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Yttria stabilized tetragonal zirconia ceramics: preparation, characterization and applications

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Abstract

In this study, yttria stabilized tetragonal zirconia (YSTZ) ceramics were prepared and were sintered at different temperatures to find out the optimum sintering temperature for their better tetragonality and mechanical properties for their application as optical ferrule. Vicker's hardness was performed by micro hardness tester and it was found to increase with the increase of sintering temperature to a maximum value, then it was decreased with *higher sintering temperature.* Water absorptivity and porosity were also seen to decrease as the densities of the specimens were increased. X-ray diffraction was employed to determine *crystal structure* of sintered samples. Surface morphology of the sintered samples was examined through field emission scanning electron microscope.

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Introduction

Conventional zirconia ceramics usually have coarse grained structure. By comparing it with fine grained YSTZ ceramics, it is found that YSTZ ceramics are quite different and possess improved sinterability, mechanical properties and super plastic behavior (Yang *et al.,* 2001). YSTZ is one of the most promising ceramic materials that are used in structural applications for its high strength and toughness. As it also possesses high oxygen ion conductivity, it can also be used in electrical applications at elevated temperature and also as fiber optic connector like ferrule (Badwal, 1984; Wu and Wu, 1998; Kubo *et al.,* 2000, Kim and Hwang, 2011). Several studies about the relationship between microstructure and mechanical properties in YSTZ ceramics have been performed over the past decades (Zhang *et al.,* 2004; Lazar *et al*., 2008; Ha *et al.,* 2010). These researches showed the mechanical properties of YSTZ are attributed to the stress-induced martensitic transformation of the metastable tetragonal to the monoclinic phase (Kitano *et al.,* 1988).

Optical ferrules are the fiber optic connector that can protect and align mating fibers for optimum transmission of the optical signal. A set of conditions are needed to be satisfied by the ferrule materials, such as thermal expansion coefficient, stability during environmental changes, hardness close to that of SiO_2 glass, low Young's modulus and quality polished surface. As YSTZ fulfills the above requirements, it is best suited for making ferrules (Badwal, 1984). In this study, YSTZ polycrystals were prepared by mixing aqueous binder and yttria stabilized zirconia powder with a composition of 97 mol% ZrO_2 and 3 mol% Y_2O_3. YSTZ samples were sintered at three different temperatures and their properties were compared to find out the most suitable one for making optical ferrule.

Materials and methods

Preparation of YSTZ

Yttria stabilized Zirconia (YSZ) powder and zirconium oxide nano powder were received from Inframat Advanced material, USA and used as received. All chemicals used in this study were of analytical grade quality with purity of 99.9%. Polyvinyl alcohol (PVA) was used as aqueous binder. Yttria stabilized zirconia (YSZ) powders having composition of 97 mol% ZrO_2 and 3 mol% Y_2O_3, mixture of YSZ and pure zirconia, and zirconium oxide nano powder were taken.

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Before compaction, polyvinyl alcohol (PVA) as binder was mixed with the powder to provide some green strength for subsequent handling. Binder solution was prepared by dissolving 5 gm of PVA in 100 ml distilled water and subsequent heating in a hot plate at about 70-90 °C for 45 minutes with continuous stirring. Green bodies of YSZP were prepared by mixing aqueous binder PVA and YSZ powder. The resultant mixture was then dried at 100 °C for 24 hrs.

Universal testing machine (UTM) (FS-300 kN, Testometric England) was used to prepare pellets at ambient temperature. A 300 kN load cell which is connected to a computer and data analysis software was used to measure the compression force. Ceramic powder was loaded in amounts of 1.0 gm into the die and then compressed at a rate of 2 mm/min until desired pelletizing load of 16 kN was reached. Following compression, the pressure was released after 10 second and the piston was removed. By the same arrangement used to compress samples, was applied to remove the pellet from the die applying compression rate of 2 mm/min.

Sintering of YSTZ

The specimens for optical ferrule were sintered in a high temperature furnace (Z18-40, Micropyretics Heaters Inc., USA), where sintering temperature up to 1800 °C can be achieved. However, for every sintering schedule all samples with similar composition were put in the furnace at the same time. Thus, in one batch at least 6 samples (2 samples of each composition) were placed on a hearth plate. A typical sintering schedule is shown in Fig. 1. The heating rate was 5 °C/min form 0 °C to 600 °C, then holding for 2 hrs at 600 °C, after that heating rate was 10 °C/min up to sintering temperature and holding at sintering temp for 2 hrs. Cooling was done at 10 °C/min. Specimens for optical ferrule were sintered at 1375 °C, 1425 °C and 1475 °C for 2 hrs, followed by furnace cooling.

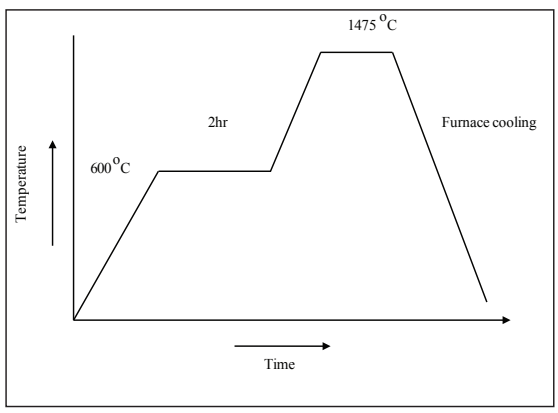

Fig. 1. Sintering schedule of YSTZ

Characterization of YSTZ

Density of the sintered sample was measured by Archimedes' law. Density of the sintered sample was measured by Archimedes principle using the following formula:

$$\rho_s = \frac{m_s \rho_w}{m_s - m_w} \quad \text{.....(1)}$$

Where, is the bulk density of the sintered sample, is the density of water, is the mass of sintered pellet and is the mass of pellet in water. The percentage of porosity was calculated from the bulk density and the theoretical density according to the standard formula:

$$\varphi = \left(1 - \frac{\rho}{\rho_0}\right) \times 100 \quad \text{.....(2)}$$

where, is the porosity,is the density of the sintered pellet and is the theoretical density of the samples.

Micro-hardness of the composites was measured by Vickers indentation using micro-hardness tester (HMV-2, Simadzu, Japan HV-114, Mitutoyo, Japan). The applied load was 50 Kgf for 15 seconds over polished surface. The values of hardness were calculated from the following equation:

$$H_v = 0.01819 \times \frac{F}{d^2} \quad \text{.....(3)}$$

X-ray diffractometer (D_8 Advanced, Bruker GmbH, Germany) employing CuKα radiation of wavelength 1.54046 A˚ with a current of 40 mA and voltage of 40 KV was used in this study. The diffraction intensity was in the range of 12° to 45° of 2θ (Bragg angle), and the scanning speed was 2°/min. The SEM micrograph of YSTZ was done using scanning electron microscope (JSM-6490LA, JEOL Ltd, Japan) at 20 kV.

Results and discussion

Physical and Mechanical properties of YSTZ

YSTZ were sintered at different temperatures. Fig. 2 shows the effect of sintering temperature on physical parameters of YSTZ. It shows that the bulk density of the YSTZ increases with the increase of sintering temperature to a maximum value, then it decreases. As the sintering temperature

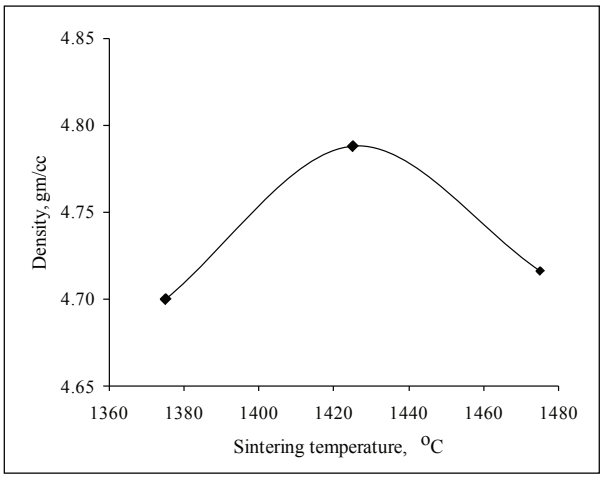

Fig. 2. Effect of sintering temperature on density of YSTZ

increases, enhancement of the process of removing the pores between adjacent particles accompanied by shrinkage of the component occurs. This is combined with growth together and strong bonding between the adjacent particles. The densification/sintering process is activated by the source of energy (e.g. pressure, temperature or chemical). In this case higher sintering temperature, the process is accompanied by more thermal energy. So, higher sintering temperature yields more densification. As the process involves removal of pores and growth process, for sintering temperature higher than a critical value the growth process is faster than the pore removal event. As a result intra-granular void remains in the component resulting in lower density (Exner, 1979).

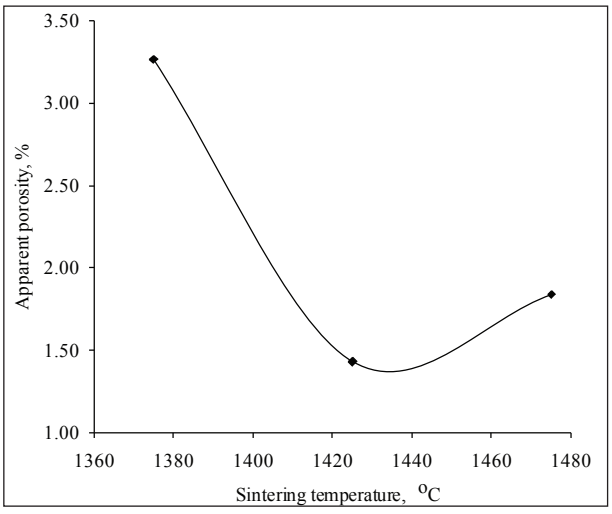

Fig. 3. Effect of sintering temperature on porosity of YSTZ

Fig. 3 and 4 show the effect of sintering temperature on apparent porosity (AP) and water absorption (WA) of the YSTZ composites. Both the figures reveal that with the increase of sintering temperature AP and WA decreased to a minimum value, then it increased. As the porosity is calculated from equation (1), the porosity is reverse to density. The higher the density of the specimen, the lower its porosity. Water absorption depends on the amount of void (open pore) in the structure of the composites. Higher the amount of void more water can be absorbed. So, WA-Sintering temperature curve is very similar to AP-Sintering temperature curves. As a result Fig. 3 and 4 are similar (Naylor and Page, 1983).

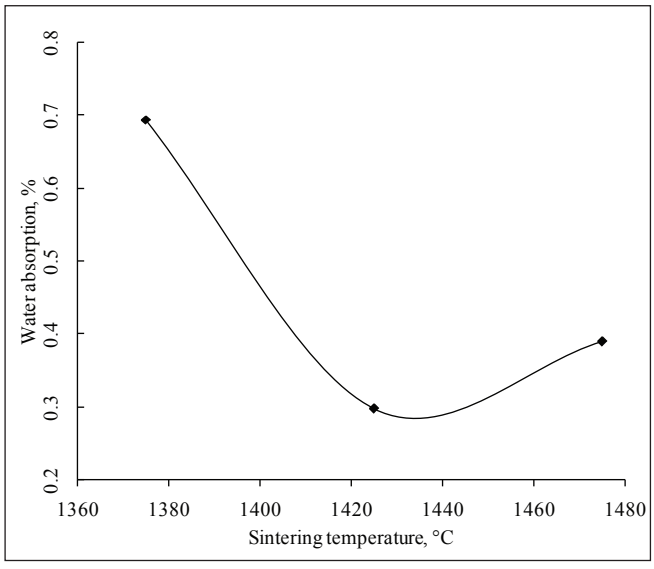

Fig. 4. Effect of sintering temperature on water absorption of YSTZ

Fig. 5 shows the of sintering temperature on the hardness of YSTZ composites It shows that the hardness of the YSTZ increases with the increase of sintering temperature to a maximum value, then it decreases. Hardness of a material is the resistance to indentation. Denser the body, higher is the hardness. As a result Hardness-Sintering Temperature curve follows the Density-Sinsitering Temperature curve (Gogotsi and Bashta, 1992; Naylor and Page, 1983).

X-ray diffraction study of YSTZ

The XRD patterns of YSTZ samples sintered at 1475 °C, 1425 °C, and 1375 °C are shown in Fig. 6. The XRD pattern matched with ICDD file no: 072-7115. The crystal strcture is tetragonal having space group P42/nmC (137). The lattice parameters are a=3.598A^{o}and b=5.185A^o. The major peaks

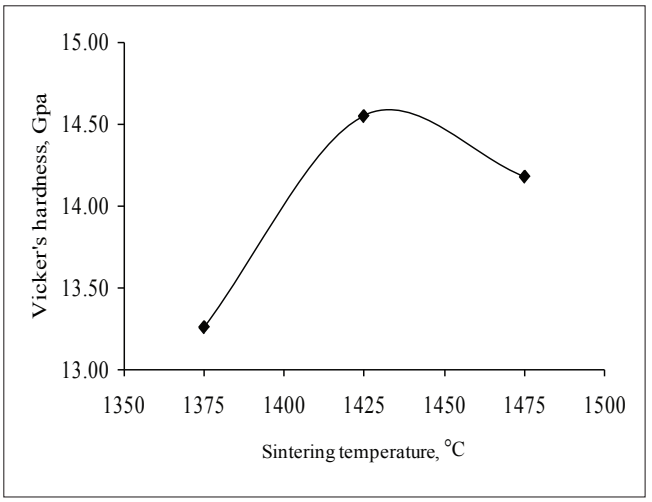

Fig. 5. Effects of sintering temperature on Vicker's hardness of YSTZ

are designated by the miller indices of the planes. The peak intensities are also found to increase with an increase in sintering temperature. A small peak is found at 43° whose plane is 002. Similar results are found in literature published earlier (Lawson, 1995).

Morphological characterization

Fig. 7 depicts the surface morphology of the sintered specimens through field emission-scanning electron microscope (FE-SEM) images at a magnification of ×10,000 for different sintering temperature. 7a. represents the SEM image of sample sintered at 1475 °C, 7b is for the sample sintered at 1425 °C and 7c is the SEM image of YSZT sample for 1375 °C sintering temperature. From the image it is observed that as the sintering temperature increased from 1375 to 1425 °C, the compactness increases by removing the pore between the particles which is supported by Density-Sintering curves. But when the sintering temperature is higher than a critical value, the grain growth is faster than the pore removing rate, leaving pore in the structure. Fig. 7c is accompanied with pores with less denser structure. The grain size is larger than those of 1425 °C and 1375 °C sintering temperature samples.

It was observed that all YSTZ samples had well crystallized grains with 0.1-0.4 μm in size. It also revealed that both grain size and pore size increased during the heat treatment. This partially results from the presence of agglomerates of the fine particles which sinter rapidly, leaving inter-agglomerated pores, and is partly due to the rapid grain growth during which pores are agglomerated by moving with the boundaries

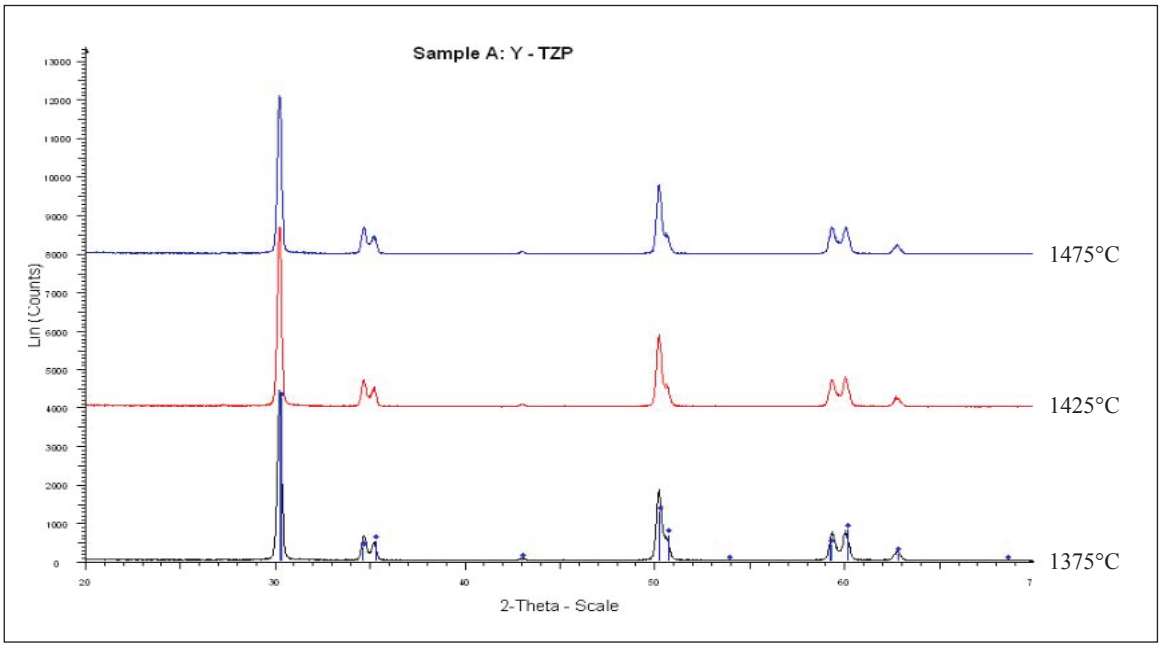

Fig. 6. XRD spectra of YSTZ sintered at different temperature

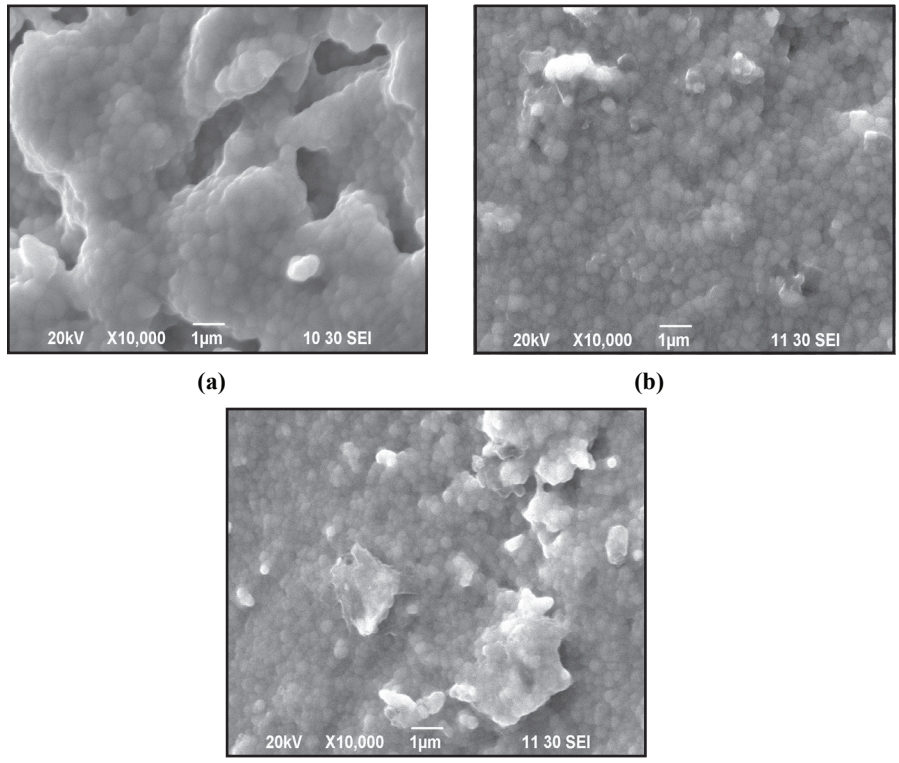

Fig. 7. FE-SEM images (×10,000) of YSTZ surface sintered at (a) 1475 °C, (b) 1425 °C and (c) 1375 °C

(Kingery *et al*., 1991). However for specimen sintered at 1475 °C, some pores were also found because of over sintering. Among three sintering temperatures, well sintered sample was obtained at 1425 °C. The grain sizes of the specimens sintered at 1375 °C, 1425 °C and 1475 °C were almost uniform within 0.1-0.4 µm.

Conclusion

In this work, yttria stabilized tetragonal zirconia (YSTZ) ceramic was prepared from yttria stabilized zirconia. All the specimens were sintered at three different temperatures to find out the effective sintering temperature that best represents the microstructure and mechanical strength. This study reveals the following conclusions:

It is observed that as the sintering temperature was increased to optimum sintering temperature, density increased to maximum value; whereas percentage of water absorption and porosity decreased to a minimum value. Highest hardness value was obtained for the YSTZ composite with the highest density. With further increase of sintering temperature, density and hardness decreased, but apparent porosity and water absorptivity increased. For all the specimens, microstructures contained well-crystallized grains with 0.1-0.4 µm in size, as characterized by FE-SEM. Hardness of the YSTZs were found to be sufficient for making them suitable for their potential applications as optical ferrule.

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