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Effect of magnesium titanate content on microstructures, mechanical performances and dielectric properties of Si_3N_4-based composite ceramics

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ABSTRACT

Silicon nitride-based composite ceramics with different contents of magnesium titanate have been fabricated via gas pressure sintering method. The phase compositions, microstructure, mechanical performances and dielectric properties of the composite ceramics were investigated. The density of the Si_3N_4-based composite ceramics firstly increased with additive of magnesium titanate powder up to 5 wt% and then gently decreased, and the mechanical properties firstly increased and then declined. Besides, the dielectric constant and dielectric loss increased with the increase of magnesium titanate contents. For the Si_3N_4-based composite ceramics with 5 wt% magnesium titanate powders, the flexural strength, elastic modulus, dielectric constant and dielectric loss reached 451 MPa, 274 GPa, 7.65, 0.0056, respectively. These results suggested that the magnesium titanate was beneficial for the improvement of mechanical performances and dielectric constant of Si_3N_4-based composite ceramics.

1. Introduction

Silicon nitride ceramics have high strength, high modulus, high temperature resistance, oxidation resistance and wear resistance, etc., and have the special value in high temperature, high speed, strong corrosive media work environment. Furthermore, duo to its excellent properties such as thermal conductivity, thermal shock resistance, low dielectric constant and dielectric loss, as well as good high-frequency electromagnetic wave transmission performance, silicon nitride ceramics get wide applications in aerospace electronics [1–3]. However, because of the strong covalent bonding between silicon and nitrogen atoms, solid-state diffusion is very slow, thus preventing the densification of the Si_3N_4 during sintering. We usually add sintering additives to the Si_3N_4 ceramics in order to form liquid phase, and the liquid formed via the chemical reactions between the additives and SiO_2 on surface of Si_3N_4 phases could enhance the diffusivity of atoms and lower the sintering temperature [4,5]. It is well known that Y_2O_3 has been a promising sintering additive for both pressureless and gas-pressure sintered Si_3N_4 [6–11].

Although Si_3N_4 ceramics have excellent dielectric properties, the dielectric constant of silicon nitride can not meet the requirement of some wave-transparent radome materials with higher dielectric constant in modern society. Therefore, in order to satisfy the needs of high dielectric constant of the wave-transparent composite radomes, we will choose materials with high dielectric constant as the additive to improve dielectric constant of Si_3N_4 ceramics. $MgTiO_3$ has a limonite-type structure with ε=17 (at 7 GHz) and tanδ reaches ~10^{-4} [12]. Also, Mg_2TiO_4 ceramic has a spinel structure with ε=14 and tanδ reaches ~10^{-4} [13]. Thus we can add magnesium titanate to silicon nitride ceramics.

In this study, we fabricated the Si_3N_4-based composite ceramics with different amounts of magnesium titanate powders by gas pressure sintering method using 4 wt% Y_2O_3 as sintering additive. The influences of magnesium titanate content on the phase compositions, microstructures and mechanical performances, as well as dielectric properties were studied.

2. Experimental procedure

2.1. Materials processing

Commercial P grade Si_3N_4 powder (D_{50}=1.30 μm, α phase content: 95%) and magnesium titanate (D_{50}=2.50 μm) were used as the starting powders. Y_2O_3 (D_{50}=0.70 μm, 99.99% purity) was added as the sintering additive. The amount of adding Y_2O_3 particles was 4 wt%. And the mass fraction of magnesium titanate varied from 0% to 25% (Table 1). Fig. 1(a) shows the SEM micrograph of magnesium titanate particles with the size of 0.5–2 μm which clearly revealed that

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Table 1
Identification, chemical compositions, sintering linear shrinkage and density of the samples.

Sample code	Composition of powder mixture (wt%)			Sintering shrinkage Linear (%)	Density (g/cm^3)
	Si_3N_4	Y_2O_3	Magnesium titanate		
M0	96	4	0	12.18	2.276
M1	96	4	5	19.69	3.334
M2	96	4	10	19.50	3.331
M3	96	4	15	19.48	3.330
M4	96	4	20	19.13	3.327
M5	96	4	25	18.73	3.323

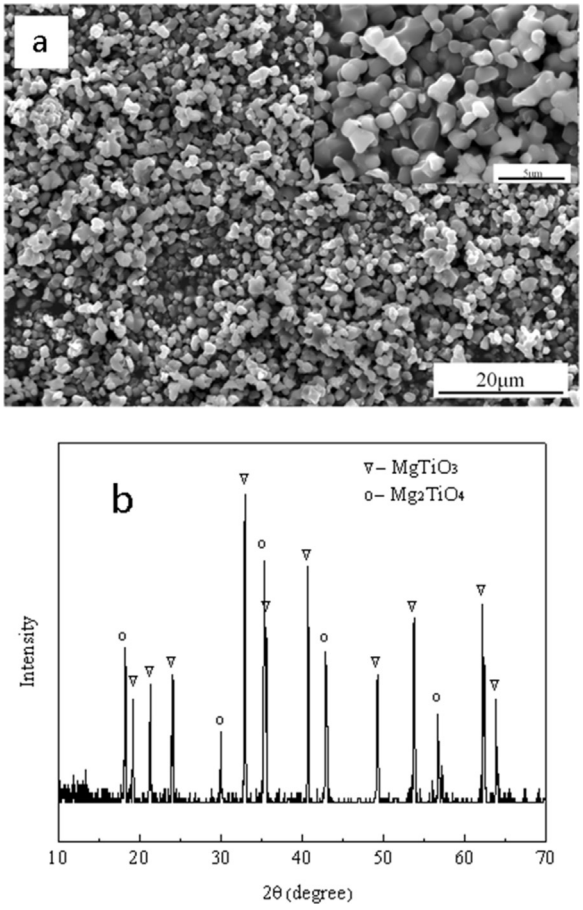

Fig. 1. SEM micrograph and XRD pattern of magnesium titanate powders.

magnesium titanate was homogeneous dispersion basically in spite of a small amount of particles were agglomerated. Besides, Fig. 1(b) shows the XRD pattern of magnesium titanate powders which contained $MgTiO_3$ and Mg_2TiO_4 phase, and the content of $MgTiO_3$ phase was 71 wt%.

The above raw powders were mixed and wet milled in anhydrous ethanol for 24 h with speed of 220 rpm/min in high-energy ball miller. The slurry was dried, and then passed through 50 mesh sieve. The mixed powders were formed into compacts of about Ø40 mm×120 mm in size under an isostatic pressing of 80 MPa. And the green compacts were dried at 110 °C for 3 h in order to reduce the humidity in the ventilation drying. And they were marked and weighed. Finally, the green bodies were embedded in the Si_3N_4 powders in a graphite crucible and sintered in a graphite furnace at 1700 °C for 2 h under a steady pressure of 0.3 MPa in N_2 atmosphere with a heating rate of 5 °C/min.

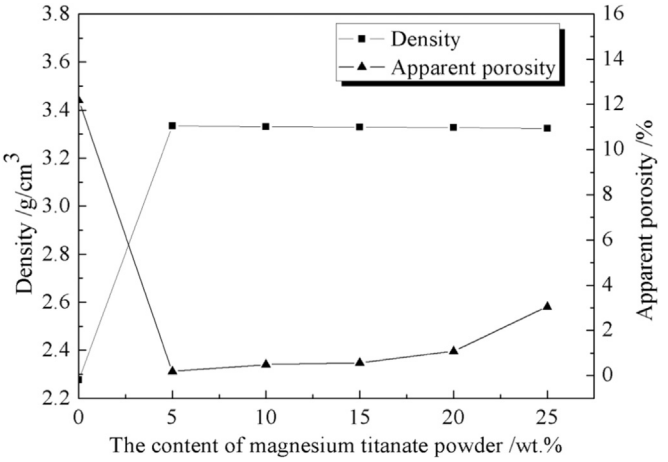

Fig. 2. Influence of different magnesium titanate powder contents on the density and porosity of the Si_3N_4-based composite ceramics.

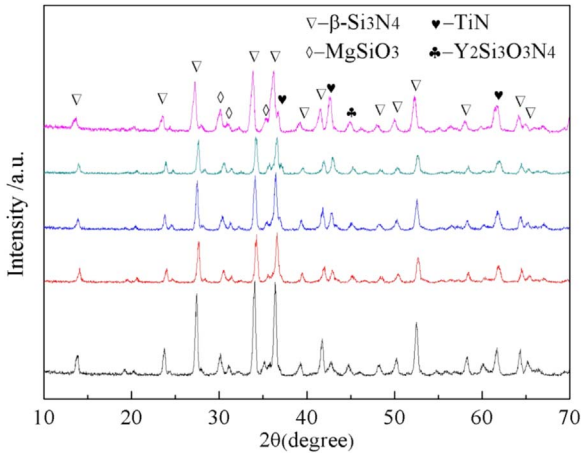

Fig. 3. XRD patterns of the Si_3N_4-based composite ceramics with different magnesium titanate powder contents. (a) 5 wt%, (b) 10 wt%, (c) 15 wt%, (d) 20 wt%, (e) 25 wt%.

2.2. Materials characterization

The density and apparent porosity of the specimens were determined by the Archimedes principle, which means that the samples were placed in distilled water under vacuum environment. X-ray diffraction (XRD, D/max 2500, Tokyo, Japan) was carried out using Cu Kα (λ=0.15405 nm) radiation to determine the phase composition. The microstructure of fracture surface of samples was observed by scanning electron microscopy (SEM, FEI Quanta 200, Brno, Czech Republic) after being sputtered with gold film. The microstructures were observed on polished sections after etching in molten NaOH at 350 °C for about 5 min. The flexural strength and elastic modulus were measured on test specimens of 3 mm×4 mm×36 mm using a three-point bend method with a span of 30 mm and a cross-head speed of 0.5 mm/min, and five specimens were tested to obtain the average values. For the measurements of dielectric properties, specimens with a size of 6.96 mm×3.48 mm×1.00 mm were tested in the 35 GHz frequency at room temperature by the short circuited waveguide technique with the help of an Agilent Network Analyzer (model N5230A, Agilent Technologies Inc, Palo Alto, CA, USA).

3. Results and discussion

3.1. Density and porosity of Si_3N_4-based composite ceramics

Variations in the density and apparent porosity of the samples are shown in Fig. 2. The density of the Si_3N_4-based composite ceramics