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Fabrication and mechanical properties of carbon short fiber reinforced barium aluminosilicate glass–ceramic matrix composites

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Abstract

Dense $BaSi_2Al_2O_8$ (BAS) and $Ba_{0.75}Sr_{0.25}Si_2Al_2O_8$ (BSAS) glass–ceramic matrix composites reinforced with carbon short fibers (C_{sf}) were fabricated by hot pressing technique. The microstructure, mechanical properties and fracture behavior of the composites were investigated by X-ray diffraction, scanning and transmission electron microscopies, and three-point bend tests. The carbon fibers had a good chemical compatibility with the glass–ceramic matrices and can effectively reinforce the BAS (or BSAS) glass–ceramic because of associated toughening mechanisms such as crack deflection, fiber bridging and pullout effects. Doping of BAS with 25 mol% $SrSi_2Al_2O_8$ (SAS) can accelerate the hexacelsian to celsian transformation and result in the formation of pure monoclinic celsian in C_{sf}/BSAS composites, which can avoid the undesirable reversible hexacelsian to orthorhombic transformation at ~300 °C and reduce the thermal expansion mismatch between the fiber and matrix.

Keywords: A. Barium aluminosilicate; A. Carbon fibres; A. Short-fibre composites; B. Mechanical properties

1. Introduction

Advanced glass–ceramics, such as barium aluminosilicate ($BaSi_2Al_2O_8$ or BAS) typically exhibit high melting points, low coefficient of thermal expansion, and good oxidation resistance and dielectric properties [1]. For these reasons, they have potential as matrix materials for ceramic matrix composites with applications in electronic packaging, structural components and nuclear shielding [2–6].

Although celsian BAS is the stable phase below 1590 °C and hexacelsian polymorph is stable above 1590 °C, hexacelsian BAS can metastably exist below 1590 °C due to the sluggishness of the hexacelsian to celsian transformation [7]. Hexacelsian shows a large thermal expansion coefficient of $\sim 8.0 \times 10^{-6}\,C^{-1}$ and undergoes a rapid, reversible hexacelsian to orthorhombic transformation at ~300 °C, accompanied by a large volume change of ~3%. This structural transformation during thermal cycling would result in microcracking of the BAS matrix. Therefore, it is needed to promote the kinetics of transformation of hexacelsian into monoclinic celsian in order that BAS glass–ceramic and its composites can be successfully applied.

On the other hand, like other glass–ceramics, the pure BAS ceramic matrix exhibits relatively low mechanical properties, and hence limits its use in many structural applications [8]. Over the past years, various kinds of BAS based composites, including particulate-, whisker-, platelet- and fiber reinforced BAS composites have been extensively investigated [5,6,9,10]. Continuous fiber reinforced composites exhibit superior properties compared with the monolithic BAS matrix [5,6]. The high strength and modulus of the fibers provide superior mechanical properties and can prevent catastrophic brittle failure in composites. While improvements in mechanical properties have been demonstrated, ease of producibility and low cost have been sacrificed.

Short fiber reinforced composites have generated a great deal of attention due to their adaptability to conventional

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manufacturing techniques and low cost of fabrication [11]. Another advantage of the use of short fibers instead of conventionally used long fibers is that the short fibers allow the development of an inner oxidation protection, i.e. the inner short fibers can be protected by the matrix [12]. However, fabrication and properties of short fibers-reinforced BAS composites have not been reported.

In present work, carbon short fiber reinforced BAS glass–ceramic matrix composites were prepared by hot pressing. The role of doping of BAS with strontium aluminosilicate (SAS) on the BAS crystal phase formation was investigated. The effects of sintering pressing parameters and fiber volume fraction on the mechanical performance were also discussed.

2. Experimental

The materials used in this study were BAS glass–ceramic composites reinforced with different volume fractions of short carbon fibers (from 20 to 40 vol%). The BAS matrix powders were synthesized through hydrolysis of alkoxides [13]. The mixed $0.75BaO \cdot 0.25SrO \cdot Al_2O_3 \cdot 2SiO_2$ (BSAS) powders were also synthesized using the same process to improve the hexacelsian-to-celsian phase transformation during sintering. The compositions of BAS and BSAS matrix powders are shown in Table 2.

The carbon fibers used in this study (Jiling Carbon Indus., China) have a diameter of 6–8 μm, and its properties are summarized in Table 1. The fibers were cut to a length of 3–5 mm before fabricating the composites. The short carbon fibers were firstly ultrasonically dispersed into ethanol. Then the matrix powders were added to the above solution. The mixed solutions were further mixed by ball-milling for 4 h. The slurries were dried and aggregates were dispersed by hand as required. The dried blends were then hot-pressed in graphite dies at 1300–1600 °C for 1 h under a pressure of 35 MPa in a nitrogen atmosphere into disks of 50 mm diameter and 6 mm thickness. The densities of the samples were measured by Archimedes' method in distilled water at 20 °C.

The fracture toughness and flexural strength of the composites were measured in air at room temperature. All flexural bars were machined with the tensile surface perpendicular to the hot-pressing axis direction. Flexural strength measurement were performed on bar specimens (3 mm × 4 mm × 36 mm) using a three-point bend fixture with a span of 30 mm. Fracture toughness measurements were performed on single-edge-notch beam specimens (SENB) with a span of 16 mm, and a half -thickness notch was made using a 0.33 mm thick diamond wafering blade. Six bars were tested for each composition.

Table 1
Typical properties of carbon fiber

Diamater (μm)	Density ($g\,cm^{-3}$)	Tensile strength, (MPa)	Tensile modulus (GPa)
6–8	⩾1.76	2930	200–220

Table 2
Compositions of the investigated composites and hot-pressing conditions

Sample	C_{sf} content (vol%)	BAS content (vol%)	Hot-pressing conditions	
			Temperature (°C)	Time (h)
B	0	0	1500	1
30B13	30	70	1300	1
30B14	30	70	1400	1
30B16	30	70	1600	1
20B	20	80	1550	1
30B	30	70	1550	1
40B	40	60	1550	1
30S	30	70 (25 mol% SAS)	1550	1
40S	40	60 (25 mol% SAS)	1550	1

The constitution phases of the composites were determined by X-ray diffractometry (XRD). Fracture surface and crack propagation path produced by Vickers indenter on the composites were examined by scanning electron microscopy (SEM). The microstructures of the composites were characterized by transmission electron microscopy (TEM). Thin foil specimens taken normal to the hot-pressing axis were prepared by dimpling and subsequent ion-beam thinning.

3. Results and discussion

3.1. Densification and phase characterization

The effect of sintering temperature and C_{sf} content on the densification of the composites are shown in Fig. 1. It reveals that the relative density increases with increasing the sintering temperature and decreases with increasing C_{sf} content. The composites with different C_{sf} content could be densified to over 95% of the theoretical density after sintering at 1500 °C for 1 h. The BSAS composites possess a higher relative density than that of BAS composites with same fiber content, as shown in Fig. 2. It is due to the lower eutectic temperature in BSAS system during sintering, which can more effectively promote the densification.

The typical XRD patterns taken from the polished surfaces of the hot-pressed composites are given in Fig. 3. It reveals that the only crystalline phase present in BAS matrix composites is hexacelsian phase, and no celsian was detected, as shown in Fig. 3a. Apparently, it is due to the sluggish kinetics of hexacelsian to celsian phase transformation [7]. On the contrary, for BSAS composites, celsian becomes the predominant crystalline phase, indicating that the incorporation of 25 mol% SAS can effectively promote the hexacelsian- to celsian-$BaAl_2Si_4O_8$ transformation (as shown in Fig. 3b). In both the BAS and SAS systems, hexacelsian is always the first to form, but the kinetics of hexacelsian to celsian transformation in SAS is very fast [14]. BAS and SAS can form solid solutions in the entire composition range, and doping of BAS with SAS could assist the breaking of Ba–O or (Al,Si)–O bonds and hence promoting the polymorphic transformation of