FABRICATION OF YTTRIA STABILIZED ZIRCONIA PELLETS CONTAINING LUMPED GD_2O_3 FOR BURNABLE ABSORBER APPLICATION

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ABSTRACT: *Oxide pellets containing either one of a lumped Gd_2O_3 rod, a minipellet, or a sphere were fabricated by the combination of uni-axial pressing and cold isostatic pressing (CIP) and sintered by microwave (MW) sintering at 1500°C. The effect of MW sintering on the densification of Gd_2O_3 and 8 mol. % yttria stabilized zirconia (8YSZ) was investigated and the results showed a higher densification of 8YSZ due to the higher dielectric constant of 8YSZ. The optimized conditions for the fabrication of Gd_2O_3 spheres by the drop casting method were investigated. Spherical beads with Gd_2O_3 content of 75 wt. % were produced when the Na alginate concentration was 4 wt. %, and the dropping distance was 3-8 cm. The fabricability of the oxide pellets containing lumped Gd_2O_3 was investigated and the results showed that the formation of interfacial gaps and cracks can be eliminated by controlling the initial density of the Lumped Gd_2O_3.*

KEYWORDS: *Lumped Gd_2O_3, microwave sintering, drop casting.*

I. INTRODUCTION

Burnable absorbers (BA) are one of the control mechanisms for nuclear fission reactors to improve the reactor performance through longer fuel cycle length and higher burnup. The BAs are required to have a high neutron absorption cross section and a burnout matching the fuel depletion.[1] Urania-gadolinia mixed fuel is widely used as BA; because it offers a decreased water displacement and reduced handling and personnel exposure.[2] Urania-gadolinia fuel is usually fabricated by mixing UO_2 with Gd_2O_3 (Ref. 3). However, a better reactivity control can be achieved by lumping the Gd_2O_3 into UO_2. Yahya et al. found that the lumping Gd_2O_3 into UO_2 had a better neutronic performance than the conventional urania-gadolinia fuel.[4] The objective of this study is to investigate the fabricability of oxide pellets containing lumped Gd_2O_3 sphere, rod, and minipellet by MW sintering using 8YSZ as a surrogate for UO_2 for the application of new BA design. 8YSZ was used as a surrogate for UO_2 due to its similar fluorite structure, melting, and thermal expansion coefficient.[5,6] Firstly, the sinterability of 8YSZ and Gd_2O_3 by MW was investigated. Secondly, the production of Gd_2O_3 spheres was investigated. Finally, we investigated the fabricability of oxide pellets containing lumped Gd_2O_3 rod, minipellet, and sphere.

II. EXPERIMENTAL PROCEDURES

II.A. Starting Materials

8YSZ (Sigma Aldrich, 99.9 %, ~ 700 nm), Gd_2O_3 (Alpha Aesar, 99.9 %, < 10 µm), Alginic acid sodium salt (Sigma Aldrich) with 15-25 cP viscosity for 1 wt.% in H_2O, and $CaCl_2$ solution (Sigma Aldrich, 1 M) were used as starting materials.

II.B. Gd_2O_3-x wt. % 8YSZ composites

Gd_2O_3 and 8YSZ powder were uni-axially pressed under a pressure of 33.5 MPa followed by CIP under a pressure of 400 MPa, and then sintered in air by MW sintering at 1400-1600°C for 20 min to investigate their sinterability by MW. The densities of the sintered composites were measured using Archimedes' principle.

II.C. Fabrication of Oxide Pellets Containing a Gd_2O_3 Rod or a Minipellet

For oxide pellets containing Gd_2O_3 rod, 8YSZ powder was poured into an annular steel mold with a 2 mm inner hole and 10 mm in diameter and uni-axially pressed under a pressure of 59 MPa. Then, Gd_2O_3 powder was poured in the hole of the 8YSZ annular pellet and pressed again under the same pressure.

For Gd_2O_3 minipellet containing oxide pellets, Gd_2O_3 powder was firstly poured in the hole of the 8YSZ annular pellet. In order to investigate the effect of the initial density of the minipellet on the fabrication process, a 3 mm CIPressed under a pressure of 400 MPa and sintered (at 1600ºC for 2 h in air with heating rate of 10ºC/min) minipellets were inserted into the annular pellet as well. Then, 2 layers of the of 8YSZ powder were poured and uni-axially pressed under a pressure of 57 MPa.

The pressed pellets were CIP at 400 MPa for 5 min. to fabricate green pellets. The green pellets were sintered using MW sintering at 1500ºC for 20 minutes. The microstructure of the sintered pellets was characterized using SEM and EDAX.

II.D. Fabrication of Oxide Pellets Containing a Gd_2O_3 sphere

Firstly, Gd_2O_3 spheres were fabricated using the drop casting method. A slurry was prepared by adding 25-75 wt. % of Gd_2O_3 powder to water under continuous mixing and then adding 3-5 wt.% of Na alginate and continue mixing until all Na alginate was dissolved. The prepared slurry was dropped from different heights into the $CaCl_2$ solution using a syringe with 1 and 2 mm tip size. Then, the synthesized Gd_2O_3 spheres were washed with water for 30 min to remove extra Ca and dried after washing at a temperature of 120ºC for 12 h to remove the remaining water. The effect of Gd_2O_3 content, Na alginate content, dropping height, and the tip size of the syringe on the sphericity and size of Gd_2O_3 spheres were investigated. The sphericity was calculated from the ratio of the short diameter and the long diameter of the spheres and the size was calculated from the average value of them. The diameter of the spheres was measured using a caliper. At least five measurements were performed. Secondly, the fabricated spheres were sintered using conventional sintering at 1600ºC for 2 h in air with a heating rate of 5ºC/min up 1000ºC and 10ºC/min from 1000ºC to the sintering temperature. The density of 10 sintered spheres was measured according to Archimedes principle. Thirdly, the oxide fuel pellets containing sintered Gd_2O_3 sphere were fabricated by pouring half of the total amount of 8YSZ inside the annular steel mold of 10 mm diameter. Then punching the pellet with a 2 mm steel rod and inserting the Gd_2O_3 sphere and pouring the other half in the steel mold and uni-axially pressing under a pressure of 57 MPa to prepare the green pellets. Finally, the green pellets were CIPressed and MW sintered using the same conditions for the oxide pellets containing Gd_2O_3 minipellet and rod. The microstructure of the sintered pellets was characterized using SEM and EDS.

III. RESULTS and DISCUSIONS

III.A. Densification and Phase Transformation of 8YSZ and Gd_2O_3 by MW Sintering

Fig. 1 and Fig. 2 show the effect of the sintering temperature on the densification of Gd_2O_3 and 8YSZ. MW sintering was more effective in sintering 8YSZ than Gd_2O_3. This is due to the higher dielectric constant of 8YSZ for MW and the smaller particle size of YSZ than that of Gd_2O_3.[7,8]

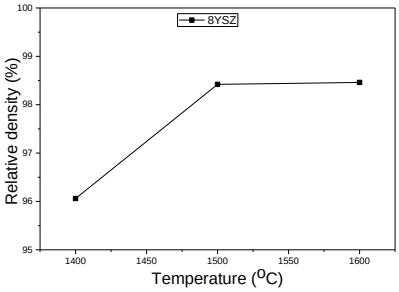

Fig. 1. Densification of 8YSZ by MW

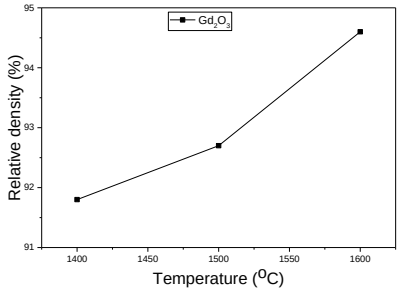

Fig. 2. Densification of Gd_2O_3 by MW.

The density of 8YSZ increased with increasing the sintering temperature from 1400 to 1500°C and then remained almost constant after increasing the sintering temperature to 1600°C. A 98.4 % densification was achieved for 8YSZ powder when sintered at 1500°C for 20 minutes. The density of Gd_2O_3 increased gradually with increasing the sintering temperature. Gd_2O_3 was about 92.5% densified when the sintering temperature was 1500°C. The neutronic performance analysis of Gd_2O_3-cored fuel with 80 % densified Gd_2O_3 showed a better reactivity control than the conventional Urania-Gadolinia fuel.[4]

III.B. Properties of Gd_2O_3 Spheres

The effect of Gd_2O_3 content, Na alginate content, tip size, and dropping distance on the sphercity and size of Gd_2O_3 spheres fabricated by the drop casting method. With increasing the Na alginate content from 3 to 4 wt. %, the sphericity slightly increased and size of the Gd_2O_3 decreased. Concentrations higher than 4 wt.% and less than 3 wt. % produced spheres with a tail shape and deformed shape due to the high and low viscosity of the Na alginate solution. The sphericity was independent of the Gd_2O_3 content. Whereas, the size increased with the increasing of Gd_2O_3 content. With the increasing tip size, the sphericity of the bead decreased and the size increased. However, the sphericity of Gd_2O_3 spheres was found to increase with the decreasing dropping distance from 8 to 3 cm. dropping distance less than 3 cm was found to produce a tail shape spheres and higher than 8 cm was found to produce a flat shape spheres. The highest sphericity of 0.90±0.01 was achieved with 3 cm dropping distance, 4 wt.% of Na alginate, and 75 wt. % of Gd_2O_3 with a sphere size of 2.29±0.01.

III.C. Fabricability of Oxide Pellets Containing Lumped Gd_2O_3

The microstructure of 8YSZ oxide pellets containing Gd_2O_3 rod and minipellet is shown in Fig. 3. No interfacial gap was observed at the interface (Fig. 3 a,c) and an oxide solid solution with a thickness of ~ 1 µm and composition of Gd, Zr and O were observed at the interface between Gd_2O_3 rod/Gd_2O_3 minipellet and 8YSZ annular pellet (Fig. 3 b,d).

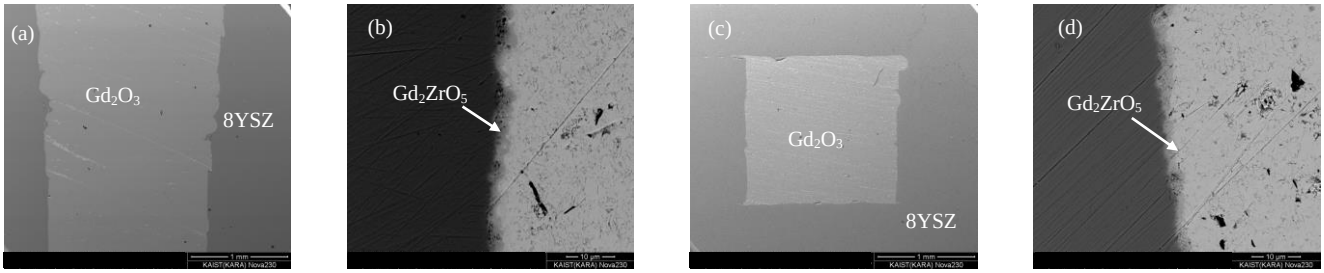

Fig. 3. Microstructure of 8YSZ pellets containing Gd_2O_3 rod and minipellet; (a) SE image of Gd_2O_3 rod, (b) BSE image of Gd_2O_3 rod-8YSZ interface, (c) SE image of Gd_2O_3 minipellet, and (d) BSE image of Gd_2O_3 minipellet-8YSZ interface.

Fig. 4 shows the effect of the initial density of Gd_2O_3 mini-pellets on the fabrication process using CIPressed minipellet and sintered minipellets with a relative density of 66.4±1.0 and 94.0±0.9 respectively. As can be seen from Fig. 4, with increasing the densification of the Gd_2O_3 minipellet, an interfacial cracks and gaps were observed due to the different shrinkage rate of Gd_2O_3 and 8YSZ by MW sintering. The different shrinkage rate is due to their different dielectric constant for MW.[5, 6]

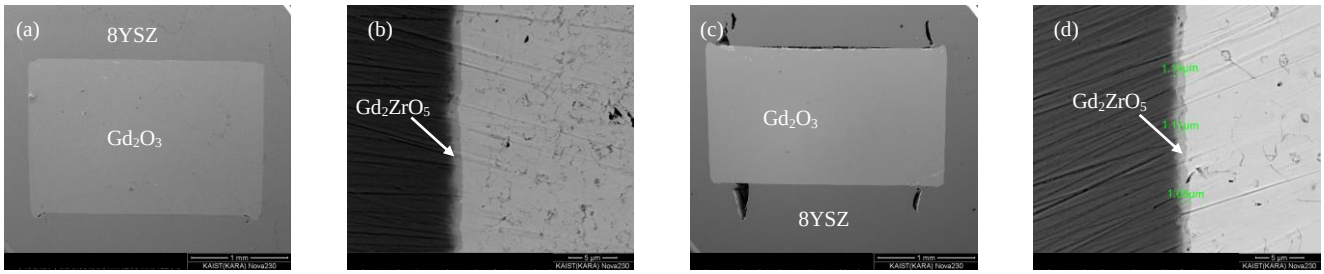

Fig. 4. Microstructure of 8YSZ pellet containing Gd_2O_3 minipellet; (a) SE of CIPressed minipellet, (b) BSE image of CIPressed minipellet -8YSZ interface, (c) SE of sintered minipellet, and (d) BSE image of sintered minipellet -8YSZ

interface.

The microstructure of 8YSZ pellets containing Gd_2O_3 sphere is shown in Fig. 5. No interfacial gaps were observed (Figure 5a) and an oxide solid solution was observed (Fig. 5b). The absence of the interfacial cracks could be attributed to the low relative density of the sintered Gd_2O_3 spheres before insertion. The relative density of the sintered Gd_2O_3 spheres before insertion was around 73.0±1.0.

Fig. 5. Microstructure of 8YSZ pellets containing Gd_2O_3 spheres; (a) SE image of the Gd_2O_3 sphere and (b) BSE image of the interface between Gd_2O_3 sphere and 8YSZ.

IV. CONCLUSIONS

MW sintering of Gd_2O_3 and 8YSZ composites was investigated. The results showed that 8YSZ had a better sinterability due to its higher dielectric constant and smaller particle size. Gd_2O_3 spheres were fabricated by the drop casting method. The optimum conditions for the fabrication of Gd_2O_3 spheres; 3 cm dropping distance, 2 mm tip size, and 4 wt.% of sodium alginate. The size and sphericity of the dried Gd_2O_3 spheres fabricated with optimum conditions were about 2.3 mm and 90 % respectively. The fabrication of oxide pellets containing Gd_2O_3 rod, minipellet, and sphere using microwave sintering was investigated. The results indicated that with controlling the initial density of the lumped Gd_2O_3, the interfacial gaps can be eliminated. Gd_2ZrO_5 phase was observed in the microstructure of 8YSZ pellet containing Gd_2O_3 rod and minipellet due the interaction between Gd_2O_3 and 8YSZ.

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