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Effects of interlayer composition on joining of 25%$CePO_4/ZrO_2$–ZrO_2 ceramics on green state

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Abstract

Green bodies of 25%$CePO_4/ZrO_2$ and ZrO_2 ceramics were joined without applied pressure by using mixed powders slurries composed of $CePO_4$and ZrO_2. The effects of $CePO_4/(CePO_4+ZrO_2)$ ratio of the interlayer on the bond strength and microstructure of the joints were investigated. Maximum bond strength of 414 MPa was obtained by joining with an adhesive with the ratio of 0.5. Under the experimental conditions, the grain size of the particles grown in the joint was smaller than those in joined ceramics. The microstructure of the joint was more homogeneous than that of the matrix.

Keywords: A. Joining; B. Interface; Green body; Slurries

1. Introduction

The methods in ceramic joining include active metal brazing [1,2], diffusion bonding [3], pyrolysis of pre-ceramic polymers [4,5], combustion reaction [6], and joining by microwave heating [7–9]. At present, two promising methods for joining advanced ceramics are brazing and diffusion bonding [10]. The filler materials can be metals, alloys or glasses [11]. However, the strength of the joints at elevated temperature is rather low because of the relatively low softening temperature of the most commonly used alloy interlayer materials. Materials joined with pure glass are more likely to crack at the joint due to the low resistance to crack propagation of the layer of glass [12]. So, it is very important to select suitable adhesives for obtaining ceramic joints with high strength at elevated temperatures.

Green body joining is commonly used in the clay-based ceramic industry to produce complex parts. The super plasticity induced by alkali ions in layered structures renders green body joining of clays relatively easy [13]. Advanced ceramics, such as ZrO_2, do not exhibit plasticity, and, hence, the green state joining of ZrO_2 has been overlooked as a method of fabricating complex shapes. However, some recent research shows that, by dispersing powders in a carrier medium, the unique behavior of clay-based ceramics can be approximated. The work of Zheng et al. [13], on joining of SiC using polymer precursors has indicated that green state joining is potentially a viable method. The green state joining offers several advantages over joining of dense ceramics. First, shape forming is easier in the green state with a suitable binder. Second, particle rearrangement at the joint may be achieved by capillary or external pressure, which may lead to a joint that has a similar microstructure as the bulk material. Third, mechanical properties of the joint may be improved because of small differential shrinkage and absence of impurity phases. Although there have been some successful experiments in the green state joining of ceramics, they were limited in the area of joining similar carbide and nitride ceramics. Therefore, joining similar, especially dissimilar, oxide ceramics is truly challenging but necessary.

The main purpose of this work is to develop adhesives in the $CePO_4$–ZrO_2 system for joining $CePO_4$ and ZrO_2 ceramics in the green state. The interlayer material developed possesses a thermal expansion coefficient closely matched to

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the matrix materials and the joints formed have a microstructure similar to that of the ceramics joined. Experimental results of joining $CePO_4$ and ZrO_2 ceramics by using three kinds of adhesives with different $CePO_4/(CePO_4+ZrO_2)$ ratios are described.

2. Experimental procedures

2.1. Materials

Commercially obtained ZrO_2 (Baotou Insitutute of Rare Earths, China) and $CePO_4$ (Farmeiya Advanced Materials Co. Ltd., China) powders were used. The average particle sizes of ZrO_2 and CePO4 powders were 0.055 μm and 0.06 μm, respectively, which were measured by Master Sizer Laser Analyzer 2000. The components were mixed and ball milled with ZrO_2 balls for 48 h to assure homogenous distribution of additives, using polycarbosilane (PCS, molecular weight of 1580, Tianjin Guoheng Co. Ltd., China) as a binder and polyethylene glycol (PEG, molecular weight of 1000, Tianjin Guoheng Co. Ltd., China) as a dispersant. The mixture was dried at 60 °C after ball milling and then sieved through a 100-mesh screen. Green compacts were obtained by uniaxially pressing 20 g of powder at 100 kPa in a 30 mm diameter stainless-steel die and then CIPing them at 200 MPa. The compacts formed had 43–52% of the theoretical density of materials. The adhesives with different $CePO_4/(CePO_4+ZrO_2)$ ratios are listed in Table 1.

2.2. Joining method

Before joining, the paste was de-gased for about 10 min. About 0.2 ml of paste was applied to each of the joined surfaces using a spatula. The compacts were joined after 5 min. Only slight hand pressure was needed to squeeze out the excess paste and then remove the additional slurry around the joint. The joined samples to be sintered were mended using ceramic knives and dried in the room temperature.

2.3. Sintering process

The samples were sintered at 1450 °C for 120 min. The sintered density of the joined sample was measured using the Archimedes' method. The final densities of the samples were measured to be in the range of 93–94% of the theoretical density of ZrO_2.

Table 1
Chemical composition of adhesives (wt.%)

	Powder [$CePO_4/(CePO_4+ZrO_2)$ ratio]	Binder	Dispersant	Water
S1	80 (0.25)	2	3	15
S2	80 (0.5)	3	4	13
S3	80 (0.75)	3	3	14

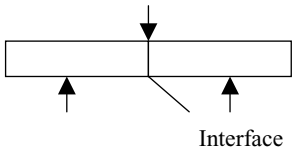


Fig. 1. Three-point bending test to evaluate the strength of the interface.

2.4. Mechanical tests and microscopy

The joined plates were cut perpendicularly to the joint interfaces to prepare bending bars (25 mm × 5 mm × 2.5 mm) for evaluating the bond strength. The joining strength of the joint was determined by a 3-point bend test and was calculated as follows:

$$\sigma_f = \frac{3PL}{2bh^2} \text{ (MPa)}$$

where L, b, and h are the sample length, thickness, and width, respectively. P is the load applied when fracture occurs. The loading situation in shown in Fig. 1. Thirty-six test bars (12 for one kind of samples using a specific adhesive) were prepared to obtain the average bond strength of joints using three different adhesives, respectively. The microstructures of the joint were investigated by scanning electron microscope (SEM).

3. Results and discussion

3.1. Mechanical property

Bond strengths as a function of $CePO_4/(CePO_4+ZrO_2)$ ratios are plotted in Fig. 2. As shown in the figure, the joint using adhesive S3 exhibits the lowest bond strength with an average value of 333 MPa. A maximum strength of 414 MPa is obtained as the ratio reaches 0.5 in the adhesive S2. The joint formed by using adhesive S1 demonstrates a bond strength of 365 MPa. Normally, the bond strength was mainly determined by two factors—residual stress in the interface and the mechanical strength of the region near the joint. Residual stresses produced in the interfaces have their origins in the coefficients of thermal expansion (CTE) mismatch of the bonded materials.

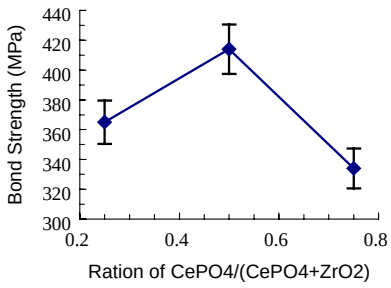


Fig. 2. Effects of $CePO_4/(CePO_4+ZrO_2)$ ratio of the adhesive on bond strength.

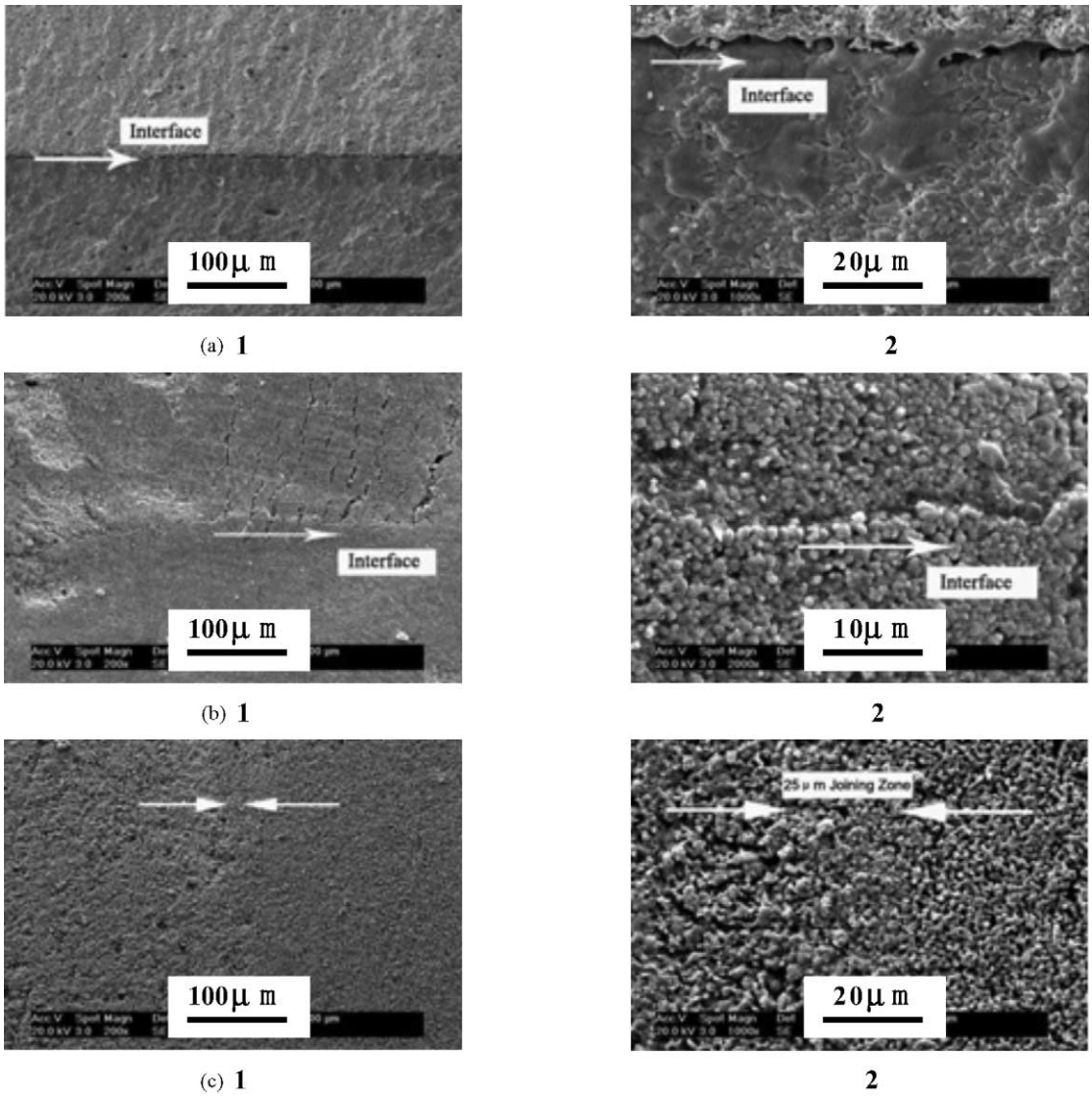
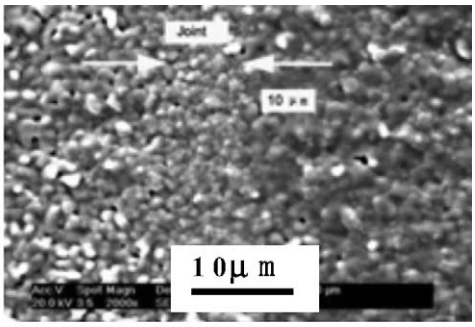


Fig. 3. Interfaces formed by using different adhesives at 1450 °C for 120 min: (a) adhesive S3; (b) adhesive S1; (c) adhesive S2.

Fig. 3 shows us the results of the interfaces formed by using different adhesives. It can clearly be seen from Fig. 3a that almost no joining zones formed because of the great difference between the CTE of $CePO_4$ and ZrO_2. Also, with a high $CePO_4$ content (75%) in the adhesive, the mechanical property of the joint reduced greatly due to the low strength of $CePO_4$ ceramics. Therefore, high interfacial shear stresses can induce delamination; meanwhile; high residual tensile stresses can cause cracking, lower the bond strength and even keep the interface apart. In Fig. 3b, it was clear that the material couples were joined together, but the joint was not dense and had too many defects. In addition, there were many visible cracks in the $CePO_4$ matrix due to the fact that the residual stress formed by the CTE mismatch of the materials joined exceeded the strength of $CePO_4$ matrix. However, using adhesive S2 with $CePO_4/(CePO_4+ZrO_2)$ ratio of 0.5, the interface formed is comparatively good. The CTE of the adhesive was similar to those of both matrix materials, which weakened the effect of the CTE mismatch. Also, the comparable microstructure between the adhesive and the matrix assured the diffusion formed easily in the interface, which can eliminate the defects and pores in the joint and therefore caused a dense joining zone with homogenous particle distribution, and then increased the bond strength. As shown in Fig. 3c, there were no visible cracks, pores and defects in the joining zone. The interface was also very dense and smooth.

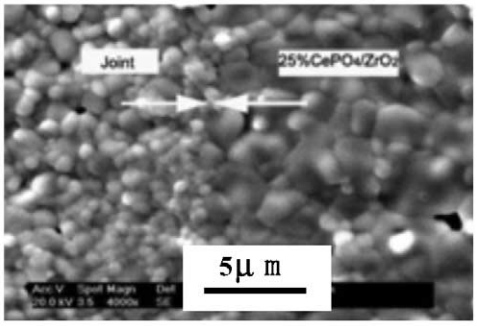
3.2. Joining mechanism and microstructural observation of the joints (adhesive S2)

To get some ideas of the joining mechanism and microstructures of the sound joint formed by using adhesive S2, the polished samples were treated at 1420 °C for about 1 h. the SEM micrographs of the joint were shown in Fig. 4. Note that the interfacial contact was excellent.



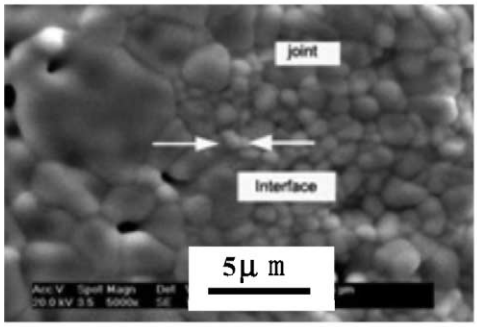
(a)

Joint between joined material couples



(b)

Interface between the joint and 25%$CePO_4/ZrO_2$



(c)

Matrix grains interlocking with grains of the joint

Fig. 4. SEM morphology of the interfacial regions of the samples S2 prepared at 1450 °C for 120 min.

In Fig. 4a, it is demonstrated that the joint thickness for the materials joined using adhesive S2 is about 10 μm. This bonding thickness is due to the fact that the ceramic powders in the adhesive penetrate into the ceramic matrix and vice versa. Because of the diffusion reaction between the adhesive and the matrix, a joint with uniform particle distribution and smaller grain size formed in the interfacial region, which join the dissimilar materials together. Fig. 4b and c show that not only grains within the joint and within the joined material couples clearly differ in size, but the transition between the two sets of grain sizes is gradual. As a green state joined process, the particles both in the adhesive and the matrix can grow together as the temperature increases. As a result, the particles in the interface closely inlayed and integrated each other in the joint. Due to all of these factors, a joint with almost perfect microstructure formed between the two materials, which had less deformation, cracks and pores than the matrix. Consequently, it is considered that these features of the joint may contribute to the formation of high-strength joints.

4. Conclusions

$25CePO_4/ZrO_2$ and ZrO_2 green state joining was accomplished by using mixed powder slurries composed of $CePO_4$ and ZrO_2 at 1450 °C for 120 min. Some conclusions can be drawn as follows:

1. Using gelatin-contained mixed powder slurries to join $25CePO4_4/ZrO_2$ and ZrO_2 on green state could reduce the shrinkage mismatch and, hence, eliminated the processing defects, which assure the formation of a sound joint.
2. With increasing $CePO_4$ powder in the adhesive, the bond strength of the joints increased. When the $CePO_4/(CePO_4+ZrO_2)$ ratio reached 0.5, a maximum bond strength 414 MPa was obtained, which was approximately 85% of that of unbonded materials. There was a drop in strength when further increasing the $CePO_4$ powder in the adhesive.
3. Microstructural analysis showed that the particle size in joint region was smaller than that of the matrix materials. The joint was denser and had less defects, cracks and pores than the matrix. Joining mechanism was determined to be diffusion reaction in the interface and the particle inlaying in the joining zone as the temperature increased.

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