Journal of the European Ceramic Society 37 (2017) 4015–4023

Contents lists available at www.sciencedirect.com

Journal of the European Ceramic Society

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

Microstructure evolution during air bonding of Al_2O_3 to Al_2O_3 joints using bismuth–borate–zinc glass

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ARTICLE INFO

Article history:
Received 30 November 2016
Received in revised form 28 March 2017
Accepted 2 April 2017
Available online 30 April 2017

Keywords:
Sealing
Bismuth-borate-zinc glass
Al_2O_3
$Bi_{24}B_2O_{39}$
$ZnAl_2O_4$

ABSTRACT

Alumina ceramics with 95 wt.% purity were sealed together using a bismuth based glass, $40Bi_2O_3–40B_2O_3–20ZnO$ (mol.%). The wettability of the glass on the Al_2O_3 substrate was investigated. The results showed a contact angle of ≤36.5° was achieved when the temperature was ≥630 °C. Subsequently, sealing cycles were performed at temperatures of 520–700 °C for 30 min. The dependence of microstructure evolution of the joints on temperature was investigated. $Bi_{24}B_2O_{39}$ was detected to be the product in the joints sealed at 530–580 °C, while $ZnAl_2O_4$ was identified to be the main product when sealing at temperature of ≥650 °C due to the reaction between the Al_2O_3 substrate and ZnO from the glass. The influence of dwelling time at 700 °C on microstructure evolution of the joints was also studied. The results showed that the size of $ZnAl_2O_4$ increased with increasing holding time.

1. Introduction

Attributing to excellent mechanical strength, high hardness, high melting point, high resistance to wear and corrosion, and high electrical insulation, alumina ceramics are becoming more and more important in various applications as a structural, engineering, and an electrical material [1–3]. Alumina ceramics are often joined to themselves to form assemblies for these applications [4–7]. Joining is a reliable technology to economically manufacture large or complex-shaped components that exhibit reliable performance through controlled integration of smaller, less complex, more easily manufactured, simple-shaped ceramic parts to expand its practical uses [8–11]. Additional, joining can also provide an avenue for reparation of damaged structures through the replacement of defective component [11].

Of the established and emerging technologies for joining alumina ceramics, a wide variety of bonding methods such as active metal brazing [12], Mo-Mn metallization process [13], and glass joining [14] were employed. The Mo-Mn metallization process, it involves two important stages: (i) metallization of Al_2O_3 ceramics, and (ii) joining the metallized ceramic using a suitable brazing alloy [4,15]. However, this technique is complex and expensive, and always involves high temperature processes and a high cost. Active brazing is the most widely used in industry for joining alumina ceramics. Ti or another reactive metal (e.g., Zr, Nb, Cr or Y) is added to metal alloys to promote wetting, braze spreading and interfacial bonding [16]. Generally, brazing of alumina ceramics to themselves or to metallic substrates is often performed using Cu or AgCu alloys containing a few percent of Ti [8,17]. Ti dissolves in these alloys and reacts with alumina forming Ti oxides or Cu—Ti—O compounds, which can bond to Al_2O_3 ceramics strongly [18]. However, the brazing process is always performed at a high temperature over 800 °C as well as in an ultrahigh vacuum (UHV). Residual stresses were induced upon cooling after joining at a high temperature, which would cause a substantial amount of internal thermal stress to build up in the joint during brazing and crack formation in the joints during the service [19,20]. Additionally, the extent of degradation or reaction often increases with increasing temperature [21,22]. Materials degradation during the joining and interfacial reactions can limit the properties and reliability of joined assemblies [23]. Moreover, a higher joining temperature, as well as an ultrahigh vacuum brings a high cost too.

Among various bonding techniques for ceramic materials, glass joining is a cost-effective and relatively simple method of air bonding various ceramics [24–28,29]. As most glasses pertain to the ceramics category since the bonds are either ionic or covalent [30]. The related properties of a glass fit well to that of the ceramics

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http://dx.doi.org/10.1016/j.jeurceramsoc.2017.04.002
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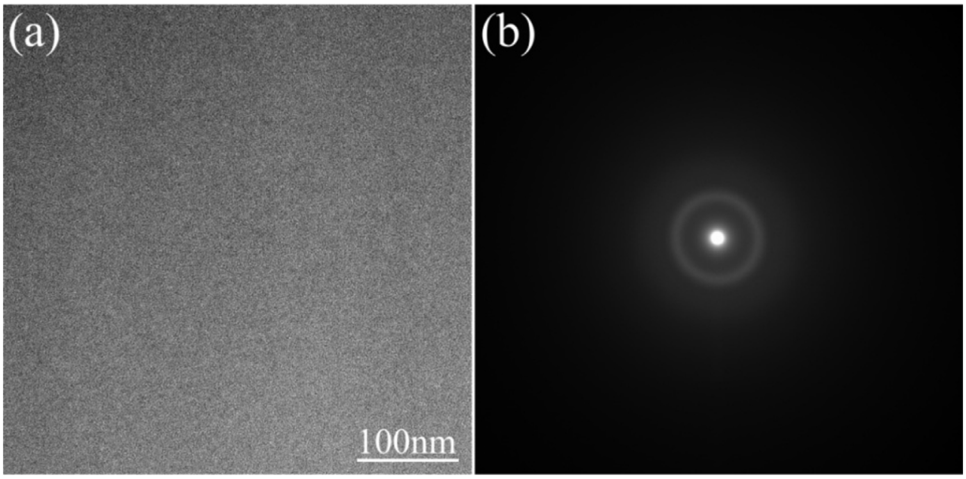

Fig. 1. (a) TEM micrograph and (b) corresponding selected area electron diffraction pattern (SAEDP) of the $40Bi_2O_3$-$40B_2O_3$-20ZnO glass.

group [30]. Therefore, glasses have better chemical compatibility with ceramics compared with metal solders [31].

The use of glass solders is desirable to join alumina in air. Several glass systems [14,32–34] have been employed to join alumina ceramics. However, their bonding temperatures ranged from 1200 to 1600 °C, which may cause irreversible changes in the structure of alumina ceramics.

In the present study, we intended to use bismuth-borate-zinc (Bi_2O_3–B_2O_3–ZnO) glass, which is a type of glass with a low melting temperature, as a joining media, to join 95 wt.% purity alumina ceramics in air. The wettability of the glass on the Al_2O_3 substrate was investigated. The microstructures of the Al_2O_3/Al_2O_3 joints were characterized, and the effects of joining temperature and holding time on the microstructures of the Al_2O_3/Al_2O_3 joints were studied.

2. Experimental procedures

Glass in the ternary system of Bi_2O_3–B_2O_3–ZnO having the chemical composition of $40Bi_2O_3$–$40B_2O_3$–20ZnO (mol.%) was prepared using conventional melt-quenching method. Commercial powders of reagent grade Bi_2O_3, H_3BO_3 and ZnO (Aldrich Industrial Corporation, Shanghai, China) were mixed well in a roller-type mill and then melted in an alumina crucible at 1000 °C for 2 h in a muffle furnace (KSL1400, Hefei Kejing Materials Technology Co., Ltd.). To prevent the occurrence of crystallization, one part of the melt was quickly poured into cold deionized water to form yellowish glass particles with sizes of 1–2 mm in diameter. Subsequently, these glass particles were ball-milled with agate balls in an agate mortar for 10 h, then screened through a 400 mesh sieve. The glass powders with an average size of ≤ 10 μm were obtained then. The remaining part of the melt was poured into a 250 °C pre-heated stainless steel mould to prepare glass bars for the coefficient of thermal expansion (CTE) measurement (φ 5 mm × 25 mm).

Commercial alumina ceramics with 95 wt.% purity ($95Al_2O_3$) were used as the base ceramic to be joined in our study. Al_2O_3 plates with size of 6 mm × 6 mm × 3 mm were used for microstructure investigation. The plates were polished with a 1 μm diamond paste, and then ultrasonically cleaned in an acetone bath for 30 min prior to joining.

Glass powders and terpi-neol were well-mixed at a weight ratio of 1:2 to obtain homogenous glass slurry. The slurry was coated onto surfaces of Al_2O_3 plates to form an Al_2O_3 plate/Bi_2O_3–B_2O_3–ZnO glass/Al_2O_3 plate sandwich structure. The Al_2O_3 ceramic substrates were ground flat before the slurry was applied. Sealing process was performed in a muffle furnace in air: (i) the samples were first heated up to 300 °C at the rate of 3 °C/min and held for 30 min to burn out the organic components; (ii) the specimens were continually heated up to joining temperature at a rate of 5 °C/min and held at that temperature for 30 min; (iii) the furnace was cooled down to 400 °C at a rate of 5 °C/min, then was left naturally cooling to room temperature.

The wetting experiment was carried out in air at a heating rate of 5 °C/min in a high-temperature microscope (HTM) from room temperature to 750 °C. The glass pellet (φ 6 mm × 3 mm) adopted for the wetting experiment was produced by pouring the melt into a 250 °C preheated stainless mold. An alumina plate (20 mm × 20 mm × 2 mm) was used as the substrate. A computerized image analysis system automatically recorded and analyzed the sample geometry during heating. The contact angle at a certain temperature was confirmed by averaging the testing values of repeated measurement of the left and right contact angles using measuring software.

The nature of the glass as well as the products or phases in the joints was confirmed by X-ray diffraction (XRD, Philips X'pert, Holland) with CuKα (wavelength = 1.5418 Å). The glass transition temperature (T_g), the crystallization temperature (T_c) and the melting temperature (T_m) were determined using a differential scanning calorimeter (DSC, STA449F3, Netzsch, Germany) at a heating rate of 5 °C/min in the range of 20–900 °C in air. A dilatometer (DIL 402C, Netzsch, Germany) was employed to evaluate the coefficients of thermal expansion (CTEs) of the glass and $95Al_2O_3$. Besides, the glass transition temperature (T_g) and the softening temperature (T_d) of the glass could be also obtained from the CTE curve.

The microstructures of the Al_2O_3/Al_2O_3 joints were examined by scanning electron microscope (SEM, Helios Nanolab 600i, FEI Co., Ltd, USA) equipped with an energy dispersive spectrometer (EDS). The transmission electron microscopy (TEM, Talos F200X, FEI Co., Ltd, USA) was adopted to further characterize and identify the products or phases in the joints. TEM specimens were prepared using a focus ion beam (FIB) micro sampling method with Helios Nanolab 600i (FEI Co., Ltd, USA).

3. Results and discussion

3.1. Characterization of prepared Bi_2O_3–B_2O_3–ZnO glass

Fig. 1 shows TEM micrograph and corresponding selected area electron diffraction pattern (SAEDP) obtained from prepared glass. The results presented fully amorphous nature of the glass.