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Effect of trace alumina on mechanical, dielectric, and ablation properties of fused silica ceramics

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ABSTRACT

The effect of a small amount of alumina on the mechanical strength, thermal shock resistance, room temperature and high-temperature dielectric, and ultra-high temperature ablation properties of fused silica ceramics prepared by aqueous gelcasting was investigated. Alumina addition promoted the dissolution of silica during slurry preparation. Compared to a pure fused silica sample, the flexural strength and thermal shock residual strength of fused silica ceramics with 0.5 wt% alumina (sintered at 1250 °C) reached 79.9 MPa and 82.0 MPa, showing increases of 19.3% and 19.9%, respectively. In addition, the ablation resistance increased. The mass loss rate and linear recession rate of fused silica ceramics with 0.5 wt% alumina were 0.018 g/s and 0.133 mm/s at 10 s and 0.026 g/s and 0.089 mm/s at 60 s ablation time, respectively. Importantly, the presence of alumina did not decrease the excellent room temperature and high-temperature dielectric properties of fused silica ceramics.

1. Introduction

Materials for radomes and electromagnetic wave transparent windows are essential for components of spacecrafts. The harsh re-entry environment requires these materials to exhibit good mechanical and dielectric properties, excellent thermal shock resistance, and very high ablation resistance [1,2]. Fused silica ceramics, BN, and Si_3N_4 are potentially used for these applications [3–5]. Fused silica ceramic shows prominent corrosion and thermal shock resistance (no breakage after thermal shock between 25 °C and 1000 °C for more than 20 repetitions), a low and stable dielectric constant (3–4, from room temperature up to 1000 °C), and a very low mass loss rate (0. 0025 mm/s at 2500 °C) [6–10]. However, the relatively low mechanical strength of fused silica ceramics are insufficient for use in advanced spacecrafts with ultra-high speed [11,12]. In effort to improve the mechanical strength, materials including BN [13], Si_3N_4 [14], silica glass fiber [15] and ceramic fiber [16] have been used to reinforce fused silica ceramics. Although desirable dielectric property is maintained in the fiber/SiO_2 composites, little effect on either mechanical properties or ablation resistance has been observed. In addition, high-temperature oxidation of BN and Si_3N_4 can occur, causing deterioration of the properties of the composites, especially of BN/SiO_2 composite materials.

Alumina is a common additive for silica-based ceramics and glasses. Alumina is often found as an impurity in fused silica raw powders and its removal can be expensive, especially in the preparation of high purity fused silica powders. Wilson et al. [17] found that the alumina contamination resulting from the ball-milling process obviously increased high-temperature flexural strength, creep resistance, and thermal contraction properties of sintered fused silica ceramics. However, the effect of low levels of alumina and other reinforced materials on other common properties such as mechanical strength at room temperature, thermal shock resistance, high-temperature dielectric, or ablation properties has been studied in less detail. Here, we investigate the effect of a small amount of alumina on these properties of sintered fused silica ceramics. To do this, fused silica ceramic material was prepared by a low-toxic aqueous gelcasting followed by pressureless sintering.

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Addition of a small amount of alumina (~0.5 wt%) seriously inhibited the densification of fused silica during the sintering process, maintained the excellent high-temperature dielectric property of fused silica ceramics, and surprisingly resulted in increased mechanical strength, thermal shock resistance, and ablation properties. We discuss the detailed mechanism of these effects of trace alumina on fused silica ceramics.

2. Experimental

2.1. Starting materials and experimental procedures

Commercial amorphous silica (purity: 99.93%, d_{50}: 3.74 μm) and α-alumina (purity: 99.84%, d_{50}: 1.72 μm) powders were provided by Donghai Silica Powders Co., Ltd., China, and Zibo Lituo Alumina Co., Ltd., China, respectively. *N'N*-dimethylacrylamide (DMAA, Kowa Co. Ltd., Japan), *N,N'*-methylenebisacrylamide (MBAM, Tianjing Chemical Reagent Research Institute, China), acrylic acid-2-acrylamido-2-methypropane sulfonic acid copolymer (AA/AMPS, Taihe Water Treatment Co., Ltd., China) and ammonium persulfate (APS) were used as gel monomer, crosslinker, dispersant and initiator, respectively, for the gelcasting process. The optimum conditions for the gelcasting process of fused silica ceramics were investigated previously [8,18]. The typical gelcasting process is as follows.

Firstly, a premix solution was prepared by dissolving DMAA (10 wt%) and MBAM (1 wt%) into distilled water. Next, the premix solution, fused silica and aluminum powders, dispersant and mill balls were added into a nylon jar, which was followed by ball-milling for 5 h in a planetary mill to obtain a slurry. After adding APS (2 wt% of DMAA), the slurry was degassed in a vacuum deaeration vessel and then cast into a stainless steel mold (with lid) and soaked in a water bath at 75 °C for an hour. Next, the solidified wet green bodies were demolded and dried in an oven at a constant humidity of 100% and temperature of 40 °C. Finally, the dried green bodies were sintered in an ordinary electric furnace with a holding time of 4 h assisted by heating at the rate of 1 °C/min from 400 °C to 600 °C to decompose the organics.

2.2. Characterization and measurements

The phases of the specimens were identified by an X-ray diffractometer (XRD, RIGAKU, CuK$_\alpha$, Japan). Si^{4+} and Al^{3+} concentration in the slurry was detected by an inductively coupled plasma spectrometer (ICP, OPTMA20000V). The slurry was centrifuged for several times and the upper clear liquid was removed out and diluted for the ICP test. Fourier transform infrared spectra (FT-IR) measurements were taken using a Nexus 670 infrared spectrometer. The microstructures and surface morphology were observed under a field emission scanning electron microscope (FE-SEM, HITACHI, SU8010, Japan) and a transmission electron microscope (TEM, Tecnai G2 F30 S-TWIN, USA). Flexural strength was examined using an universal testing machine (WT-6002, Shenzhen Reger instrument Co. Ltd., China) by the three-point flexural method with a sample dimension of 3 mm × 4 mm × 40 mm and a cross-head speed of 0.5 mm/min. Ten samples were measured to determine an average value. Archimedes method was employed to determine the bulk density and apparent porosity. The thermal shock experiment was carried out by heating sintered samples to 600 °C and then quickly quenching them in water (25 °C). Residual strength is defined as the flexural strength of fused silica samples after thermal shocking.

For dielectric measurements, both sides of the ceramics pellets (thickness: 1.5 mm, diameter: 12 mm) were polished, coated with silver conductive paste, and then heated at 600 °C for 30 min. Room temperature dielectric properties were measured using an impedance analyzer (Agilent 4294A, America) at 1 MHz. High-temperature dielectric properties were measured using a Wayne Kerr 6500B precise impedance analyzer (Wayne Kerr Electronic Instrument Co., China) coupled with a PST-2000HL dielectric measuring system (Wuhan Pusite Instrument Co., China) and a PCT10 Stanford temperature controller (Stanford Research System Instrument, America).

The ultra-high temperature ablation test was carried out under a flowing oxyacetylene torch environment according to the national standard GJB323-96A (China). The pressure and flux of O_2 and C_2H_2 were controlled as 0.4 Mpa and 0.095 Mpa, 0.24 L/s and 0.18 L/s, respectively. Heat flux and flame temperature reached approximately 4187 kW/m^2 and 3100 °C. The distance between the gun tip and the sample was 10 mm, and the flame was perpendicular to the surface of the samples. The ablation samples were Φ30 mm × 10 mm. The mass loss rate and linear recession rate were calculated by the following formulas, respectively.

$$R_m = (m_0 - m_t)/t \quad (1)$$

$$R_l = (l_0 - l_t)/t \quad (2)$$

where R_m and R_l are the mass loss rate and linear recession rate, respectively; m_0 and m_t are the mass before and after ablation; l_0 and l_t represent the thickness at the ablation center before and after ablation; and t is the ablation test time.

3. Results and discussion

Table 1 shows the effect of alumina on density, mechanical strength, thermal shock resistance, and room temperature dielectric properties of sintered fused silica ceramics. Alumina significantly altered the densification of fused silica ceramics. Adding only 0.25 wt% alumina results in a 10.8% increase in apparent porosity (from 6.2% to 17.0%) for fused silica ceramics sintered at 1250 °C. Surprisingly, the mechanical strength and thermal shock resistance increased after adding alumina. When no alumina was added, the fused silica ceramics sintered at 1225 °C possess relatively high flexural strength (67.0 MPa), the highest residual strength (68.4 MPa) after thermal shock, and the lowest room-temperature dielectric loss (tanδ: 2.88×10^{-3}). For the samples in which alumina was added, all sintered (1250 °C) samples exhibited higher flexural strength and residual strength than the pure samples sintered at 1225 °C or 1250 °C. The sample with 0.5 wt% alumina (sintered at 1250 °C) showed the optimal comprehensive properties. The flexural strength and residual strength reached 79.9 MPa and 82.0 MPa, for increases of 19.3% and 19.9%, respectively, compared to the values for the pure fused silica ceramics sintered at 1225 °C. Additionally, the dielectric constant (ε': 3.54) and loss (tanδ: 3.03×10^{-3}) of this 0.5 wt% alumina sample were as good as the pure sample sintered at 1225 °C. The explanation for this positive effect of alumina addition will be analyzed in the measurements presented in Tables 2 and 3 and Figs. 1–5. As shown in Table 2, alumina addition strongly promoted the dissolution of amorphous silica in the ball-milling process during preparation of fused silica slurry. The dissolution mechanism of amorphous silica remains unclear [19,20]. However, the rate of the dissolution of amorphous silica may be related to three main parameters: the composition of the solution, the temperature, and the local interfacial free energy, or the energy needed to create new surfaces. During the high energy ball-milling process, the Si-OH groups in silica particle surface will be broken and Si^{4+} will be released into the solution. At the same time, some Si–OH groups will be peeled off. The pH of the slurry will be weakly acidic owing to the hydrolysis of Si–OH