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Joining of Si_3N_4/Si_3N_4 with in-situ formed Si-Al-Yb oxynitride glasses interlayer

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ABSTRACT

Si_3N_4 ceramic was successfully joined to itself with in-situ formed Yb-Si-Al oxynitride glass interlayer. The joints were composed of three parts: (I) Si_3N_4 matrix, (II) oxynitride glass interlayer in which hexagonal or fine elongated β-sialon grains and a few ball-like β-Si_3N_4 grains exist, and (III) diffusion zone in Si_3N_4 matrix containing a thin dark layer and a ~ 25 μm thick bright layer. The seam owned similar microstructure to matrix and was inosculated with the matrix as a whole. The strength of the joint tended to increase with the increase of bonding temperature and reached the value of 225 MPa, when the joints were prepared at 1600 °C for 30 min under a pressure of 1.5 MPa. The high-temperature strength remained 94.7% and 75.2% of R.T. strength when the joints were tested at 1000 °C and 1200 °C, respectively. It may be contributed to the high softening temperature of the Yb-Si-Al oxynitride glass phase formed in the seam. Even suffered to the air exposure for 10 h at 1200 °C, the residual strength of the joints was still 143 MPa, attributed to the existence of YbAG phase.

1. Introduction

Si_3N_4 ceramic is one of the most promising engineering materials owing to its excellent over-all properties such as high mechanical properties, good thermal stability and resistance to wear and corrosion [1–3]. Many investigations have been carried out on the preparation and properties of silicon nitride ceramics. The existing sintering techniques [4–6] of fabricating silicon nitride ceramics contain reaction bonding sintering, spark plasma sintering and hot pressing sintering, etc. Although the sintering methods are various, the fabrication of large complex Si_3N_4 ceramic products is quite difficult due to their intrinsic brittleness. Hence, significant effort is being applied to other fabrication techniques, including joining.

A number of joining techniques have been applied to join Si_3N_4 ceramics. Lemus-Ruiz et al. [7] self-bonded Si_3N_4 at 1200–1600 °C under a pressure of 20 MPa using Nb-foil as the interlayer and Nb_5Si_3 and $NbSi_2$ were detected at the joining interface. However, high-pressure diffusion joining limits the size and shape of components. Active brazing, due to its convenient operating process and high room-temperature joining strength, is often used to join Si_3N_4 to itself or metals. Ag-Cu-Ti [8], Cu-Zn-Ti [9] and composite solders [10] were developed and used as active brazing filler, with which strong joints are ready to form. Nevertheless, the techniques mentioned above have a common characteristic that metals were used as the interlayer to realize the joining behavior. As a result, these joints cannot be used above 800 °C, which greatly limits their application.

A satisfactory method to join Si_3N_4 ceramics with oxide and oxynitride glass solders has been developed [11–16]. Y_2O_3, MgO, Al_2O_3, SiO_2, etc, are often used to form the glass phases, which have been also applied to the densification of bulk Si_3N_4 ceramics as sintering aids. Owing to the similarity in composition and properties of glass to the intergranular phase in Si_3N_4 matrix, glass adhesive bonding does benefit to decreasing the mismatch of physicochemical properties between metal filler and ceramics. But ceramics bonded with oxide glasses is still more likely to break at the interface because of the difference in thermal expansion coefficient [17]. Compared to the oxide glass, the oxynitride glass formed by adding Si_3N_4 powders owns superior mechanical properties, better resistance to thermal and mechanical shock, and lower thermal expansion coefficient [13,18,19]. Xie et al. [13] have joined Si_3N_4 to itself using Y-Si-Al-O-N glass adhesive with near eutectic composition, and the joint retains a high strength at 1000 °C. The study on Si-Al-RE-O-N (RE = rare-earth) glass by Becher et al. [20] indicated that the glass transition temperature (Tg value) was increased by substituting smaller RE ions, whereas the thermal expansion coefficient (α) was decreased. In addition, the oxidation resistance of Si-Al-RE oxynitride glass increased with the decrease in size of RE cations. And in recent papers [1,21], Yb_2O_3 has been studied for enhancing the physical properties of bulk Si_3N_4 due to the formation grain boundary phase (Yb-Si-Al-O-N glass). Because the radius of Yb^{3+} (0.086 nm) is almost the smallest among RE ions, the formation of Si-Al-Yb-O-N glass

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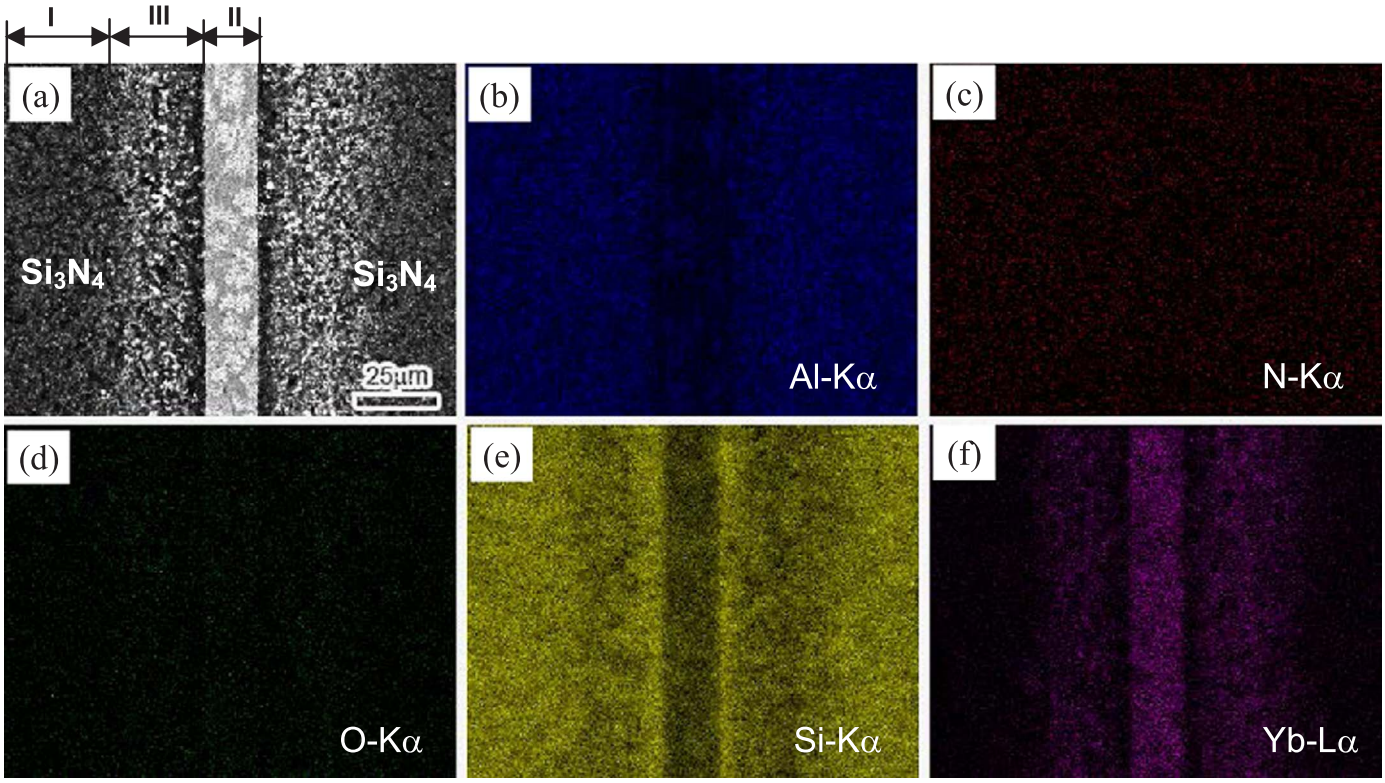

Fig. 1. BSE micrograph (a) and element maps of Al, N, O, Si and Yb (b–f) for the Si_3N_4 joint bonded at 1550 °C for 30 min under 1.5 MPa pressure.

interlayer is expected to improve the high-temperature properties of the joint. Hence, in our current work, Yb_2O_3 was selected and the mixtures combined with SiO_2, Al_2O_3 and Si_3N_4 powders were applied to bond Si_3N_4 to itself. The microstructure and properties of the joints were investigated and the high-temperature properties were also evaluated.

2. Experimental procedures

Hot-pressed Si_3N_4 ceramics (density, 3.2 g/cm^3, Shanghai Institute of Ceramics, China) were used as the bonded materials, in which there are no open porosities. The bending strength of Si_3N_4 at room temperature is 512 MPa. The Si_3N_4 ceramic was cut into bars with the dimension of 3 × 4 × 17 mm^3 and 3 × 4 × 3 mm^3 for property and microstructure samples, respectively, by diamond cutting machine. The joining surfaces of the ceramic bars were coarsely ground on SiC sand papers and then polished using diamond paste of 0.5 μm. Before joining, the ceramic bars were cleaned ultrasonically in acetone and dried by air blowing.

Solder was designed using powder mixtures of the composition (in eq.%) of 53Si-18Al-29Yb-70O-30N, and extra 15 wt% Si_3N_4 was added to form ceramic phases in glass. Starting powders used in this study are α-Si_3N_4 (E10 grade, UBE Industries Ltd., Tokyo, Japan), Al_2O_3 (grade A16SG, Alcoa, Pittsburgh, PA), Yb_2O_3 (99.99%, grade fine, H.C. Starck Ltd., Berlin, Germany) and SiO_2 (grade fine, China). The mixed powders were ball-milled in absolute alcohol for 8 h with Si_3N_4 balls as the mixing media, and then dried at 40 °C in a rotary evaporator and sieved. Powder mixtures were compacted and placed between two Si_3N_4 bars by a kind of organic glue. The assemblies were put into a graphic jig. During the bonding process, a pressure of 0.6 MPa or 1.5 MPa was put on the joining bars and a 0.09 MPa N_2 pressure was kept. The bonding temperature ranged from 1450 to 1600 °C. At the beginning of bonding process, the temperature was increased to 300 °C at a heating rate of 30 °C/min and held there for 10 min to make the organic glue volatilize in order to clean the brazing surfaces. The temperature continued to rise to 1000 °C at a heating rate of 30 °C/min and then to joining temperatures at 20 °C/min and held there for 30 min. At last, the samples were cooled down at a rate of 20 °C/min to 1000 °C and then cooled down spontaneously in the furnace.

The phase assemblages in the joint were determined by X-ray diffraction (XRD, Philips X'Pert, the Netherlands). The microstructure of the joints was observed by scanning electron microscopy (SEM, Quanta 200FEG, FEI Co., USA) in backscattered electron mode. The fine microstructure of the interface was also observed by Transition electron microscopy (TEM, TECNAI G2 F30, FEI Co., USA) fitted with Energy dispersive spectrometer (EDS). TEM specimen with 0.2 μm thickness and 5 μm length was prepared by focused ion beam (FIB, HELIOS NanoLab 600i, FEI Co., USA) technique.

The strength of the butt joints was evaluated by three-point bending test in air from room temperature to 1300 °C with a cross-head speed of 0.5 mm/min. The oxidation resistance of the joints was examined by holding 10 h at 1200 °C in air and then the 3-point bending strength was measured at room temperature. An average of at least three samples was used to determine the bending strength for each joining condition. The error was derived from the mean standard deviation. The fractural surfaces were observed by SEM.

3. Results and discussion

3.1. Typical microstructure of joint

Yb-Si-Al oxynitride glass interlayer was in-situ formed during the bonding process, which successfully promoted the Si_3N_4 ceramic joining. The typical microstructure and element maps of Si_3N_4 joint were indicated in Fig. 1. According to the variety in contrast (Fig. 1(a)), it is clear that the joint consisted of three parts, e.g. (I) Si_3N_4 matrix, (II) oxynitride glass interlayer in which bright and gray phases existed, and (III) diffusion zone in Si_3N_4 matrix containing a thin dark layer close to interface and a ~ 25 μm thick bright layer far from interface. Observed from the element maps, Al mainly distributed in the Si_3N_4 matrix attributing to the Al_2O_3 sintering additive, but it presented nonuniform in