

Influence of Borosilicate Addition on Mechanical Properties and Sinterability of 8YSZ

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The influence of borosilicate (BS) addition on the mechanical properties and sinterability of yttria-stabilized zirconia (8YSZ) was investigated using scanning electron microscopy (SEM) and Vickers-hardness test. Undoped and 0.5–10 wt.% BS-doped 8YSZ powders were prepared using a colloidal process. The powders were then pelletized under a pressure of 200 MPa. The BS-doped 8YSZ specimens were sintered at 1200–1400 °C for 1 h. SEM results showed that intergranular glass phases were formed at the grain boundaries of 8YSZ, particularly in the 5 and 10 wt.% BS-doped 8YSZ specimens. These intergranular glass phases caused a decrease in grain size of 8YSZ. The grain sizes of the undoped and 0.5 wt.% BS-doped 8YSZ specimens were 2.68 μm and 1.83 μm , respectively. The addition of BS to 8YSZ caused a decrease in the relative density. In addition, the relative density was found to be increased with higher sintering temperature. The hardness of 8YSZ decreased with the increase in BS dopant amount, due to presence at the grain boundaries of a BS glassy phase that has a lower hardness than 8YSZ.

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1. Introduction

Grain growth occurs in ceramic materials during sintering and annealing at high temperatures. The mechanical properties of coarse-grained ceramic materials are inferior to those of the fine-grained ones. Many methods have been developed to prevent the grain coarsening and to make the structure stable during the sintering and heat treatment at high temperatures. These methods include making composite ceramics, using additives that facilitate sintering (liquid phase sintering), and cold and hot isostatic pressing processes [1].

8 mol.% yttria-stabilized cubic zirconia (8YSZ) has a wide range of applications in many areas, such as aerospace, automotive industry, healthcare, manufacture of high-temperature turbine blades, and solid electrolytes. Because 8YSZ has high oxygen ionic conductivity in a wide temperature range and oxygen partial pressure, it is often used as a solid electrolyte in solid oxide fuel cells (SOFCs) [2–7]. For these applications, 8YSZ should have high ionic conductivity, high strength, and considerable chemical and electrical stability. The fact that it has low mechanical strength at high temperatures limits the use of 8YSZ as a solid electrolyte. Due to excessive grain growth at high temperatures and long-term working conditions, the mechanical strength of the electrolyte material decreases, making the fuel cell unworkable [8].

As mentioned above, 8YSZ exhibits low mechanical strength, due to the grain growth at high operating temperature in the SOFCs. The prevention of grain growth in 8YSZ is important in order to improve its

mechanical properties. In this study, the influence of borosilicate (BS) addition on the sinterability and mechanical properties of 8YSZ is investigated using Vickers hardness test and scanning electron microscopy (SEM).

2. Materials and procedure

8YSZ powder (Tosoh, Japan) was used as the matrix material, and the BS powder (Specialty Glass, Florida), up to 10 wt.% was used as dopant. The average grain sizes of the powders were 0.3 and 0.25 μm for 8YSZ and BS, respectively.

The specimens used for the microstructural and mechanical investigations were prepared by colloidal processing. Doping was carried out in a plastic container by mechanically mixing up to 10 wt.% of BS with 8YSZ using zirconia balls and ethanol. Mechanical mixing was performed in a “speks” type mixer at 200 rpm for 12 h. Following mixing, the container lid was removed, and the prepared slurry was left in air for drying during 24 h. After drying, the agglomerated powders were ball milled for 10 min to achieve a uniform dispersion. The powders were then passed through a 60 μm sieve and pressed at 200 MPa in a single-axis die with a radius of 10 mm and a height of 4 mm. The inner surface of the steel die was cleaned after each dry pressing, and stearic acid was applied to its side walls. Sintering was carried out in a box-type furnace under atmospheric pressure. The pressed pellets were pre-sintered at 1000 °C and then sintered at 1200–1400 °C for 1 h. The surfaces of the sintered specimens were ground and polished using metallographic method after the sintering process, and then thermally etched at 1350 °C for 1 h. Microstructural investigations of the sintered specimens were performed using a scanning electron microscope (SEM, JEOL LV 6060). The grain sizes of the specimens were measured using a mean linear intercept method.

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The average grain sizes of the specimens were determined with the following equation:

$$D = \frac{L_i}{N_i M}, \quad (1)$$

where L_i is the length of the line, N_i is the number of grain-boundary intercepts, and M is the magnification in the photomicrograph of the material.

Both the hardness and fracture toughness of the specimens were determined using a Vickers hardness tester with a load of 2 kg and duration of 15 s. Hardness values were calculated using the following equation:

$$H_v = \frac{1854P}{d^2}, \quad (2)$$

where P is the applied load, and d is the mean value of the diagonal length.

3. Results and discussion

The microstructures of specimens doped with various amounts of BS after sintering at 1400 °C for 1 h are presented in Fig. 1. As can be observed from the figures, the undoped 8YSZ shows coarse and co-axial polygonal grains (Fig. 1a). However, BS-doped 8YSZ specimens show smaller grains (Fig. 1b–e). The SEM images

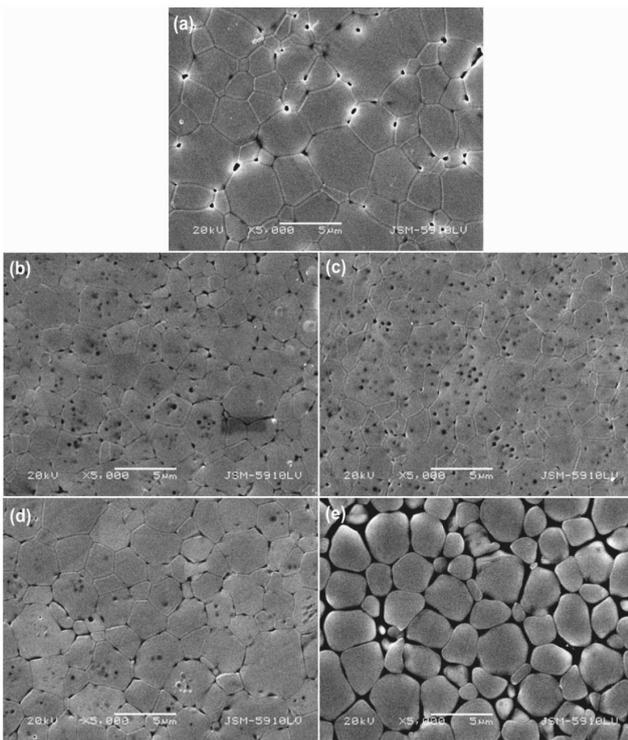


Fig. 1. Microstructures of undoped and BS-doped 8YSZ specimens sintered at 1400 °C for 1 h : (a) Undoped 8YSZ, (b) 0.5 wt.% BS-doped 8YSZ, (c) 1 wt.% BS-doped 8YSZ, (d) 5 wt.% BS-doped 8YSZ, and (e) 10 wt.% BS-doped 8YSZ.

(Fig. 1d–e) and the energy-dispersive X-ray spectroscopy (EDS) results of the 5 and 10 wt.% BS-doped 8YSZ samples show thin glassy phases at the grain boundaries.

The presence of the BS glassy phases at the triple junction of intersecting grain boundaries led to conversion to spherical grains from co-axial polygonal grains. As the BS addition is increased, an increase in thickness of the BS glassy phase layer is observed (Fig. 1d–e).

Figure 2 shows the relationship between the addition of BS and the relative densities of the resulting materials at different temperatures. A relative density of 93% was reached at 1400 °C in the undoped 8YSZ, while the relative densities of 0.5, 1, 5, and 10 wt.% BS-doped 8YSZ specimens were 87%, 91%, 82% and 88%, respectively. As a result, the BS addition to 8YSZ caused a decrease in the relative density, due to presence of the glassy phase layers at the grain boundaries.

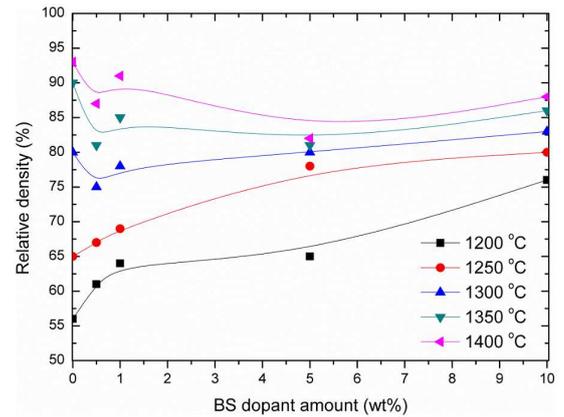


Fig. 2. Relative densities of undoped and BS-doped 8YSZ specimens sintered at various temperatures for 1 h.

As BS amount was increased, it has segregated as the second phase (glassy phase) at the grain boundaries of 8YSZ, leading to an increase in thickness of the glassy layer. This BS glassy phase layer caused loss of contact between the grains of 8YSZ, and a decrease in diffusivity of grain boundary. As is known, the grain boundary diffusion is a powerful agent transport mechanism. The BS glassy layers in the grain boundaries lead to longer grain boundary diffusion paths, and therefore slowed down the diffusion of the atoms along the grain boundaries between the BS glassy phase layers and 8YSZ. Furthermore, another reason for the decrease in densities may be caused by the smaller density of the BS (2.2 g/cm³) compared to 8YSZ (5.95 g/cm³).

Figure 3 shows how grain size of 8YSZ changes with BS content. From this figure, it can be observed that the average grain size decreases with the BS content. The grain size of BS-doped 8YSZ samples decreases, due to the presence of the glassy phase at the grain boundaries of 8YSZ. This BS glassy phase with high viscosity at the grain boundaries causes a decrease in the grain boundary diffusion of 8YSZ. Due to the slow diffusion at the grain boundaries in the BS-doped 8YSZ samples, the grain size is decreased. Intergranular and grain boundary phases with high solubility are known to have low

viscosity and high diffusion rate at high temperatures. BS is known to have a high viscosity because it contains B^{3+} ions network formation. As a result, it is expected to have a low diffusion rate [9].

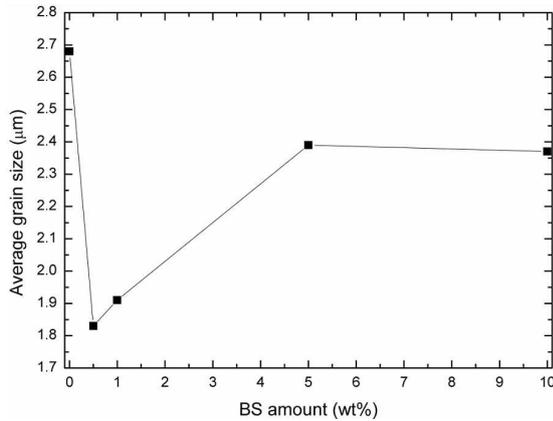


Fig. 3. Plot of grain size versus BS content of doped and undoped 8YSZ specimens after sintering at 1400 °C for 1 h.

The results of Vickers hardness tests of the undoped and BS-doped 8YSZ samples sintered at 1400 °C for 1 h are presented in Fig. 4. The hardness of the undoped, and 0.5, 1, 5, and 10 wt.% BS-doped 8YSZ samples is 12.35, 13.17, 11.04, 5.64, and 8.53 GPa, respectively.

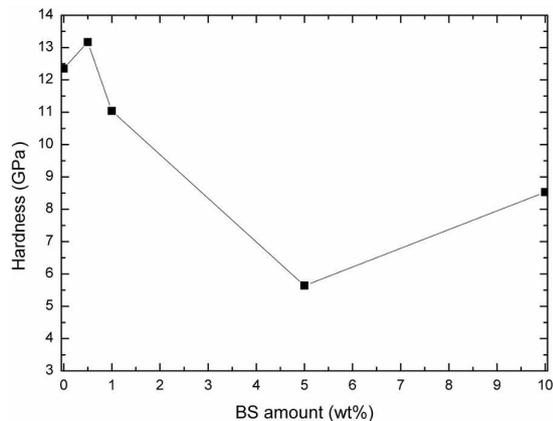


Fig. 4. Effects of BS content on the hardness of 8YSZ samples sintered at 1400 °C for 1 h.

In general, the addition of BS to 8YSZ has led to a decrease in hardness. The reason for this decrease may be the low hardness of the BS glassy phase. The hardness of ceramics is usually affected by the intrinsic deformability of the ceramic and microstructural features, such as multiphases, grain size and orientation, porosity, and grain boundary constitution [10].

4. Conclusions

BS additive has precipitated as a glassy phase around the grains and grain boundaries of 8YSZ. The thickness of glassy phase layer at the grain boundaries has increased with the increase of the BS additive amount. This glassy phase causes slow diffusion of grain boundaries, resulting in a reduction in grain size. The addition of BS to 8YSZ leads to a decrease in the relative density. The relative density also increases with the sintering temperature. The hardness of 8YSZ has decreased with BS addition, due to the existence of the glassy phase.

Acknowledgments

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