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Fabrication and characterization of cordierite-based glass-ceramic adhesives for bonding solar heat transmission pipelines

Xiaoyang Xu[a,*], Xiaohong Xu[b], Jianfeng Wu[b], Xinbin Lao[c]

[a] *Ceramic Intellectual Property Information Center, Jingdezhen Ceramic Institute, Jingdezhen 333000, PR China*
[b] *State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, PR China*
[c] *National Engineering Research Center for Domestic & Building Ceramics, Jingdezhen Ceramic Institute, Jingdezhen 333000, PR China*

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ABSTRACT

Cordierite-based glass–ceramic adhesives for bonding solar heat transmission ceramic pipelines were fabricated successfully using Suzhou kaolin, talc and commercial alumina as the main raw materials, TiO_2 as nucleation agent, waste glass powder and frit 242 as high-temperature fluxes. Bonding behavior, phase transformation, microstructure, thermal shock resistance and thermal cycling properties of the adhesives were investigated, and bonding mechanism was studied. The results indicated that adhesives B4 (composed of 60 wt% cordierite glass–ceramic and 40 wt% frit 242) after heat treatment (nucleated at 800 °C for 2 h and then crystallized at 950 °C for 2 h) exhibited the optimal performances with a shear strength of 10.26 MPa, which exceeded the industrial standard (JC/T 547-2005) of 1 MPa. The analysis of bonding mechanism showed that the glass of adhesive that filled up the pores in matrix surface and penetrated inside contributed to the high shear strength. Adhesives B4 had good thermal shock resistance and thermal cycling properties, as the shear strength of samples after 30 thermal shock cycles (1100 °C-room temperature, air cooling) was 8.51 MPa, which increased to 26.93 MPa with an increase rate of 162.48% after 100 thermal cycles (200–1100 °C). During thermal shocks or thermal cycles, cordierite particles crystallized at the interfaces to form a pinning-in effect, contributing to a higher shear strength.

1. Introduction

Owing to the high density, excellent mechanical property, creep and thermal shock resistance, ceramic materials, such as andalusite [1], mullite-cordierite [2], corundum-mullite-spinel [3], corundum-spinel-ZrO_2 composite and cordierite-mullite- corundum [4], are suitable for producing heat transmission pipeline in solar thermal power generation. However, due to the nature of low ductility and brittleness, the productions of ceramic pipelines with large volume and complex shapes are so difficult and costly that the complex preparation procedures and a large amount of energy are required [5,6]. Therefore, it is quite prevalent to produce ceramic pipelines with medium or small sizes, which then are bonded with each other using high-temperature adhesives, aiming at simplifying the transportations of ceramic pipelines. Accordingly, advanced bonding technology for ceramics becomes a key factor in the industrial applications of the ceramic pipeline materials.

Nowadays, many bonding technologies such as mechanical connection, diffusion bonding, green-state joining, active metallic brazing and plastic deformation joining are applied for ceramic bonding [7]. Drawbacks of these bonding technologies as low operating temperature, mismatch of thermal expansion coefficients, stress concentration, high porosity, accurate and complex treatment processes and high cost have highly limited their feasibility for ceramic bonding. Conzone et al. [8] obtained the tense joints of $MoSi_2$ to 316L stainless steel by active metallic brazing, while the operation temperature was limited due to the Cu/Si phase with a relatively low melting temperature (852 °C) that was produced at the $MoSi_2$/Nb interface. Comparing with the aforementioned techniques, the adhesive bonding technique is more advantageous, which allows for a more homogeneous stress distribution in the bonding area and maintains the nature of ceramic materials [9–11]. Inorganic and organic adhesives are two kinds of common adhesives for bonding structural components. Although organic adhesives possess outstanding bonding behavior, their low operating temperatures (usually under 400 °C) have restricted their applications in bonding ceramic heat transmission pipelines due to the inevitable pyrolysis reactions occurring at high temperatures [12]. Besides, some heat-resistant adhesives such as polysiloxane and phenol-formaldehyde resin also need to be heat-treated at a reducing atmosphere for bonding SiC, Al_2O_3 ceramics to obtain satisfactory shear strength [13].

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[*] Corresponding author.
E-mail address: zalexu1991@126.com (X. Xu).

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Since the inorganic ones are capable of enduring the high temperature working conditions, they show great potential to bond ceramic heat transmission pipelines which has lots of advantages as high-temperature resistance (> 1000 °C), good durability and satisfactory shear strength, etc. [14]. However, many inorganic adhesives such as phosphate and silicate also suffer from brittleness, poor water resistance [14,15]. Therefore, it is necessary to develop new inorganic adhesives with better comprehensive properties for overcoming the drawbacks mentioned above and glasses or glass-ceramic for ceramic bonding are under great consideration. Xie et al. [16,17] had fabricated Y-Si-Al-O-N oxynitride glass adhesive to bond Si_3N_4 ceramics, and the similarity in the microstructure between the joined layer and the bulk ceramic resulted in high shear strength and good bonding behavior. $CaO-Al_2O_3-SiO_2$ glass-ceramic adhesive for bonding electro-ceramics was prepared by Wu et al. [18], and the optimal samples possess a bending strength of 88.6 MPa and a high density after suitable heat-treatment process with heating rate as 5 °C /min and a holding time as 1 h at 950 °C. However, the thermal shock resistance of glass adhesives had been proven to be poor due to their high thermal expansion coefficients, which would also weaken the thermal cycling properties.

In this study, cordierite glass-ceramic with a wide range of thermal expansion coefficient, chemical stability, thermal cycling properties and thermal shock resistance were prepared as a kind of inorganic adhesive from Suzhou kaolin, talc, commercial alumina and different high-temperature fluxes using TiO_2 as nucleation agent, which could be applied for bonding heat transmission ceramic pipelines. The bonding effect and mechanism of cordierite glass-ceramic were investigated, while the mechanical property, thermal shock resistance and thermal cycling properties of the as-prepared high-temperature adhesive were studied.

2. Experimental

2.1. Starting materials

Suzhou kaolin (~58 μm, China Kaolin Clay Co., Jiangsu, China), talc (~58 μm, Guilin Talc Development Co., Ltd., China), commercial alumina (~58 μm, Dongda High Temperature Energy-saving Material Co., China) were used as starting materials to form cordierite crystallite. TiO_2 powder with a purity of 99.9% was chosen as nucleation agent (supplied by Sinopharm Chemical Reagent Co., Ltd., China). Waste glass powder (~58 μm, the softening temperature from 900 °C to 1000 °C, the waste from Wuhan Ghangli Glass Co., Ltd., China) and frit 242 (~58 μm, the softening temperature from 700 °C to 900 °C, Zibo Guosheng Glaze Co., Ltd., China) were used as high-temperature fluxes to decrease the melting point of the cordierite glass-ceramic. The chemical compositions of the starting materials were listed in Table 1. In order to obtain a high content of α-cordierite, the designed formula compositions of cordierite glass-ceramic were 15 wt% MgO, 30 wt% Al_2O_3, 55 wt% SiO_2 and 1 wt% TiO_2 in addition, namely formula B0 listed in Table 2. The other formulae were designed containing two kinds of fluxes, of which formulae B1 and B2 contained 30 wt%, 40 wt% waste glass powder, respectively, and formulae B3 and B4 were distinguished by 30 wt%, 40 wt% frit 242.

2.2. Sample preparation

The starting materials of Suzhou kaolin, talc, commercial alumina and TiO_2 summarized in Table 2 were mixed and ball-milled for 1 h, and then melted at 1500 °C for 2 h in a corundum crucible in an electric furnace with a heating rate of 5 °C/min. By quenching the melts of the mixed raw materials, $MgO-Al_2O_3-SiO_2$ glass was obtained, which was ball-milled for 2 h, sieved by 250-mesh sieve and mixed with waste glass powder or frit 242 powder.

Rectangle cordierite-mullite-corundum composite ceramics of 40×25×8 mm (length×width×height) used for bonding experiments were fabricated by solid-state sintering with a density of 3.16 g cm^{-3}, which had been reported in our previous study [19]. The room-temperature bending strength of cordierite-mullite-corundum samples was 123.48 MPa. After the samples were polished, they were ultrasonically cleaned in water-free ethanol and dried at 80 °C for 2 h, and a slurry of the powdered adhesive added with carboxymethylcellulose solution (1 vol%) was coated on the 40×25 surface of the rectangle composite ceramics. In order to control the thickness of adhesive, 0.2 MPa pressure was applied to bond two rectangle samples. According to the TG-DSC results shown in Fig. 1, the first and second holding temperatures for nucleation and crystallization of cordierite were determined as 800 °C and 950 °C, respectively, whilst the holding time at each temperature was 2 h with a heating rate of 3 °C/min..

2.3. Characterization

The shear strength of bonded samples was measured by Computer Control Electronic Universal Test Machine (Shenzhen Reger Instrument Co., Ltd., China) at a speed of 0.5 mm/min. The testing method performed was based on Ref. [6,20,21]. The thermal expansion coefficients were measured by WTC-1 quartz thermal expansion analyzer (Wuhan University of Technology, China). X-ray diffraction (XRD) patterns were collected using a D/MAX-IIIA diffractometer (Rigaku Corporation, Japan) equipped with a copper target, scintillation detector and graphite monochromator with Cu Ka (λ=1.54 Å) radiation. SEM images were recorded in a JEOL JSM-5615LV (JEOL Ltd., Japan) to investigate the microstructure of the samples. The morphology and semi-quantitative analysis of fractured surfaces of the samples were performed by field emission scanning electron microscope (FE-SEM) (Ultra Plus-43-13, Zeiss, Germany) with an energy dispersive spectroscopy (EDS) (X-Max 50, Oxford, Britain).

3. Results and discussion

3.1. Shear strength of the adhesives

Adhesives for bonding solar heat transmission ceramic pipelines are required to possess a high mechanical property. Therefore, the shear strength of samples bonded with adhesives of series B after heat treatment (nucleating at 800 °C for 2 h and then crystallizing at 950 °C for 2 h) was tested and listed in Table 3. It is seen that the shear strength of adhesives has surpassed the industrial standard (JC/T 547–2005) of 1 MPa. In terms of shear strength, adhesives B4 (using 40 wt% frit 242 as flux) show better bonding behavior. As indicated by Table 2, the higher flux content, the higher shear strength. This shall be attributed to the good wettability of flux at high temperature and can be verified by the SEM analysis.

3.2. XRD analysis of adhesives

Fig. 2(a) and (b) are the XRD patterns of adhesives B2 and B4 after heat treatment, respectively, both of which demonstrate the formation of cordierite obtained from the crystallization of $MgO-Al_2O_3-SiO_2$ glass in the mixture of Suzhou kaolin, talc, commercial alumina and TiO_2. In contrast to the testing results of thermal shock resistance, the formation of cordierite with low thermal expansion coefficient will improve the thermal volume stability and endow the adhesives with good thermal shock resistance. As seen in Fig. 2(a), with the addition of waste glass powder, anorthite, forsterite, α-cristobalite and α-tridymite are also generated, indicating that $MgO-Al_2O_3-SiO_2$ glass after heat-treatment will not transform to cordierite phase completely. The MgO residual reacts with SiO_2 to form forsterite, which is illustrated by Eq. (1). The generation of anorthite is attributed to the reaction of CaO, SiO_2 and Al_2O_3 in waste glass and $MgO-Al_2O_3-SiO_2$ glass (as shown in Eq. (2)). Moreover, the SiO_2 residual can transform into α-cristobalite and α-tridymite at high temperature. In Fig. 2(b), apart from cordier-