [3,4]. Metal fuel is manufactured using an injection casting and uses an induction heating process for the melting due to various advantages. [5,6]. The problem of loss of nuclear material due to the high reactivity of metal fuel and melting crucible in the metal fuel casting process must be solved for loss control and waste reduction. To this end, it is necessary to optimize the process for reducing the reaction loss and secure a crucible manufacturing technology capable of controlling the reaction loss. In particular, it is necessary to develop a new coating material that can minimize high reactivity in order to develop new material casting parts capable of reducing reaction loss.

Y₂O₃ is a conventional crucible coating material for preventing the reaction of the melt with various crucibles. However, the Y₂O₃ coating layer was interacted with the RE's (Rare earth) elements, damaged, and detached after casting at high temperature process. In order to reduce the interaction, researchers improved density and adhesion of the Y₂O₃ coating layers. Dense and strongly bonded coating layers reduced the interaction, but

the protection effects were not remarkable, and coating materials were detached after the cast. It indicates that an enhanced reaction preventing system is necessary for a crucible of casting. Some researchers also attempted to develop the alternative materials, but reported materials did not improve the reaction preventing effect of the Y₂O₃.

In this study, fabrication test was conducted to develop a new material NdYO₃ to improve the degree of anti-reactivity of the crucible. For the powder composition, five conditions of Nd₂O₃ and Y₂O₃ molar ratio standards (60:40, 50:50, 45:55, 40:60, 30:70) were selected to investigate the formation behavior and phase transformations in NdYO₃. The correlation between phase and pore rates were also considered.

2. Experimental

As for raw material powder specifications, Nd₂O₃ powder had a purity of 99.999% and an average particle size of 2.5 μm, and Y₂O₃ powder had a purity of 99.9% and an average particle size of 0.5 μm. Five types of molar composition ratios of Nd₂O₃ and Y₂O₃ powders were selected follows; NdY1(60:40), NdY2(50:50), NdY3(45:55), NdY4(40:60) and NdY5(30:70). Due to the severe hygroscopicity of Nd₂O₃-Y₂O₃, the raw

¹ KOREA ATOMIC ENERGY RESEARCH INSTITUTE, NEXT-GENERATION FUEL TECHNOLOGY DEVELOPMENT DIVISION, 989-111, DAEDEOK-DAERO, YUSEONG-GU, DAEJEON, 34057, REPUBLIC OF KOREA

* Corresponding author: sgpark82@kaeri.re.kr



material needs to be preheated at 1000°C for 1 hour to remove moisture before weighing.

Powders were milled for 24 hours with alcohol and ceramic balls in a plastic ball mill container to obtain a mixed composite powder. While stirring and heating the slurry in which the composite powder was mixed, the alcohol solvent in the slurry was heated and evaporated. The dried composite powder was heated at 1000°C for 10 hours to remove foreign substances including moisture. The obtained Nd-Y-O synthetic powder was fabricated in the cold isostatic pressing (CIP) process.

The dried powder was put into an electric furnace through a classification process (powder between #100 and #400 mesh) and sintered at 1550°C for 10 hours. The prepared specimens were subjected to density measurement by Archimedes method. X-ray diffraction (XRD) patterns of the specimens were recorded using an AXS D8 Advance instrument (Bruker, Billerica, MA, USA), and scanning electron microscopy (SEM Philips, XL series) analysis of the bonded specimens.

3. Results and discussion

Fig. 1 shows the phase diagram according to the molar composition ratio of Y_2O_3 in the Nd_2O_3 - Y_2O_3 system [7]. Based on the sintering temperature of 1550°C used in this study, the predicted phases of each sample were monoclinic for NdY1, monoclinic+cubic for NdY2, NdY3, and NdY4, and cubic for NdY5. In general, in a metal fuel manufacturing process with a U-10Zr composition, the metal of the U-Zr composition is heated to 1600°C and melted, and then manufactured through a casting process. When comparing the composition and phase diagram of the materials used in this study, all samples are considered to be able to maintain a uniform phase under processing conditions.

In order to confirm these expected results, XRD pattern analysis was performed on the prepared samples. For pattern analysis, refer to 41-1105 for Nd_2O_3 having a monoclinic

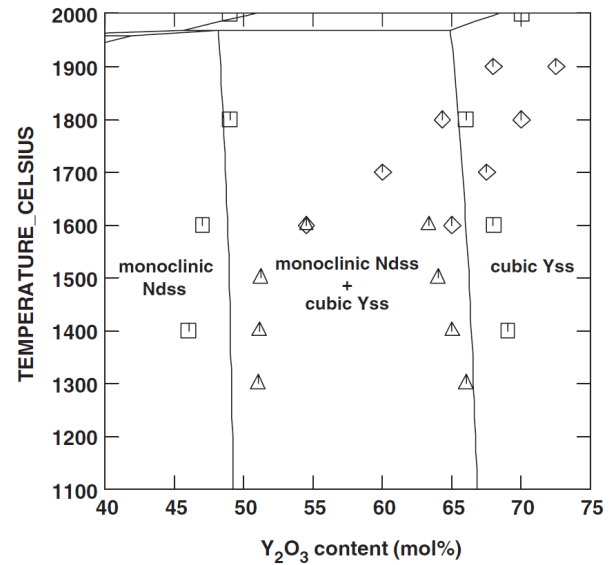


Fig. 1. Phase diagram of Nd_2O_3 - Y_2O_3 system from Shuigen Huang et al. [7]

structure and 28-671 for Y_2O_3 having a cubic structure in the JCPDS card. In the case of NdY1, which was expected to have a monoclinic phase, it was shown that it had a typical monoclinic phase through the 31° peak (501), 42° peak (013), and 47° peak (702). In the case of NdY2 and NdY3 samples, the trend was similar to that of NdY1, but it was analyzed that the intensity of the 47° peak (440) in the simple cubic became stronger and the monoclinic and cubic phases began to coexist. When analyzing the XRD pattern of NdY4, the peak of the cubic phase appears much more clearly than that of NdY2 and NdY3, and peak (501) is observed in 30°~32° region, confirming that the cubic phase and monoclinic phase are still mixed. Finally, NdY5 was analyzed to have a typical cubic phase. The XRD analysis results were found to be in good agreement with the previously reported results of the phase diagram, and it was confirmed that the cubic crystal structure gradually changed as Y_2O_3 was added.

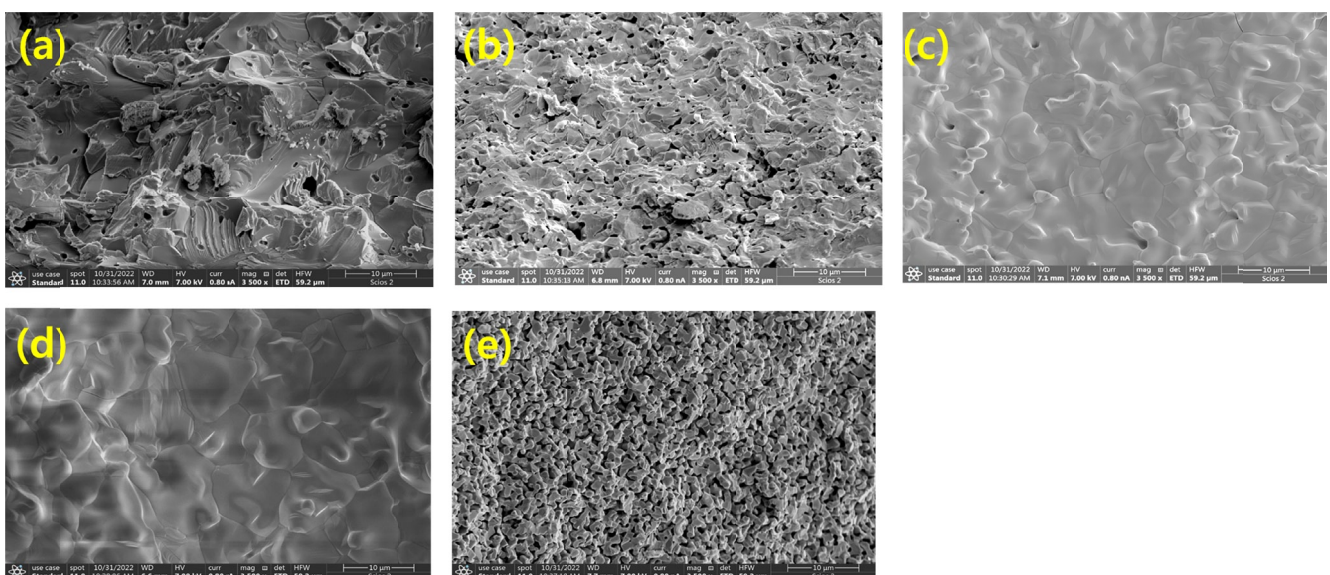


Fig. 2. SEM images of $NdYO_3$ pellets cooled in the furnace after sintering at 1550°C: (a) NdY1, (b) NdY2, (c) NdY3, (d) NdY4, and (e) NdY5

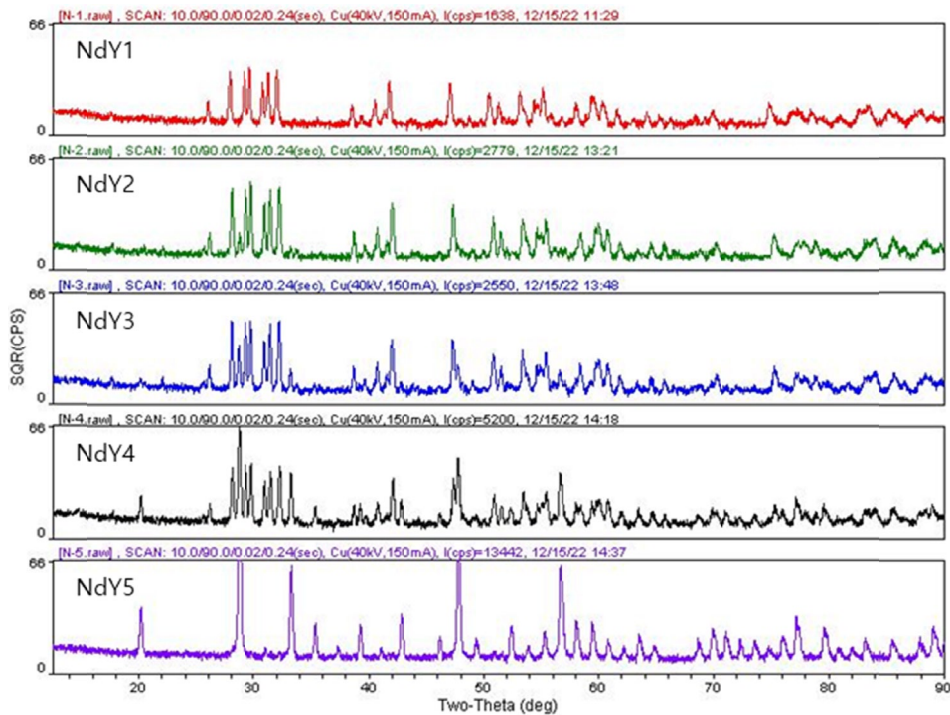


Fig. 3. X-ray diffraction patterns of the samples

The cross section of each sample was analyzed by SEM and shown in Fig. 3. In NdY1 having a monoclinic structure, the overall fracture surface showed a tendency of ductile fracture, and it was confirmed that a small amount of pores were formed inside the fracture surface. In the case of NdY2, the tendency of ductile fracture is more reduced than that of NdY1, and it is seen that it exhibits grain boundary fracture behavior that is destroyed along some grain boundaries. And the internal pores are distributed more than NdY1. Compared to NdY2, NdY3 and NdY4 show a distinctly denser structure, and the internal pores are greatly reduced. On the other hand, in the case of NdY5 having a cubic structure, unlike the other specimens, a high percentage of pores is observed with a fine grain size.

TABLE 1 shows the density and porosity of each sample. Comparing the density change of each sample, it can be seen that the density increases as the mole fraction of Nd_2O_3 increases regardless of the phase change due to the atomic mass of Nd that is relatively higher than that of Y.

TABLE 1
Density & porosity of the samples

Specimen	NdY1	NdY2	NdY3	NdY4	NdY5
Compact ρ (g/cm^3)	6.48	6.27	6.09	5.82	5.25
Apparent ρ (g/cm^3)	6.36	5.99	5.89	5.74	5.07
Pore (%)	1.77	4.36	3.27	1.46	3.43

The density and porosity changes according to the Nd_2O_3 content are shown in Fig. 4. In the section where the Nd_2O_3 content is low (30 mol%), it shows the relatively lowest density and high porosity. Looking at the SEM picture (NdY5), it is thought that high porosity appears inside because the sintering

is not performed completely after the powders are mixed. This will be supplemented through additional experiments in the future. In the section where the cubic and monoclinic phases were mixed (40-50 mol% Nd_2O_3), the density and porosity increased linearly as the content of Nd_2O_3 increased, which was due to the continuous inflow of Nd_2O_3 (Monoclinic) into Y_2O_3 (Cubic). The pore % and density of NdYO_3 showed opposite trends in the single-phase region at 30 mol% Nd_2O_3 and 60 mol% Nd_2O_3 , which is consistent with the phenomenon seen in general ceramic materials. However, the pore % in the section of cubic + monoclinic mixed phases (40-50 mol% Nd_2O_3) show a tendency to increase regardless of the density. It is considered to be increasing lattice mismatch between cubic and monoclinic phases. As proof, it can be confirmed that the difference between the apparent

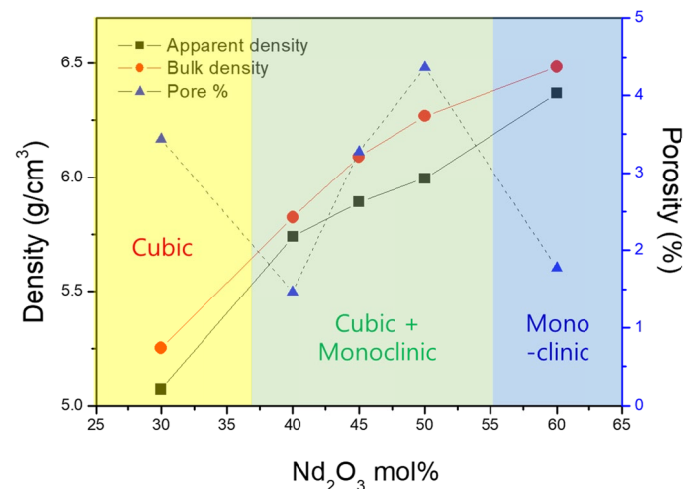


Fig. 4. Summary of the density & porosity of NdYO_3 with increasing Nd_2O_3 contents

density and the bulk density increases as the pore % increases in the 40-50 mol % Nd_2O_3 section in Fig. 4. When examining the characteristics of each sample, from the viewpoint of porosity, NdY4 may be advantageous, but the condition of NdY1, which shows a stable low porosity in the complete monoclinic single phase region, is considered to be the most advantageous in terms of stability and porosity.

4. Conclusions

In this study, fabrication test was conducted to develop a new material NdYO_3 to improve the degree of anti-reactivity of the crucible. Cubic crystal structure gradually changed as Y_2O_3 was added. The density and porosity increased linearly as the content of Nd_2O_3 increased, which was due to the continuous inflow of Nd_2O_3 (Monoclinic) into Y_2O_3 (Cubic). NdY1 (60 mol% Nd_2O_3 – 40 mol% Y_2O_3) seems to be the most advantageous material in terms of stability and porosity.

Acknowledgments

This work was supported by the National Nuclear R&D program funded by Ministry of Science and ICT (RS-2022-00155198).

REFERENCES

- [1] T. Abram, S. Ion, *Energy Policy* **36**, 4323-4330 (2008).
- [2] Generation IV International Forum, *A Technology Roadmap for Generation IV Nuclear Energy Systems*, 2002.
- [3] H. Lee, G.I. Park, E.H. Kim, *Nucl. Eng. Technol.* **43**, 317-328 (2011).
- [4] J.I. Jang, *Nucl. Eng. Technol.* **43**, 161-170 (2007).
- [5] G. Ledergerber, F. Ingold, R.W. Stratton et al., *Nucl. Tech.* **114**, 194-203 (1996).
- [6] G. Bart, F.B. Botta, C.W. Hoth, G. Ledergerber, R.E. Mason, R.W. Stratton, *J. Nucl. Mater.* **376**, 47-59 (2008).
- [7] S. Huang, O.V. Biest, J. Vleugelsw, *J. Am. Ceram. Soc.* **89**, 2596-2601 (2006).