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Fabrication of dense β-Si_3N_4-based ceramic coating on porous Si_3N_4 ceramic

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Abstract

β-Si_3N_4-based coating was prepared on the porous Si_3N_4 ceramic using the liquid infiltration and filling method. The effect of sintering temperature on microstructures, water absorption, mechanical properties and erosion resistance of the coatings were investigated. The results show that phase components of all the coatings consisted of β-Si_3N_4, $Y_{10}Al_2Si_3O_{18}N_4$ and Y–Si–Al–O glass. The coatings prepared at 1700 and 1800 °C had a good water resistance, which can be attributed to the high density of the coatings. The defects, including large pores and micropores, were produced within the coating when the sintering temperature was up to 1900 °C, thus leading to the reduction of the water resistance. Hardness and elastic modulus of the coating increased gradually as the sintering temperature increased from 1700 to 1800 °C, but it began to decrease when the sintering temperature was 1900 °C. The coating prepared at 1800 °C displayed a good erosion resistance.

Keywords: Porous Si_3N_4; β-Si_3N_4-based coating; Mechanical properties; Erosion resistance

1. Introduction

Silicon nitride (Si_3N_4) ceramic is widely used as high temperature structural materials due to its excellent mechanical properties at high temperature, but it is difficult to be machined into complex components because of its good wear resistance and high hardness.[1–3] More recently, porous Si_3N_4 ceramic is gaining more and more interest as a substitute for dense Si_3N_4 ceramic, because it is lighter and possesses better machinability, lower dielectric constant and thermal conductivity.[4–7] However, the pores of porous Si_3N_4 ceramic can absorb moisture seriously when it is applied in atmosphere, leading to the decrease of dielectric properties and thermal insulation. Besides, the surface pores of porous Si_3N_4 ceramic can result in the degradation of surface hardness and erosion resistance.[8] Therefore, the application of porous Si_3N_4 ceramic as some structural/functional components, such as radome and insulation materials, is usually limited. In order to obtain a Si_3N_4 ceramic with not only good machinability and erosion resistance but also good dielectric properties and thermal insulation, it is a significant design that porous Si_3N_4 ceramic is coated with a dense Si_3N_4 ceramic in order to improve the tightness and the resistance to erosion.

Up until now, various methods, such as chemical vapor deposition (CVD),[9–11] liquid sintering,[12] are often employed to fabricate a dense amorphous Si_3N_4 or α-Si_3N_4-based coating on the porous Si_3N_4 ceramic. However, those coatings often failed in the form of cracking due to the inherent brittleness and phase transformation in the harsh environment, which limits the applications of the coated porous Si_3N_4 ceramic enormously. β-Si_3N_4 ceramic not only shows a good high temperature stability but also has the higher strength and toughness. However, no literature published about using β-Si_3N_4-based ceramic as a protective coating for porous Si_3N_4 ceramic. Preparation of β-Si_3N_4 ceramic using liquid sintering requires very high sintering temperature (1600–1900 °C) to achieve phase transformation and densification of ceramic, which can make β-Si_3N_4 ceramic cause a very big volume shrinkage.[13,14] So, the stress cracks and/or large deformation will be inevitably produced in the coating owing to the mismatch of shrinkage between the coating and the substrate.[15]

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In this work, a dense β-Si_3N_4-based ceramic coating was fabricated by the liquid infiltration and filling method on the porous Si_3N_4 ceramic for the resistance to moisture and erosion. The double-layer powders composed of α-Si_3N_4 inner layer and oxides outer layer were deposited on the porous Si_3N_4 ceramic by the slurry spraying method. During sintering, the liquid formed by oxides can infiltrate into α-Si_3N_4 powder layer to prompt the liquid sintering of α-Si_3N_4 as sintering additive, forming a dense β-Si_3N_4-based coating. The effect of sintering temperature on the microstructure, water absorption, hardness, elastic modulus and erosion resistance of the coatings were investigated in details.

2. Experimental procedure

2.1. Preparation of the coatings

The porous Si_3N_4 ceramic fabricated in our previous work was employed as the substrate.[16] The samples were machined into the discs with the dimensions of Ø 20 mm × 5 mm, then ground with sand paper and polished with a 0.25 μm diamond pastes, at last, ultrasonic cleaned in ethanol and dried at 100 °C for 10 h in drying oven. Pore size of the porous Si_3N_4 ceramic is in the range of 0.4–1.3 μm, and total porosity is about 56%.

The α-Si_3N_4 powder (mean particle size: 0.2 μm, purity > 95%) was mixed in ethanol to prepare the α-Si_3N_4 slurry with the solid content of 5.5 vol%. SiO_2 powder (mean particle size: 0.9 μm, purity >99%), Y_2O_3 and Al_2O_3 powder (mean particle size: 0.5 μm, purity >99%) were mixed according to the mass ratio of Y_2O_3:SiO_2:Al_2O_3 = 4:3:3. The oxide mixture mixed with ethanol to prepare the Y_2O_3–SiO_2–Al_2O_3 (YSA) slurry with the solid content of 5.2 vol%.

The double-layer powders were prepared on the porous Si_3N_4 ceramic by the slurry spraying using the atomization spray gun (Type: F-2; Taiwan, China). The parameters of spraying were: nozzle diameter is 0.5 mm, air pressure is 0.4–0.7 MPa, spraying distance is 25 cm and spraying rate is 20 cm/s. The α-Si_3N_4 powder was deposited on the porous Si_3N_4 ceramic directly as the inner layer, and then the YSA powder outer layer was deposited on the α-Si_3N_4 powder inner layer. The α-Si_3N_4 slurry and the YSA slurry were sprayed four times, respectively. The porous Si_3N_4 ceramic with double-layer powders was sintered at 1700–1900 °C. All the samples were sintered for 1 h in N_2 atmosphere, and the heating and cooling rate was 5 °C/min.

2.2. Analysis of composition and structure of the coatings

The coatings were analyzed by X-ray diffraction (XRD, Type: D/MAX-2400X) to determine the phase compositions. The surface and cross-section morphologies of the coating were observed using scanning electron microscopy (SEM, Type: FEI6000), equipped with energy dispersive spectroscopy (EDS). The microstructure of the coating was observed by transmission electron microscope (TEM, Type: JEM-2100F; JEOL, Japan), and the interface between β-Si_3N_4 and glass and element distribution in the glass were studied by high-resolution electron microscope (HRTEM) equipped with an energy dispersive spectroscopy (EDS).

2.3. Characterization of properties and erosion behavior of the coatings

The water absorption and the bulk density (ρ) of the porous Si_3N_4 ceramic with and without coatings were measured by Archimedes method according to ASTM C-20 standard. The total porosity (P) was calculated according to the following equation:

$$P = \left(1 - \frac{\rho}{\rho_o}\right) \times 100\% \tag{1}$$

where ρ_o is the theoretical density of the samples. The theoretical density of porous Si_3N_4 substrate (ρ_{OS}) and the coated porous substrate (ρ_{OC}) were estimated by mixture rule according to Eqs. (2) and (3),[9] respectively.

$$\rho_{OS} = V_{Si_3N_4}\rho_{Si_3N_4} + V_{Al_2O_3}\rho_{Al_2O_3} + V_{Y_2O_3}\rho_{Y_2O_3} \tag{2}$$

$$\rho_{OC} = V_{OS}\rho_{OS} + V'_{Si_3N_4}\rho'_{Si_3N_4} + V'_{Al_2O_3}\rho'_{Al_2O_3} + V'_{Y_2O_3}\rho'_{Y_2O_3} + V'_{SiO_2}\rho'_{SiO_2} \tag{3}$$

where $\rho_{Si_3N_4}$, $\rho_{Al_2O_3}$ and $\rho_{Y_2O_3}$ are the theoretical densities of Si_3N_4 (3.20 g/cm^3), Al_2O_3 (3.97 g/cm^3) and Y_2O_3 (5.01 g/cm^3) in the porous Si_3N_4 ceramic, respectively, and $V_{Si_3N_4}$, $V_{Al_2O_3}$ and $V_{Y_2O_3}$ are the volume fractions of Si_3N_4, Al_2O_3 and Y_2O_3, respectively. $\rho'_{Si_3N_4}$, $\rho'_{Al_2O_3}$, $\rho'_{Y_2O_3}$ and ρ'_{SiO_2} are the theoretical densities of Si_3N_4 (3.20 g/cm^3), Al_2O_3 (3.97 g/cm^3), Y_2O_3 (5.01 g/cm^3) and SiO_2 (2.50 g/cm^3) within the coating, respectively, and V_{OS} is the volume fraction of porous substrate and $V'_{Si_3N_4}$, $V'_{Al_2O_3}$, $V'_{Y_2O_3}$ and V'_{SiO_2} are the volume fractions of Si_3N_4, Al_2O_3, Y_2O_3 and SiO_2 within the coating, respectively.

Nano-indentation test was performed using Nano-Indenter^{TM} XP (MTS system Corp., USA) system with a Berkovich indenter on the well-polished cross-section of the coatings. All of the measurements were made with a constant load value (6000 μN). The hardness and elastic modulus of the coating were calculated from load-penetration depth curves using the Oliver and Pharr method [17]. A total 15 indentations were recorded to get the average values of the hardness and elastic modulus. Load, depth and time were recorded continuously during the indentation process.

The erosion behavior of the uncoated and coated porous Si_3N_4 ceramic was measured using the self-made jet slurry erosion tester which consists of the water tank with a high pressure water injection pump, the spraying tube with a nozzle, sand hopper and sample table. The slurry consisted of water and silica sand. During eroding, the water is introduced into the spraying tube with the nozzle by the high pressure water injection pump, at the same time the silica sand is injected into the nozzle due to the absorption caused by the water flowing, forming the eroded slurry. The detailed information about the erosion method can be found elsewhere.[18] All parameters of erosion test were: nozzle diameter is 8 mm, spray distance is 50 mm, jet