ARTICLE IN PRESS

Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Synthesis and properties of AlN/MAS/Si_3N_4 ternary glass-ceramic composites with in-situ grown rod-like β-Si_3N_4 crystals

Zhiwei Luo*, Jing Zhang, Changwei Lin, Lei Han, Anxian Lu**

School of Materials Science and Engineering, Central South University, Changsha 410083, PR China

ARTICLE INFO

Keywords:
A. Sintering
B. Composites
C. Thermal conductivity
D. Si_3N_4
In-situ grown

ABSTRACT

The AlN/MAS/Si_3N_4 ternary composites with in-situ grown rod-like β-Si_3N_4 were obtained by a two-step sintering process. The microstructure analysis, compositional investigation as well as properties characterization have been systematically performed. The AlN/MAS/Si_3N_4 ternary composites can be densified at 1650 °C in nitrogen atmosphere. The in-situ grown rod-like β-Si_3N_4 grains are beneficial to the improvement of thermal, mechanical, and dielectric properties. The thermal conductivity of the composites was increased from 14.85 to 28.45 W/(m K) by incorporating 25 wt% α-Si_3N_4. The microstructural characterization shows that the in-situ growth of rod-like β-Si_3N_4 crystals leads to high thermal conductivity. The AlN/MAS/Si_3N_4 ternary composite with the highest thermal conductivity shows a low relative dielectric constant of 6.2, a low dielectric loss of 0.0017, a high bending strength of 325 MPa, a high fracture toughness of 4.1 MPa $m^{1/2}$, and a low thermal expansion coefficient ($\alpha_{25-300\ ^\circ C}$) of 5.11 × 10^{-6}/K. This ternary composite with excellent comprehensive performance is expected to be used in high-performance electronic packaging materials.

1. Introduction

Due to low thermal expansion, low dielectric constants, and remarkable chemical stability and mechanical stability, glass-ceramics have attracted much attention in many technological areas [1–3]. The $MgO-Al_2O_3-SiO_2$ (MAS) is a powerful ternary glass-ceramic system which is extensively investigated and classified as a potential material for low temperature co-fired ceramic (LTCC) substrates due to its lower thermal expansion coefficient, which is very close to silicon [4–6]. However, MAS glass-ceramics have low mechanical strength and thermal conductivity. So it has the disadvantage of short service life under thermal fatigue. Therefore, if the thermal conductivity and mechanical strength of the glass-ceramics can be improved, they will have a significant advantage. Aluminum nitride ceramic exhibits excellent properties, such as high thermal conductivity, low dielectric constant, and high mechanical properties. Thus, aluminum nitride is a promising candidate for high-performance electronic devices packaging due to their superior thermal, mechanical and electrical properties [7,8]. Also, some efforts have been made to prepare low-temperature sintered high thermal conductivity ceramics by sintering AlN particles with glasses [9–11].

On the other hand, low-cost Si_3N_4 ceramic composites sintered at low temperature, which exhibits high strength, high toughness, high thermal shock resistance, excellent oxidation resistance and other advantages, have attracted considerable interest for emerging high-temperature advanced engineering applications [12–15]. In recent years, it has been reported that silicon nitride reinforced glass-ceramic matrix composites due to their excellent chemical resistance, thermal shock resistance and mechanical properties [16–18]. Ma et al. studied the thermal conductivity of low-temperature sintered borosilicate glass–AlN composites with β-Si_3N_4 whiskers; the thermal conductivity of the borosilicate glass–AlN ceramic composite was increased from 11.9 to 18.8 W/m K, accompanied by flexural strength up to 226 MPa [11]. However, due to the significant thermal expansion mismatch between the glass matrix and the β-Si_3N_4 whiskers, a large number of microcracks appear in the glass matrix, which deteriorated the mechanical properties of the glass–AlN-Si_3N_4 ceramic composites. At present, the thermal and mechanical properties of AlN composites remain at a moderate level, and there is still room for further improvement.

In this paper, we propose a new approach to build a thermal conducting network in the MAS/AlN composites through in-situ grown β-Si_3N_4 crystals during the sintering process. The results obtained confirmed the feasibility of this approach. Besides, the thermal and mechanical properties of the AlN/MAS/Si_3N_4 ternary composites were also investigated.

* Corresponding author.
** Corresponding author.
E-mail addresses: loaswell@163.com (Z. Luo), axlu@mail.csu.edu.cn (A. Lu).

http://dx.doi.org/10.1016/j.ceramint.2017.10.125
Received 23 September 2017; Received in revised form 15 October 2017; Accepted 18 October 2017
0272-8842/ © 2017 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

Table 1
Composition of the MAS/AlN and MAS/AlN/Si_3N_4 composites investigated in this work.

	Samples no.	Compositions (wt%)		
		Glass frit	AlN	α-Si_3N_4
MAS/AlN composites	GA-10	10	90	0
	GA-20	20	40	0
	GA-30	30	50	0
	GA-40	40	60	0
	GA-50	50	70	0
	GA-60	60	80	0
MAS/AlN/Si_3N_4 composites	GAS-7	40	53	7
	GAS-14	40	46	14
	GAS-20	40	40	20
	GAS-25	40	35	25
	GAS-30	40	30	30
	GAS-35	40	25	35

2. Experimental procedures

The composition of MAS glass consists of 25% MgO, 5% La_2O_3, 25% Al_2O_3 and 50% SiO_2 (mol%). Addition of 5% La_2O_3 in the MAS glass composition aims to improve the transformation of α-Si_3N_4 to β-Si_3N_4 during sintering process of the composites. Analytical reagent MgO (99.9%), La_2O_3 (99.9%), Al_2O_3 (99.9%) and SiO_2 (99.9%) were melted at 1580 °C for 2 h at a heating rate of 5 °C/min. The melt was poured into deionized water to form MAS glass frit. The MAS glass frit was milled by agate balls for 10 h. AlN (D_{50} = 3.0 μm, N > 33.06 wt%, O < 0.36 wt%), α-Si_3N_4 (D_{50} = 0.50 μm, α-phase > 94%) powders and MAS glass frit and were mixed in the appropriate proportion of water-free ethanol using an agate ball as the milling medium for 24 h.

The AlN/MAS binary system was prepared firstly. The sample codes of the binary samples are shown in Table 1. According to the thermal conductivity measurements, the optimal sample of AlN/MAS binary composites was chosen as a new matrix for the ternary system. The AlN/MAS/Si_3N_4 ternary composites in the composition of 40 MAS·(60-x) AlN·x α-Si_3N_4 (x = 7, 14, 20, 25, 30, 35) in weight percent are prepared, which are also shown in Table 1. The powder mixtures were dried and sifted through a 400 mesh sieve. The mixed powders were uniaxially pressed in a steel mold of 40 mm in diameter at a pressure of 30 MPa, followed by cold isostatic pressing with 300 MPa. Sintering was carried out in a graphite-resistance furnace under a nitrogen atmosphere. A two-step sintering procedure was applied, i.e., the samples were first heated to 1650 °C and held there for 0.5 h, and they were then cooled down to 1050 °C and held there for 3 h. The AlN/MAS binary and AlN/MAS/Si_3N_4 ternary samples were prepared by the same process.

The bulk density of the composite samples was measured according to the Archimedes's principle. Phase composition was investigated by an X-ray diffractometer (D/max 2500 model) with Cu-K_α radiation (λ = 0.154 nm). Data was collected from 2θ = 10° to 80° at a scanning rate of 8°/min. Scanning electron microscopy (SEM) was carried out using an FEI Quanta 200 SEM. The bending strength was measured on a specimen bar by using an electronic multipurpose tester (CSS-44100 model). Fracture toughness was determined by the single edge-notched beam method (SENB). All the samples were polished to the 1.5–2.0 μm (2000#) final finish prior to bending strength and fracture toughness tests. Each data point represents an average of at least five specimens for flexure strength and fracture toughness test. The thermal expansion coefficient for the samples with a dimension of 20 mm × 4 mm × 4 mm was obtained by a high-temperature dilatometer (NETZSCH DIL 402C). Thermal conductivity specimens with dimensions of *Φ* 10 mm × 2.0 mm were machined. The thermal diffusivity (λ) for each sintered sample was measured, using the laser-flash method (Model NETZSCH LFA 427). The thermal conductivity was calculated by multiplying the thermal diffusivity with the density and specific heat. The dielectric properties were measured by an impedance analyzer (Wayne-Kerr, 6500B, UK) at 1 MHz.

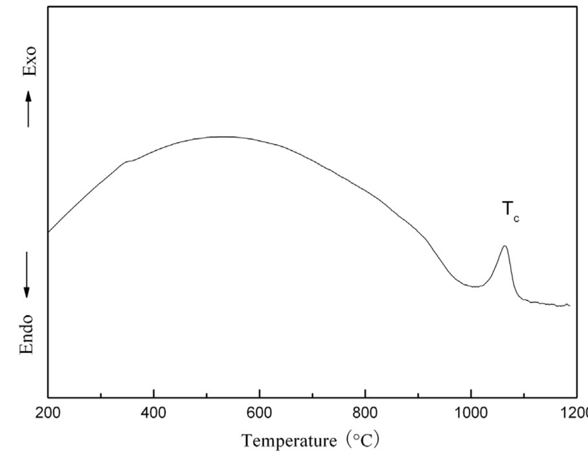

Fig. 1. DSC curve of the MAS glass containing 25% MgO, 5% La_2O_3, 25% Al_2O_3 and 50% SiO_2 (mol%).

3. Results and discussion

Fig. 1 shows the DSC curve of the MAS glass powders containing 20 mol% MgO, 5 mol% La_2O_3, 25 mol% Al_2O_3 and 50 mol% SiO_2. The exothermic peak observed at a temperature of about 1060 °C is related to the crystallization of the magnesium aluminosilicate glass, which is confirmed by XRD analysis. Suitable glass matrix for glass-AlN binary composites is very fundamental as it can determine the densification behavior and final physical and chemical properties of the composites [19]. The glass used in our work can obtain crystallized phases cordierite and $La_2Si_2O_7$. Moreover, MgO and La_2O_3 are considered as valid sintering aids to AlN or Si_3N_4 ceramics. The crystallization of the glass phase is deemed to be helpful for obtaining larger phonon mean free paths and therefore facilitating the thermal conductivity [20].

Fig. 2 shows thermal conductivity of MAS/AlN binary composites as a function of MAS glass content (wt%). As can be seen from Fig. 2, the thermal conductivity of the composite samples increases gradually with increasing of MAS content. When the MAS content achieves 40 wt%, the thermal conductivity of the composites reaches as high as of 14.58 W/(m K). When the MAS content is more than 40 wt%, the thermal conductivity of the MAS/AlN binary composites decreases instead. This may be related to that MAS glass acts as the second phase,

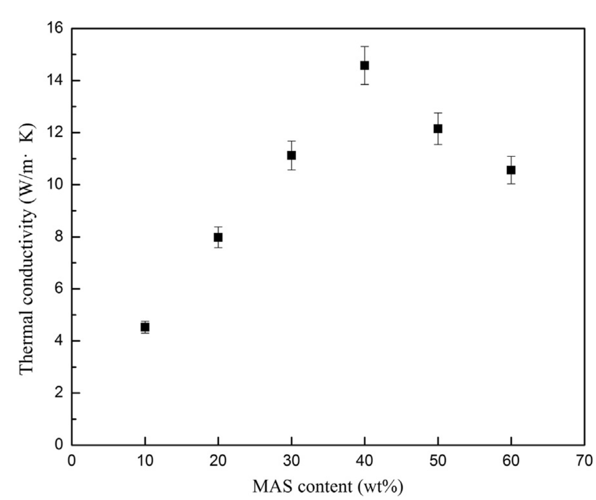

Fig. 2. The thermal conductivity of AlN/MAS binary composites as a function of MAS glass content (wt%).

Download English Version:

https://daneshyari.com/en/article/7889123

Download Persian Version:

https://daneshyari.com/article/7889123

Daneshyari.com