

Anti-oxidation behavior of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites prepared by Sol-Gel

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Abstract: $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were fabricated by sol-gel method, and then the flexural property and oxidation behavior of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were discussed. The results showed that $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were fabricated by sol-gel method. The flexural strength of the composites was homology with that of $\text{SiO}_2/\text{SiO}_2$ composites, while the elastic modulus was higher than that of $\text{SiO}_2/\text{SiO}_2$ composites. After high-temperature oxidation, the fracture mechanism of the composites changed, which showed typical brittle/sudden fracture. Compare to $\text{SiO}_2/\text{SiO}_2$ composites, $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites showed higher flexural strength and higher elastic modulus.

1. Introduction

Owe to the excellent damage tolerance and thermal shock resistance, continuous fiber-reinforced ceramic matrix composites (CFCMC) had attracted considerable attention in high-temperature structure applications [1-4]. Compared to carbide ceramics, oxide ceramics showed better oxidation resistance [5].

Compared to polymer infiltration and pyrolysis (PIP), reaction sintering-hot pressing (RS-HP) and electro phoretic deposition (EPD), sol-gel method should be the most appropriate method to fabricate oxide/oxide composites[5-9], which showed the lowest densification temperature ($\leq 1000^\circ\text{C}$), appropriate fiber/matrix interfacial bonding and near-stoichiometric composition in matrix.

The high-purity quartz fibers, which had the excellent chemical stability and dielectric properties, were considered the most attractive candidate for fiber-reinforced in the silica ($\text{SiO}_2/\text{SiO}_2$) composites.

Several studies had been carried out in order to clarify the high-temperature mechanical behavior of $\text{SiO}_2/\text{SiO}_2$ composites [10]. However, the composites showed a worse high-temperature flexural strength [11].

For long time usage at higher temperature, the present paper was to research the $\text{ZrO}_2\text{-SiO}_2$ matrix, which had higher stability and melting temperature. The mechanical behavior and microstructure of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites after higher temperature oxidation were also discussed. will be part of the journals therefore we ask that authors follow the guidelines explained in this example, in order to achieve the highest quality possible.

2. Experiment

2.5D SiO_2 reinforcements, which were finished by Z-stitching the stacked fabrics with SiO_2 fibers yarn in a 5mm*5mm space, were provided by Feilihua Institute of Glass Fibers (P.R.China). The volume fraction of SiO_2 fibers was 41.0%. Diphasic ZrO_2 sol and SiO_2 sol were used as the

precursor of ZrO_2 and SiO_2 .

$\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were prepared by sol-gel method. First, the reinforcements were placed in a closed container, and then the container was evacuated to 0.1Pa. The high pure ZrO_2 sol (volume ratio 23%, average particle size 8 nm, PH8 obtained from Snow chemical S&T Co., China) was sucked into the container, and the pressure was maintained at 0.1Pa during the entire process for 8h. The composites were dried at 200°C for 4h, and then sintered at 1000°C for 1h to remove the coupling agent and bounded water. The process was repeated for ten cycles.

Secondly, the reinforcements were placed in a closed container, and then the container was evacuated to 0.1Pa. The high pure silica sol (volume ratio 25%, average particle size 10 nm, PH9 obtained from Snow chemical S&T Co., China) was sucked into the container, and the pressure was maintained at 0.1Pa during the entire process for 6h. The composites were dried at 100°C for 4 h and sintered at 800°C for 1h. The process was repeated for four cycles.

$\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were heated at 1200°C in air for 2h. The samples were weighed before and after oxidation by an electronic balance with a sensitivity of $\pm 0.001\text{g}$. Three-point bending tests were also used to evaluate the flexural strength of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites before and after oxidation. The span/height ratio was 15 and the speed was 0.5 mm/min.

The microstructure of the samples was observed with scanning electron microscopy (SEM, FEI Quanta-200).

3. Results and Discussion

3.1. Morphology and mechanical property

The surface and cross-section images of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites are showed in Figure 1. No obvious pores are existed. SiO_2 fibers are covered by $\text{ZrO}_2\text{-SiO}_2$ sol (Figure 1 Cross-section (a) and (b)), which show a better fiber/matrix interfacial bonding.

According the research [12], the better fiber/matrix interfacial bonding is necessary for higher performance. The hypothesis is confirmed by Fig.1, which show a lot of $\text{ZrO}_2\text{-SiO}_2$ matrix attached to SiO_2 fibers.

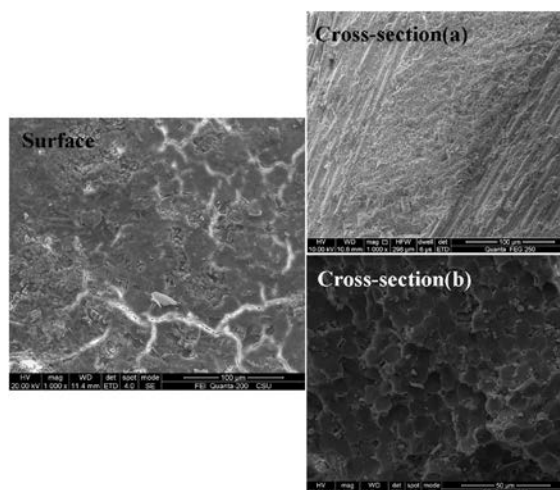


Figure1 The surface and cross-section of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites

The pores even inner fiber bundles are filled with dense $\text{ZrO}_2\text{-SiO}_2$ sol (Figure 1 Cross-section (b)). SiO_2 fibers show a uniformity form, which means no diffusion and reaction occurred, indicating good chemical and thermal compatibility. Thus, $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites can be fabricated efficiently by sol-gel method.

Figure 2 shows typical load-displacement curves of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites tested at room temperature. The flexural strength at room temperature is 100.8MPa, and the elastic modulus is 29.19GPa.

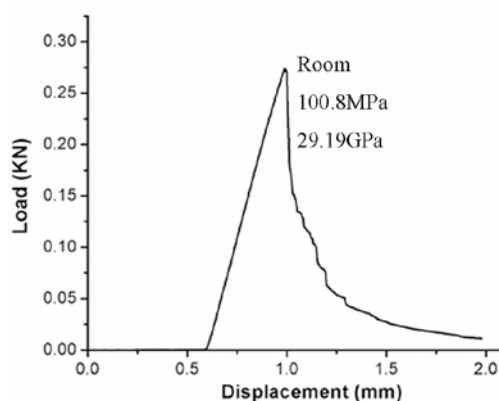


Figure 2 The load-displacement curves of the samples (Room temperature tests)

There are mainly fibers bundles pull-out when the load reaches at the maximum (Figure 3 (tested at room temperature)). The matrix fails when the stress reaches the maximum; the samples show a typical ductile/gradual fracture behavior.

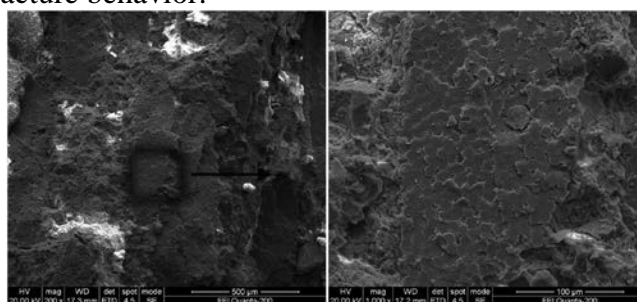


Figure 3 The cross-section of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites after three-point bending test (Room temperature)

At room temperature, the flexural strength of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ is homology with that of $\text{SiO}_2/\text{SiO}_2$, and the elastic modulus of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ is higher than that of $\text{SiO}_2/\text{SiO}_2$ [11, 13-14].

3.2. Anti-Oxidation behavior at 1200°C

The oxidation behavior of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites is tested at higher temperature (1200°C in air for 2h). The samples are weighed after oxidation, and then the flexural strength of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites is tested.

Due to the densification degree enhancement of $\text{ZrO}_2\text{-SiO}_2$ matrix during thermal atmosphere, $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites show better oxidation resistance in oxidation atmosphere [11, 15]. After oxidation at 1200°C, the mass loss of the samples reaches 0.8%.

Fig.4 shows typical load-displacement curves of the oxidation samples. The fracture mechanism is brittle mode. The flexural strength and elastic modulus of the samples after 1200°C oxidation are about 50.51MPa and 31.51GPa, respectively.

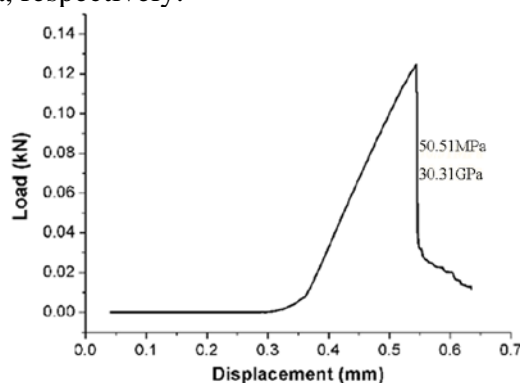


Figure 4 The load-displacement curves of the samples after oxidation

The fracture surfaces of the samples are examined by SEM (Figure 5). After high temperature

oxidation, the samples show a typical brittle fracture behavior, which is testified by the load – displacement curves (Figure 4).

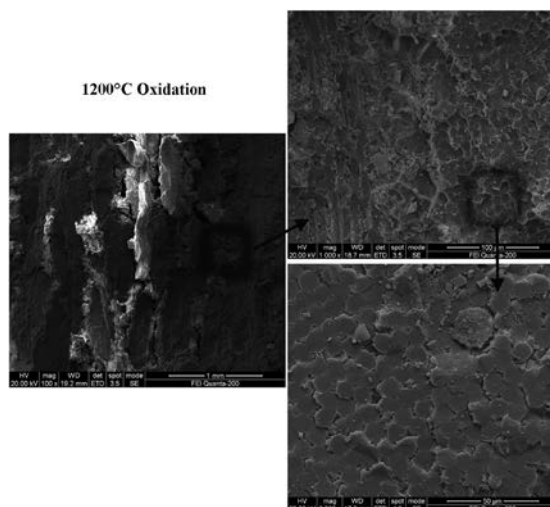


Figure 5 The cross-section of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ after 1200°C oxidation

Our earlier researches developed $\text{SiO}_2/\text{SiO}_2$ composites via sol-gel method and high-temperature mechanical properties of $\text{SiO}_2/\text{SiO}_2$ composites were discussed [11]. Moreover, for long time usage at higher temperature ($\geq 1200^\circ\text{C}$), it is necessary to develop a more protective glassy layer by using additives or replacements, which have higher stability and melting temperature.

The $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites by sol-gel method show higher high-temperature flexural property. The flexural strength and elastic modulus after 1200°C oxidation are about 50.51MPa and 31.51GPa. $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites show long time usage at higher temperature.

4. Conclusion

For long time usage at higher temperature ($\geq 1200^\circ\text{C}$), $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were fabricated by sol-gel method. The mechanical and fracture behavior of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites under higher temperature were discussed. The mechanisms were summarized as follows:

- 1) $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites were fabricated effectively by sol-gel method. At room temperature, the flexural strength of the composites is homology with that of $\text{SiO}_2/\text{SiO}_2$ composites, while the elastic modulus of the composites is higher than that of $\text{SiO}_2/\text{SiO}_2$ composites.
- 2) After higher temperature oxidation, the fracture mechanism of $\text{SiO}_2/\text{ZrO}_2\text{-SiO}_2$ composites changed to typical brittle/sudden fracture.

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