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Effects of SiC, SiO_2 and CNTs nanoadditives on the properties of porous alumina-zirconia ceramics produced by a hybrid freeze casting-space holder method

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ABSTRACT

Highly porous alumina-zirconia ceramics were produced by adding space-holder materials during freeze casting. To increase the strength of porous ceramics, different amounts of nanoadditives (silicon carbide-SiC, silica-SiO_2, and multi-wall carbon nanotubes-CNTs) were added. Space-holder materials were removed by preheating, and solid samples were produced by sintering. Up to 68% porosity was achieved when 40% space-holder was added to the solid load of slurry. Wall thicknesses between pores were more uniform and thinner when nanoadditives were added. Compressive tests revealed that SiC nanoparticles increased the strength more than other nanoadditives, and this was attributed to formation of an alumina-SiC phase and a uniform distribution of SiC nanoparticles. Results indicated that by including 20% space-holder materials and 15% SiC nanoparticles, the density decreases by 33.8% while maintaining a compressive strength of 132.5 MPa and porosity of 43.4%. Relatively low thermal conductivities, less than 3.5 W/K-m, were measured for samples with SiC nanoparticles.

1. Introduction

Porous ceramic materials with unique and unconventional properties such as low density, stability at elevated temperatures, and good compressive strength have applications in a variety of fields [1,2]. Porous ceramics have gained a great deal of attention because of their applications as thermal insulation, liquid/gas filters, bone tissue engineering scaffolds, supports for catalytic agents, and electrodes in fuel cells [3,4]. Furthermore, porous ceramics can possess inherent advantages, such as high melting points, good corrosion resistance, and high strength [5–7]. Specially, porous ceramics with interconnected pore channels are appealing because they have an extremely good permeability and large specific surface area [3,7,8]. The characteristics and applications of porous ceramics are fundamentally governed by their pore morphology and mechanical properties [7,9]. The mechanical strength of porous ceramics is tightly related to the size and wall structures of pores [10–12], which are determined by the processing techniques. Thus, the relationship between the pore structure, process parameters, and the mechanical behavior of porous ceramics must be well understood to design unique pore microstructures with improved strength [13,14].

Various processing routes have been developed [15] to produce porous ceramics. Compared to dry processing [15–17], wet forming methods are widely used for fabricating porous ceramics due to their advantages; in particular, they can give complex-shaped porous bodies with a controlled pore structure and high porosity [16,18].

Among wet forming processes, freeze casting is a promising method for producing porous ceramics, because it is cost effective, and it also reduces drying shrinkage [17,19–22] [23,24]. This method involves preparing ceramic slip, pouring it into a mold, freezing, and sublimating away the solvent (ice to steam transformation) [17,20,22,25,26]. Removing the frozen solvent by sublimation can lead to a near net shaped porous ceramic with uniquely controlled pore channels (e.g., long-range ordered and/or gradient pore structures), mainly depending on the slurry concentration, freezing solvent, freezing direction and rate, and freeze-drying operation [18,21]. Water, camphene, naphthalene-camphor, or tertiary-butyl alcohol (TPA) can be used as a freezing

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agent. However, water- and camphene-based freeze castings develop a dendritic porous network, and they can result in relatively long pore channels parallel to the freezing direction after sublimation, and this due to the unidirectional solidification characteristic of the water solvent [26–28]. To resolve this issue, we propose to add space-holder materials during freeze casting to connect the parallel channels and produce open-pore net structures [28–31]. In this way, temporary powder particles (space-holder) are developed as pore formers [13,32].

Utilizing space-holder materials is considered to be one of the viable methods for making porous materials [13,32–34]. In general, the open-pore manufacturing process utilizing space-holder materials is divided into four main steps: (i) mixing matrix powder and contract space particles, (ii) pressure granulated materials, (iii) remove the space-holder granules, and (iv) sinter porous bodies.

To increase the strength of the walls between the voids in porous ceramics, it is essential to add reinforcement materials; generally, the most effective additions are carbides and oxides in the form of particles, fibers, or tubes [35]. The size of particles may affect the pore structure, therefore to minimize this effect, it is suggested to use nano size additions [36]. In this work we investigate the effect of nano size SiC, SiO_2, and CNTs on the void structures and the strength of the walls between the voids.

In this work we propose to prepare highly porous alumina-zirconia ceramics by a hybrid method utilizing freeze casting and addition of space-holder materials (porous forming). The space-holder materials will affect the solid spaces between the large, parallel freeze casting voids, and they are expected to create some small channels between these large voids [16]. We also aim to study the effects of additions of nanoadditives, SiC, SiO_2, and CNTs, on the pore morphology, compressive strength and thermal conductivity to find out suitability of produced porous ceramics for thermal barrier coating applications.

2. Preparing porous ceramics

2.1. Materials

For this study, α-Al_2O_3 powder (CT-3000-SG and CT-3000-SDP Almatis, Germany) was mixed t-ZrO_2 (TZ-3YE Tosoh, Japan). The mean particle size of Al_2O_3 and ZrO_2 in the green compact were 2.7 μm and 4.5 μm, respectively. Silicon carbide β-SiC nanoparticle powder (with an average particle size of 30–20 nm), silica α-SiO_2 nanoparticles (with an average particle size of 50–35 nm), and Multi Wall Carbon Nanotubes MWCNTs (with an average particle size of 22–18 nm) were used as nanoadditives. Aspecial wax (paraffin wax, initially a solid plate which was melted in an oven) and cornstarch powder (Belgian starch, with particle size of 2.6 μm) were used as space-holder materials. Polyvinyl alcohol (PVA) powders (with particle size 4.2 μm) were used as the binding material. The initial density of the compacted samples (without space-holder and nanoadditives) was 4.04 g/cm^3.

2.2. Preparation of porous ceramics

The batch of powder slurries with a solid loading of 25–30 vol.% was homogenized by ball milling in distilled water using a polyethylene bottle with alumina ball media for 90 min. Different wt.% of wax and Belgian starch (1 to 1 ratio) as space-holder materials were used to set the amount of porosity. Polyvinyl alcohol was used as the binder material [37,38], and 0.35 wt.% (0.08 vol.%) ethoxylated acetylenic diol surfactant was used to enhance powder surfaces for better attachments. In cases with nanoadditive additions, 1 wt.% citric acid (0.65 vol.%) dispersant was used to inhibit agglomeration of nanoadditives. Table 1 shows the composition of slurries. Each slurry was mixed at 60 °C, then the warm slurry was poured into a cylindrical epoxy mold (with a diameter of 20 mm) protected by a heat insulating layer in which the bottom face of the mold was tightly capped. Subsequently, the mold was placed on a stainless steel plate that was temperature-controlled at less than 0 °C using liquid nitrogen; in such a condition, controlled freezing of the water gradually occurs from the bottom to the top of the specimen which results in creation of connected open pores due to addition of space-holder materials. This may result in unidirectional pore channels through the whole cast body after evaporation. The frozen samples were carefully removed from the mold, and then the suspension medium was sublimated in a freeze drier. After the green bodies were cleaned at 600 °C for 1 h in air (with a heating rate of 2 °C per minute) to remove organic materials, they were sintered in an electric tube furnace at 1550 °C for 2 h.

3. Experimental testing of porous ceramics

3.1. Apparent porosity and apparent density

Apparent density and apparent porosity of produced porous ceramics were measured by the water immersion principle based on "Archimedes' method" according to ASTM C20-00 [39]. The procedure of immersing method is to: i) weigh the dry sample, ii) weigh the immersing sample in distilled water for 25 h, and iii) weigh the saturated sample. The equations of apparent density (AD) and percentage of apparent open porosity (AP) are [18,34,39]:

$$\mathrm{AP} = \frac{W_s - W_d}{W_s - W_i}, \tag{1}$$

$$\mathrm{AD} = \frac{W_d}{W_d - W_i}, \tag{2}$$

where W_d is the dryweight, W_i is the immersed weight and W_s is the saturated weight.

3.2. X-ray diffraction and SEM imaging

Crystalline phases were identified by X-ray diffraction (XRD) using a SHIMADZU X-RAY Diffractometer, XRD-600 with monochromatic CuKα radiation. The applied voltage and current were 40.0 kV and 30.0 mA, respectively. The detection is normally from 10° to 80° with a step size of 0.02° and a speed of 1.2°/min. The size of sample for XRD analysis was 3 mm × 3 mm × 3 mm. The pore size and wall thickness were determined using a Hitachi S-570 SEM, and different areas were arbitrarily selected in each sintered specimen. SEM sample dimensions were 5 mm × 3 mm × 3 mm.

3.3. Compressive strength

The Brazilian disk test, which is a diametrical compression test, was used to measure the compressive strength for all samples [40]. This test is usually used for materials that are too hard to process or machine into ASTM standard's dog-bone samples for tensile testing [28,29,39].

The compressive strength in the direction perpendicular to the freezing direction was measured using a universal testing machine. Sintered specimens were circular disks with dimensions of 10 mm × 5 mm. A tensile machine with a crosshead speed of 5 mm/min (Instron machine/IMR) was used for compression tests.

Compressive Brazilian strength was measured using the following equation [29,39]:

$$\sigma = \frac{2P}{\pi D t}, \tag{3}$$

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