Preparation of Mullite Fiber Reinforced Mullite Composites

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Abstract. In this paper, mullite fiber reinforced mullite composites were prepared by stitched structural perform, the silica sol and alumina sol. In order to improve the compatibility of mullite fiber and silica sol matrix, interface coating was made by infiltration. The composites were prepared by the method of liquid phase infiltration, and the suitable ceramic heat treatment temperature was 750~900°C though testing the damage of fiber strength after heat treatment at different temperatures. Results showed that SiO₂-Al₂O₃-ZrO₂ interface coating could uniformly distribute on the surface of mullite fibers, and the appropriate interfacial bonding strength was obtained, which made a contribution to the high density mullite fiber reinforced mullite composite, whose density reached 1.75~1.8g/cm³, tensile strength, bending strength and compressive strength reached 48.7MPa, 65.4MPa and 82.7MPa, respectively. Dielectric constant was 3.2±0.1, Loss angle tangent was less than 8×10⁻³. So mullite fiber reinforced mullite composites became a new generation of high temperature wave-transparent material.

1. Introduction
Wave-transparent material is a kind of multifunctional material to protect aircraft communications for communication, telemetry, guidance and detonation in harsh environmental conditions, which has been widely used in spacecraft, rocket, missile and return type satellite. At present, silica fiber reinforced silica composites have been thought the best multi-functional wave-transparent material with high temperature resistance, resistance to erosion, load bearing and wave transmission.

However, when the silica fiber is used above 1200°C for a long time, the strength damage is severe, so the SiO₂/SiO₂ composites cannot be used for a long time above 1200°C. Mullite fibers is a kind of ceramic fiber with high temperature resistance, which can be used for a long time up to 1400°C. Now mullite fiber reinforced mullite composites have been studied as a new generation high-temperature wave-transparent material[1-3]. This paper studied mullite fiber reinforced mullite composites, which were prepared by the method of liquid phase infiltration. In this paper, interface coating on the fiber surface, preparation process and composite properties were studied.

2. Experiment

2.1 Sample preparation
The mullite fibers used in this paper were produced by Japan, which was composed of 72 wt% Al₂O₃ and 28wt% SiO₂. Stitched structural perform was made by mullite fibers, whose volume fraction was more than 40%. High purity Si-sol, in which silica content was 23.8% and sodium ion content was less...
than 60 ppm, and high purity silica powder were used to prepared silica slurry. The silica slurry solid content was set to 45wt% with a viscosity of 0.1Pa·S and a pH of 2 ~ 4.

On the first, organic wetting agent on the surface of mullite fiber was removed by heat treatment. In order to improve the compatibility of mullite fiber and silica slurry matrix, interface coating was made by infiltration fiber perform with SiO2-Al2O3-ZrO2 sol, and dried at room temperature. Secondly, the fiber perform was infiltrated by silica slurry in vacuum, then the perform was treated. And then alumina sol was used to densify the composites through the infiltration-dry cycles, then sintered at appropriate temperature. At last, test samples were prepared by precision machining.

2.2 Testing methods
The density of mullite fiber reinforced mullite composites samples were tested using Archimedes principle. The bending strength, compressive strength, tensile strength were tested by universal testing machine AG-IC. Netherland FEI Company Sirion200 hot field emission scanning electron microscope was used to observe the section micromorphology of composites samples. Dielectric constant and dielectric loss were tested using short circuit waveguide.

3. Results and Discussion

3.1. Effect of heat treatment temperature on mullite fiber strength
In order to select suitable ceramic heat treatment temperature and decrease the damage of fiber strength after heat treatment, the mullite fiber was respectively heat treated at 750 °C, 900 °C, 1150°C, 1250°C, 1350°C, 1450°C for 2h, heating rate is 5~7°C/min. The changes of strength with temperature were shown in Figure 1. Figure 1 showed that the strength retention rate was about 50% at 750°C, but more than 750°C, the strength damaged seriously. From 900°C to 1350°C, the strength retention had little difference, retention rate was around 30%. So the ceramic heat treatment temperature was set at 750~900°C.

![Figure 1](image)

**Figure 1.** The strength changes of mullite fibers with temperature.

3.2. Microstructure of interface coating on the surface of mullite fibers
Figure 2 showed the surface micromorphologies of quartz fibers before and after filling SiO2-Al2O3-ZrO2 interface coating. Figure 2 investigated that interface coating was uniformly coated on the surface of mullite fiber. Before interface coating, the average diameter of the silica fiber was 6.4μm, and the average diameter of the silica fiber was 6.96μm after interface coating, which indicated that the thickness of the coating was about 280nm, which was in agreement with the measurement results.
in Figure 2(c). Figure 3 showed the energy spectrum analysis of the interfacial coating. Figure 3 investigated that interface coating on surface of mullite fiber was composed of Al-Si-Zr, whose specific composition was showed in Table 1, which was agreement with the component supplied by manufacturer.

**Figure 2.** Surface micromorphologies of quartz fibers (a) before and (b) (c) after filling interface coating.

**Table 1.** Component of interface coating on the surface of mullite fiber

| Element | Weight percentage | Atomic percentage |
|---------|-------------------|-------------------|
| Al K    | 67.95             | 71.53             |
| Si K    | 26.42             | 26.71             |
| Zr L    | 5.63              | 1.75              |
| Total   | 100.00            |                   |

3.3. Microstructure of mullite fiber reinforced mullite composites

Figure 4 showed the microstructure of mullite fiber reinforced mullite composites at different temperatures of 800°C and 1250°C, respectively. Figure 1a, b revealed that the mullite matrix was uniform and compact between the fiber and the fiber, and there were lots of fibers pull-out from mullite matrix when the tensile fracture occurred. Figure 1c, d revealed that interface between the mullite fibers was fuzzy, and the fracture of composites was even, which appeared the brittle fracture. So 800°C was appropriate.

3.4. Properties of mullite fiber reinforced mullite composites

Table 2 showed density, the tensile strength, bending strength, compressive strength and dielectric properties of the composites. As can be seen, mullite fiber reinforced mullite composites had excellent mechanical properties and dielectric properties, which can be regarded as one excellent high-temperature wave-transparent materials. The typical flat sample prepared was shown in Figure 5.
Figure 4. Microstructure of mullite fiber reinforced mullite composites at different temperatures: (a,b) 800°C; (c,d) 1250°C.

Table 2. Properties of mullite fiber reinforced mullite composites.

| Item                        | Performance index |
|-----------------------------|-------------------|
| Density (g/cm³)             | 1.75 ~ 1.80       |
| Tensile strength (MPa)      | 48.7              |
| Bending strength (MPa)      | 65.4              |
| Compressive strength (MPa)  | 82.7              |
| Dielectric constant         | 3.2 ± 0.1         |
| Loss angle tangent          | ≤ 8 × 10⁻³        |

Figure 5. The typical flat sample of mullite fiber reinforced mullite composites.
4. Conclusions
Mullite fiber reinforced mullite composites were prepared with stitched structural perform, silica slurry and aluminum sol by the method of liquid phase impregnation.

The heat-treated temperature of composites was set at 750–900°C by strength testing of mullite fiber heat-treated at different temperatures.

SiO$_2$-Al$_2$O$_3$-ZrO$_2$ interface coating was prepared on the surface of mullite fiber uniformly, which improved the composites properties greatly.

Mullite fiber reinforced mullite composites had excellent mechanical properties and dielectric properties, which can be regarded as one excellent high-temperature wave-transparent materials.

References
[1] Kanka B, Schneider H 2000 Journal of the European Ceramic Society 20 619
[2] Bao Y H, Patrick S, Nicholson J 2008 Journal of the European Ceramic Society 28 3041
[3] Chen Z F, Han G F, Zhang F P, et al. 2004 Journal of Chinaese Ceramic Society 24 5