Fabrication of Neutron Absorber Containing Ceramic Composite Pellets by using Microwave Sintering

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1. Introduction

The life-time and the burn-up of Light Water Reactors (LWRs) fuel can be improved by the application of Burnable Absorbers (BAs). These BAs can control the excess of reactivity added to compensate for fuel depletion, fission product poisons build-up, and the loss of reactivity due to the changes in fuel temperature [1]. The main requirements for BA materials are: (1) a high neutron absorption cross section; (2) a burnout rate matching fuel depletion, and (3) the absence of strongly neutron-absorbing radioisotopes [2]. BAs can be classified into two categories, boron containing materials and rare earth oxides. Due to He gas emission, high sintering temperature, and the relatively low neutron absorption cross section, boron-containing materials are usually applied as a coating on fuel pellets as used in Integral Fuel Burnable Absorber (IFBA) [3] or in separate fuel pellets like Wet Annular Burnable Absorber (WABA) [4]. Rare earth oxides have a high neutron absorption cross section and low sintering temperature and therefore can be mixed with uranium oxide fuel[5]. Urania-Gadolinia mixed oxide fuel is widely used as BA; because it offers decreased water displacement and reduced handling and personnel exposure [6]. A more effective reactivity control can be achieved by lumping Gd_2O_3 into UO_2. Yahya et al. [7] found that a lumped Gd_2O_3 in the center of the fuel rod had a better neutronic performance than the conventional urania-gadollinia fuel.

There are two fabrication methods for urania- gadolinia mixed oxide fuel; dry mechanical milling and co-precipitation. The dry mechanical milling is mostly employed due to its simplicity. In the dry mechanical milling, UO_2 and Gd_2O_3 powder are firstly mixed in 1:1 ratio using shaker mixer and after that, the mix is added to the bulk UO_2 and homogenized. Then, the homogenized mixture is pressed to 50 % theoretical density and finally sintered at 1650°C for 3 h in H_2 atmosphere using the conventional sintering method. Lower sintering temperature and shorter sintering time can be achieved using non-conventional sintering methods like MW and Spark Plasma Sintering (SPS).

The objective of this study is to investigate the fabricability of gadollinia containing duplex ceramic fuel using Microwave (MW) sintering. Fig. 1 shows a schematic diagram for gadollinia containing duplex fuel. Firstly, the sinterability of Yttrium stabilized ZrO_2 (YSZ)

and Gd_2O_3 by MW was investigated. After that, we investigated the fabricability of gadolinia cored ceramic fuel using YSZ as a surrogate for UO_2.
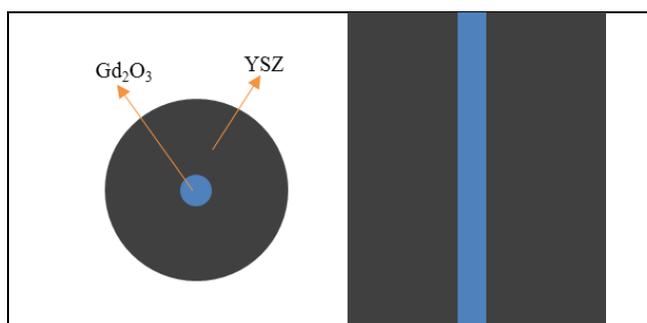

Fig. 1. Schematic diagram for Gdolinia containing fuel pellet

2. Experimental procedures:

ZrO_2- 8 mol. % Y_2O_3 (8YSZ) (Sigma Aldrich, 99.9 %, ~ 700 nm) and Gd_2O_3 (Alpha Aesar, 99.9 %, < 10 μm) were used as starting materials. In order to investigate the sinterability of Gd_2O_3 and 8YSZ using MW sintering, 8YSZ and Gd_2O_3 powder were uni-axially pressed under a pressure of 33.5 MPa, Cold isostatically pressed under a pressure of 400 MPa, and finally sintered using MW sintering at 1400-1600°C for 20 min. The crystal structure was analyzed using X-ray Diffractometry (XRD) and the densification of the sintered samples was measured using Archimedes principle.

An annular mold with an inner hole diameter of 1 mm and an outer cavity diameter of 10 mm was manufactured in order to fabricate gadollinia cored ceramic fuel. 8YSZ powder was poured into the annular mold and uni-axially pressed under a pressure of 57 MPa. Then, Gd_2O_3 powder was poured in the center hole of the YSZ annular fuel pellet and pressed under the same pressure using a steel rod. After that, the uni-axially pressed pellet was cold isostatically pressed at 400 MPa to fabricate green pellets. The green pellets were sintered using MW sintering at 1500-1600°C for 20 minutes. The densification of the sintered pellets was measured at room temperature using the Archimedes method. The microstructure of the sintered pellets was characterized using Scanning Electron Microscopy (SEM).

3. Results and discussions

Figure 1 and Figure 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 with a relative density at 1600ºC of 98.4 % and 94.6 % for 8YSZ and Gd_2O_3 respectively. This is due to the higher dielectric constant for MW of YSZ than that of Gd_2O_3 that might produce a higher densified 8YSZ samples at the same sintering temperature [8-9].
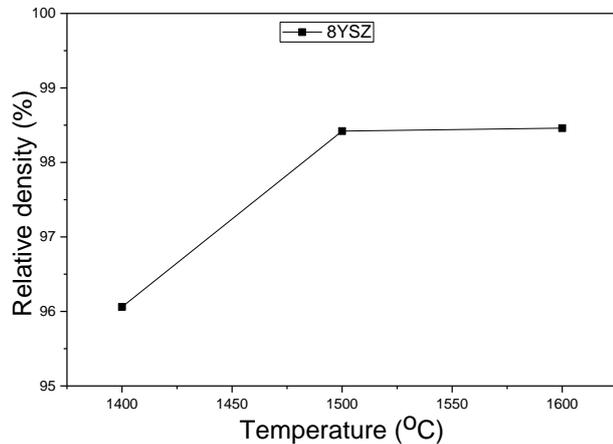

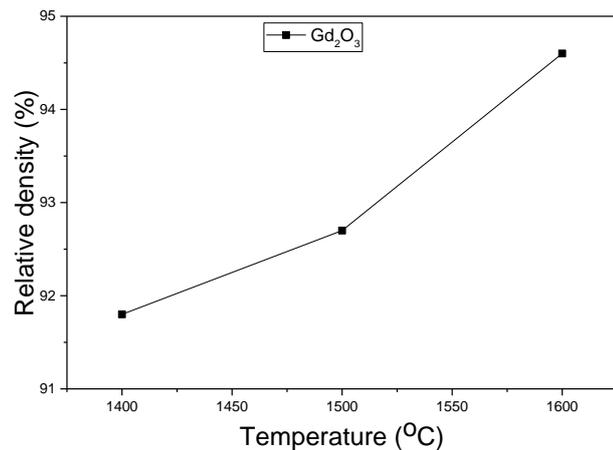Fig. 2. The effect of sintering temperature on the densification of 8YSZ by MW

Fig. 3. The effect of sintering temperature on the densification of Gd_2O_3 by MW.

The density of 8YSZ increased with increasing the sintering temperature from 1400 to 1500ºC and then remained almost constant after increasing the sintering temperature to 1600ºC. On the other hand, the density of Gd_2O_3 increased gradually with increasing the sintering temperature. Upadhyaya et al. [10] and Mazaheri et al. [11] sintered 8YSZ using MW sintering and found that a densification of 97 % for powder sintered at 1540ºC for 35 minutes with an average particle size of 0.69 μm [10] and 97.5 % for powder sintered at 1500ºC with an average particle size of 33 nm was achieved [11] which is comparable to our study where a 98.4 % densification was achieved for 8YSZ powder with 700 nm average particle size when sintered at 1500ºC for 20 min. There is no study about the sinter-ability of Gd_2O_3 by MW sintering and the studies about the sintering of bulk Gd_2O_3 are scarce. Only

one study was recently published about the sinterability of Gd_2O_3 using SPS [12]. In this study, Gd_2O_3 was about 92.5% densified when the sintering temperature was 1500ºC. 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 [7]. The partially densified Gd_2O_3 can also accommodate the nuclear fission products.

X-ray diffraction patterns for Gd_2O_3 and 8YSZ powders and pellets sintered at different sintering temperature are shown in Figure 3 4and 5.
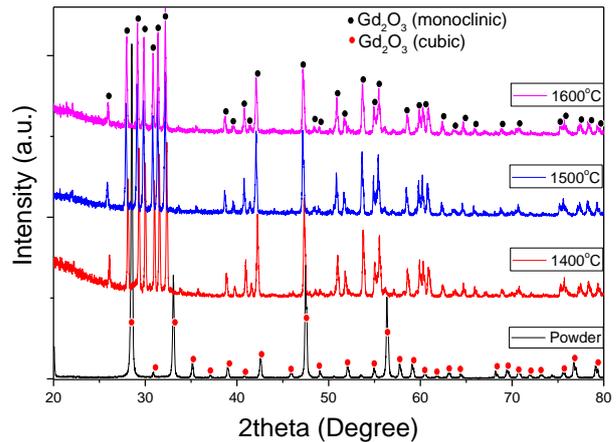

Fig. 4. X-Ray diffraction patterns for Gd_2O_3 powders and sintered Gd_2O_3 pellets.
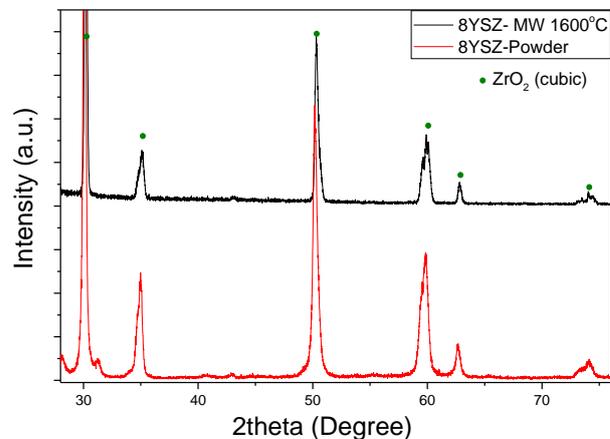

Fig. 5. X-Ray diffraction patterns for Powders and sintered 8YSZ pellets

Gd_2O_3 exists in three different crystal structures; cubic, monoclinic, and hexagonal. At room temperature, Gd_2O_3 has the cubic structure and at about 1250ºC it transforms into monoclinic structure [13]. It was experimentally found that the cubic to monoclinic phase transformation was irreversible even with a slow cooling rate of 2ºC/min [14]. The crystal structure of 8YSZ remained cubic after sintering at temperature up to 1600ºC indicating the stability of the cubic structure of 8YSZ at high temperature.

Figure 6 and 7 show the microstructure of the gadolinia cored 8YSZ samples sintered at 1500ºC and 1600ºC respectively.
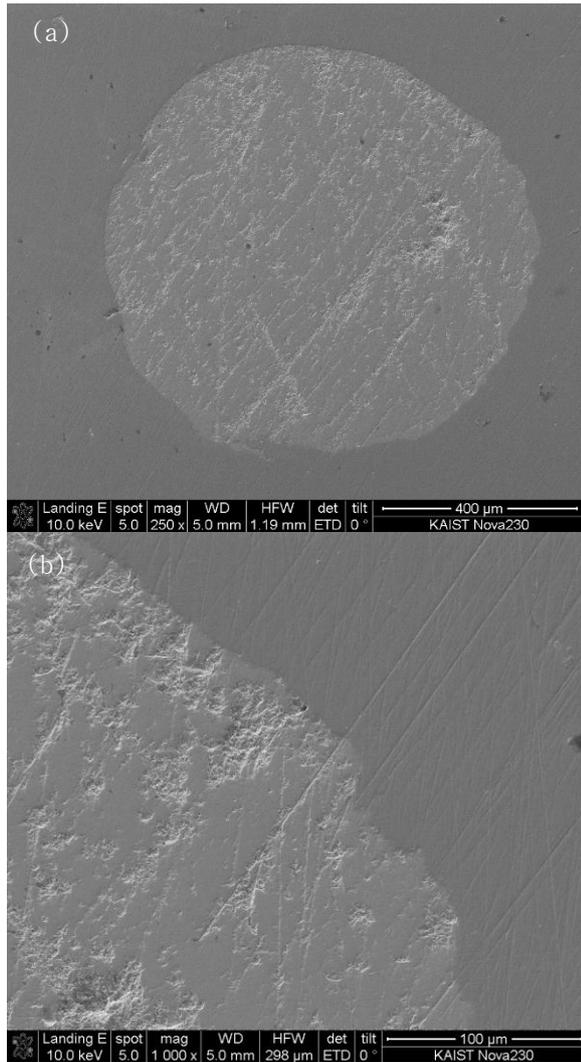
Fig. 6. Microstructure of gadolinia cored 8YSZ pellets sintered at 1500ºC (a) low magnification (b) high magnification
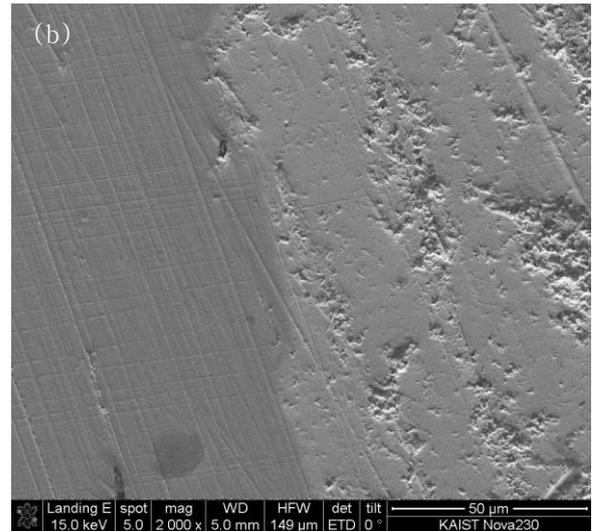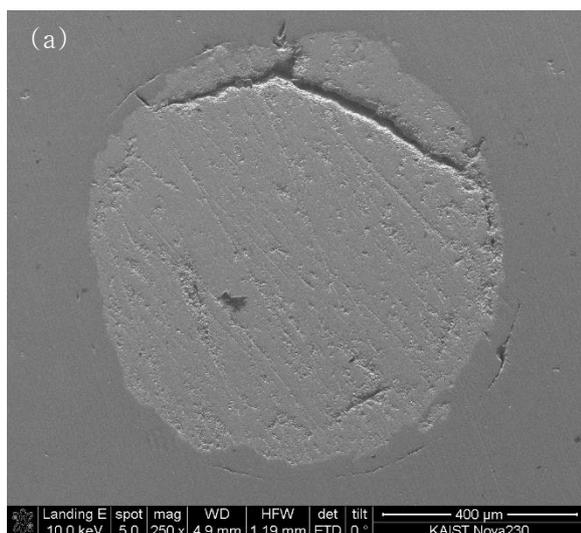

Fig. 7. Microstructure of gadolinia cored 8YSZ pellets sintered at 1600ºC (a) low magnification (b) high magnification.

As can be seen from Figure 6 and 7, No interaction was observed at the interface between Gd_2O_3 and 8YSZ. The Gd_2O_3 region was about 0.8-0.9 mm in diameter. Some cracks were observed near the surface of the Gd_2O_3 region. This might be due to the different shrinkage rate of Gd_2O_3 and 8YSZ. The different shrinkage rate is due to their different dielectric constant for MW [8-9]. A similar cracks were also observed in UO_2-ThO_2 double pellet [15]. Another reason could be due the deformation of the YSZ annular region during the pressing of Gd_2O_3. Therefore, the diamtere of the hole will be changed from 1.0 mm to 2.0 mm in order ro investigate the effects of the hole size on the fabricability and the integrity of the sintered Microstructure.

4. Conclusions

The fabrication of Gd_2O_3 cored ceramic fuel pellets were using microwave sintering was investigated. The results showed that the cracks that formed near Gd_2O_3 region during the fabrication might be due to the different shrinkage rates of YSZ and Gd_2O_3 or the deformation of YSZ region during the pressing. Future studies will investigate the effect of changing the diameter of the hole from 1.00 to 2.00, and stabilizing the cubic structure of Gd_2O_3 on the fabricability of Gd_2O_3 cored ceramic fuel pellets.

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