

Mechanical and Electromagnetic Properties of Carbon Fiber Toughened Si₃N₄ Ceramic Composites

Qinggang Li^{1*}, Yuhang Du², Ning Sun¹, Jinkai Li¹, Mengyong Sun³, Guopu Shi¹

¹Department of Material Science and Engineering, University of Jinan, Jinan, China

²Shandong Provincial Key Laboratory of Preparation and Measurement of Building Materials, Jinan, China

³Department of Material Science and Engineering, No. 52 Institute of China North Industry Group, Yantai, China

Research Article

Received: 18-Nov-2022, Manuscript No. JOMS-22-80277; **Editor assigned:** 21-Nov-2022, PreQC No. JOMS-22-80277 (PQ); **Reviewed:** 05-Dec-2022, QC No. JOMS-22-80277; **Revised:** 12-Dec-2022, Manuscript No. JOMS-22-80277 (R); **Published:** 19-Dec-2022, DOI: 10.4172/2321-6212.10.8.003.

***For Correspondence:**

Qinggang Li, Department of Material Science and Engineering, University of Jinan, Jinan, China

E-mail: mse_liqg@ujn.edu.cn

Keywords: Mechanical properties; Carbon fiber; Si₃N₄ ceramic composite; Dielectric properties

ABSTRACT

Carbon fiber-reinforced Si₃N₄ ceramic materials were prepared by hot pressing sintering process using silicon nitride as substrate and short carbon fiber reinforcement. The mechanical properties, microstructure and electromagnetic properties of the sintered samples were evaluated. Experimental results show that atmosphere hot pressing can reduce the sintering temperature of Si₃N₄ ceramics and promote phase transition. When the additional amount of carbon fiber is 3.0 wt%, the sintering temperature is 1700°C, the holding time is 30 min, forming pressure is 30 MPa, the sintered sample has the best extensive properties: hardness (>8.5 GPa) and density (>2.8 g/cm³) meet the engineering needs, fracture toughness is enhanced by 22.7%, fracture toughness can reach 8.11 MPa • m^{1/2}, dielectric properties are also improved.

INTRODUCTION

Silicon nitride ceramics have high hardness, strength and fracture toughness, high-temperature resistance, corrosion resistance and a small coefficient of thermal expansion and other excellent properties [1,2]. It is widely used in cutting tools, engine high-temperature parts, ceramic bearings, and metallurgical products, chemical corrosion-resistant and wear-resistant parts [3]. In addition, its low density, good wave permeability, stable dielectric properties, good thermal shock resistance, and rain erosion resistance make silicon nitride ceramics an ideal material for Radome [4,5]. However, most of them are composed of strong covalent bonds, which are difficult to compact and have low fracture toughness, limiting their application in a wider range of fields such as biological, military and nuclear industries [6-9]. Carbon fiber

toughened Si₃N₄ ceramics can prevent crack propagation in the ceramic matrix and decrease the inherent brittleness of ceramic materials effectively [10-12]. This research uses hot-pressing sintering technology, by choosing the appropriate proportion of each component, in the forming pressure of 30 MPa, sintering temperature of 1700°C. The effects of carbon fiber content on the density, hardness, flexural strength, fracture toughness, microstructure, and dielectric properties of Si₃N₄ ceramics were discussed.

MATERIALS AND METHODS

Commercial powders of Si₃N₄ (99.9% pure, with an average particle size of 1 μm), 0.5 wt.% Y₂O₃-Al₂O₃ (99.9% pure, with an average particle size of 0.5 μm) and 0 wt.%, 0.3 wt.%, 0.6 wt.%, 1.5 wt.%, 2.1 wt.%, 3.0 wt.% shortcut carbon fiber (99.9% pure, average length and diameter of 1 mm and 5 μm) were used as starting materials for the hot-pressing sintering process. With alcohol as the medium in the ball grinding tank for ball milling mixing, 24 hours after the slurry from the ball grinding tank and drying for 8 hours. The dried powder was placed in a graphite mold for hot pressing sintering under nitrogen atmosphere. The sintering temperature is 1700°C, applying pressure is 30 MPa, and holding time is 30 min.

The obtained ceramics were diamond-polished, chamfered, and cut into bars (3 mm × 5 mm × 30 mm) for subsequent characterizations. The flexural strength and fracture toughness of the post-ceramic composites were determined by the three-point fracture method and single-edge notched beam test by applying crosshead speeds of 0.5 mm•min⁻¹ and 0.05 mm•min⁻¹, respectively, using an electromechanical universal testing machine (Instron-5980, Instron Corporation, USA). The test of dielectric constant is measured under the K band by using dielectric constant tester. X-Ray Diffraction (XRD) measurements were performed using an X-ray diffractometer (D8-Advance, Bruker Corporation, Karlsruhe, Germany) with CuKα (1.54 Å) radiation. The microstructures on the fracture surfaces of the ceramic composites were observed using scanning electron microscopy (SEM, Hitachi S-4800, Hitachi, Japan).

RESULTS AND DISCUSSION

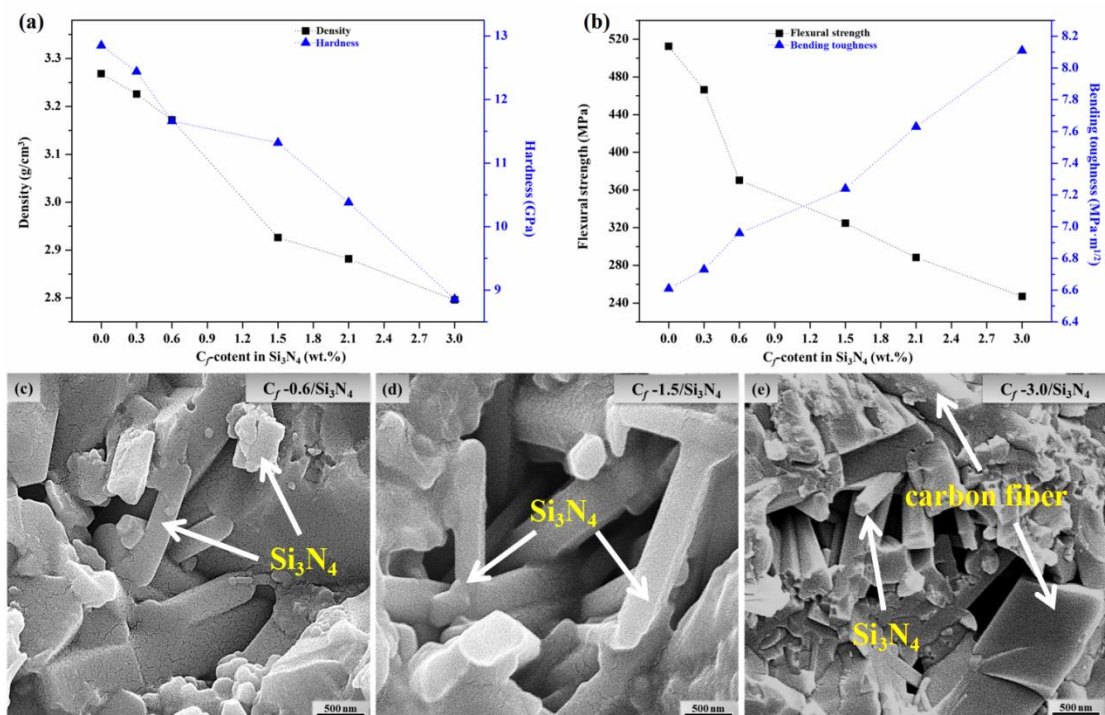
Figures 1a and 1b show the mechanical properties of Si₃N₄ composite material. The density of the sample decreases with the increase of carbon fiber content, mainly because the addition of carbon fiber affects the formation and rearrangement of different particles. It is also possible that the early dispersion effect is not good, leading to partial aggregation of carbon fiber. In addition, the sintering temperature of carbon fiber itself is lower than that of silicon nitride, which may be due to the decrease in densification caused by the lower sintering temperature. However, the sample density is closely related to porosity, and the two are negatively correlated. In the position where pores exist, the stress concentration is also quite severe, which often leads to the main cause of cracks, resulting in the reduction of mechanical properties of ceramic materials. The hardness and flexural strength of the sample will show a downward trend.

The flexural strength of Si₃N₄ ceramics decreases with the increase of carbon fiber content, while the fracture toughness is opposite. The fracture toughness of the ceramic sample was the highest when the maximum carbon fiber content was added, indicating that the introduction of carbon fiber had a good toughening effect on the ceramic sample, especially when the carbon fiber content was 3.0 wt.%, the fracture toughness of the ceramic sample was the maximum in this experiment. On the one hand, the size of carbon fiber particles belongs to the micron level, and most of the carbon fiber particles are distributed in the glass phase of the Si₃N₄ grain boundary after mixed hot pressing and sintering, which has a deflecting effect on the propagation of cracks, thus enhancing the toughness of silicon nitride

ceramic materials. In addition, the elastic modulus of carbon fiber is different from that of Si₃N₄, so the energy of applied pressure is first transferred to carbon fiber, and the fracture work is absorbed by interface friction to prevent the further expansion of cracks. However, with the increase of carbon fiber content, the strength of sintered body decreases, which may be due to the reaction of silicon nitride and carbon fiber to generate silicon carbide particles at high temperature, which increases the interface bonding force, but has a certain particle toughening effect.

Figures 1c-1e shows the specimen fracture morphology, carbon fiber and the physical and chemical compatibility of the compatibility of silicon nitride ceramic not very ideal, combined with the weak interface, and the addition of carbon fiber and the original crystalline silicon nitride particles and particle arrangement has the effect of blocking, so are the main factors influencing the bending strength. The increase in fracture toughness is closely related to the toughening mechanism of carbon fiber. On the one hand, the addition of carbon fiber can absorb the energy of the load, resist the external force and prevent the further expansion of the crack. In addition, the elastic modulus of carbon fiber is larger than that of the matrix, and the crack deviates from the original direction. It expands along the bonding surface or expands in the matrix, changing the surface energy and thus absorbing more fracture work. On the other hand, when the fiber is pulled out from the matrix, it is toughened by absorbing fracture work by interface friction.

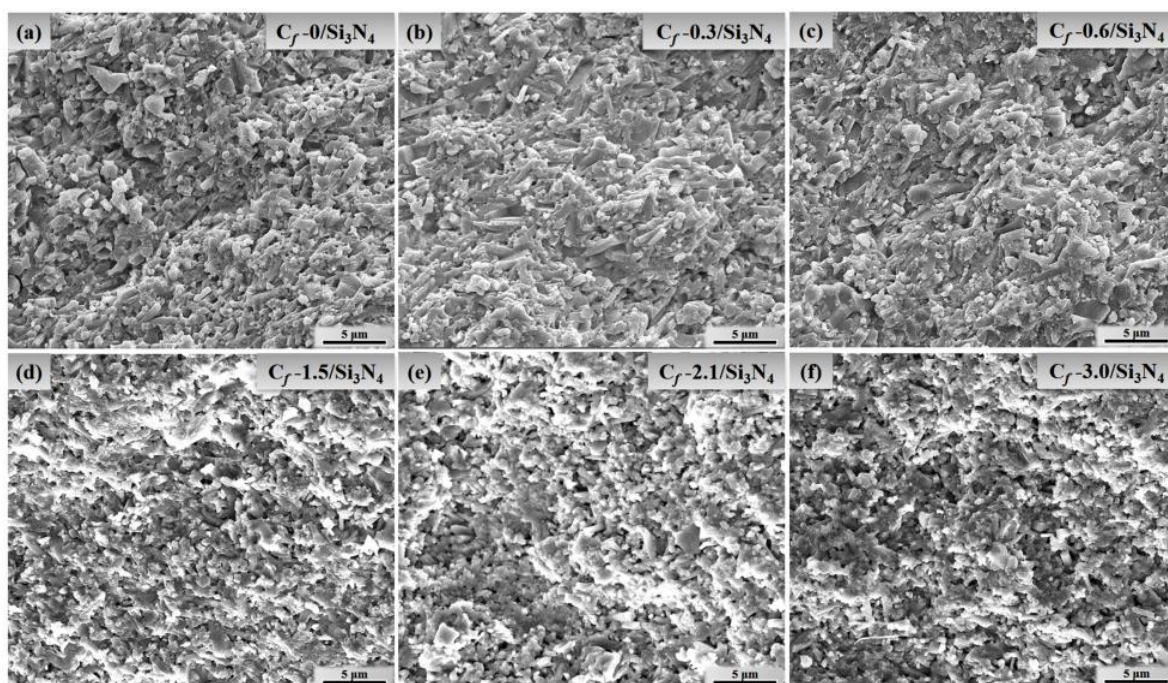
Figure 1. Mechanical properties (a and b) and fracture morphology (c, d and e) of carbon fiber toughened Si₃N₄ ceramic composites. (a) Density and hardness of Si₃N₄ (Note: ■ Density, ▲ Hardness) (b) Flexural strength and bending toughness of Si₃N₄ (Note: ■ Flexural strength, ▲ Bending toughness) (c) C_f-0.6/Si₃N₄ (d) C_f-1.5/Si₃N₄ (e) C_f-3.0/Si₃N₄



Figures 2a-2f is the SEM images of the microstructure of the composite material. From the morphology of group Cf-0, it can be seen that a large number of long columnar Si₃N₄ is generated and there is a certain porosity, which indicates that most of α silicon nitride is converted into β silicon nitride. The rapid diffusion rate of silicon nitride and the suitable holding time led to the rapid phase transformation. The morphology of Cf-0.3 also shows that long columnar β phase is

formed at 1700°C, which indicates that the phase transformation rate is similar to that of Cf-0, the porosity increases and the growth is disorderly. This is because the presence of carbon fiber affects the α Si_3N_4 particles and the shrinkage of the body leads to the existence of large pores. The fracture morphology of group Cf-0.6 shows that there are a large number of long columnar grains interwoven and connected, forming a network structure, which indicates that the growth of β phase grains has filled the void and other substances may be formed. And that's why the strength of fracture toughness has increased.

Figure 2. SEM images of the microstructure of carbon fiber toughened Si_3N_4 ceramic composite. (a) Cf-0/ Si_3N_4 (b) Cf-0.3/ Si_3N_4 (c) Cf-0.6/ Si_3N_4 (d) Cf-1.5/ Si_3N_4 (e) Cf-2.01/ Si_3N_4 (f) Cf-3.0/ Si_3N_4 .



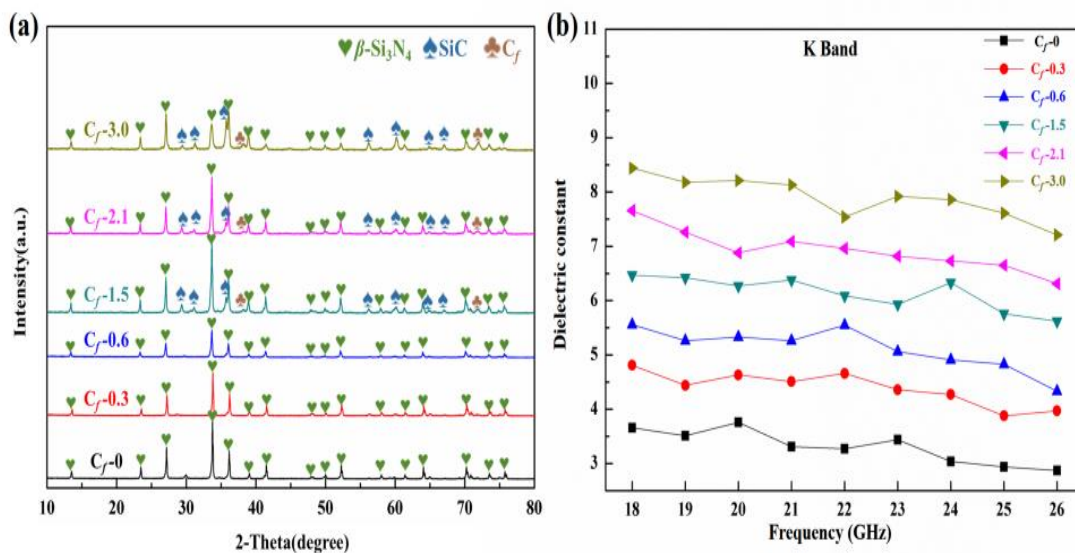
Cf-1.5 group, Cf-2.1 group and Cf-3.0 group all have good hot pressing and sintering phenomena. It can be seen from the fracture morphology diagram that there are a large number of long cylindrical grains interwoven and connected, forming a network structure, which is also the reason for the improvement of fracture toughness. With the increase of carbon fiber, the surface porosity increases gradually, and the distribution is not very uniform, indicating that α phase Si_3N_4 is transformed into β phase Si_3N_4 , and there is a mass aggregation phenomenon, indicating the existence of other phases. C phase or SiC phase may be present. The SiC content of the sample increased obviously, which was also caused by the significant increase in the addition of carbon fiber. In the three groups of samples, the number of long rod particles can still be observed to occupy the majority, that is, the morphology of β - Si_3N_4 . In the Cf-3.0 group, many large pores were observed, and the carbon fiber content in this sample was the largest, resulting in high porosity, low densification, greatly decreased hardness and strength, and obviously increased SiC phase with small-scale agglomeration and dispersed around Si_3N_4 powder.

The XRD pattern of carbon fiber toughened Si_3N_4 composite is shown in Figure 3a. In general, the main component of the composite is β - Si_3N_4 . In addition, the presence of Y and Al elements was not detected in the XRD pattern of Si_3N_4 ceramics, because the additional amount of combustion aid is very small compared with the amount of Si_3N_4 . The diffraction peaks of β - Si_3N_4 are dominant in Cf-0 and Cf-0.3 samples. As the carbon fiber content continues to increase,

when the carbon fiber content is Cf-0.6 and Cf-1.5, the peak containing C element and the peak of SiC compound may be formed can be seen in the figure. The intensity of β -Si₃N₄ diffraction peak decreases, indicating that the grain development is not good, and the densification degree of the sample decreases, which is consistent with the mechanical properties of the sample deteriorating. The addition of carbon fiber causes a small number of miscellaneous peaks. When the carbon fiber content is Cf-2.1 and Cf-3.0, there are β -Si₃N₄ and SiC phases in the sample. Compared with the above, SiC content increased and more complete. The XRD pattern is mainly β -Si₃N₄, the grain development is relatively good, the densification of the sample decreases slowly, the mechanical properties are consistent, and the density and hardness curve can be compared.

Figure 3b shows the data of the dielectric properties of carbon fiber to silicon nitride ceramic material at different frequencies in K-band. The overall effect is as follows: under the condition of the same carbon fiber content, the dielectric constant decreases with the increase of frequency, and under the condition of the same frequency, the dielectric constant increases with the increase of carbon fiber content. Carbon fiber is a dielectric absorbing material. The dielectric constant can be adjusted by adjusting the content of carbon fiber. Short carbon fiber is randomly distributed in the material, which can form harmonic oscillators or dipoles when interacting with radar waves. Due to certain content and random distribution, it cannot form a continuous conduction current, so short carbon fiber has a better absorbing effect on radar waves. This is where the dielectric constant increases.

Figure 3. (a) XRD patterns of carbon fiber toughened Si₃N₄ ceramic composites. (b) Dielectric constants in K band of carbon fiber toughened Si₃N₄ ceramic composites. (Note: ♥ β -Si₃N₄, ▲ SiC, ♣ C_f, ■ C_f-0, ● C_f-0.3, ▲ C_f-0.6, ▼ C_f-1.5, ◆ C_f-2.1, ► C_f-3.0)



CONCLUSION

The toughness of Si₃N₄ ceramics was improved by adding carbon fiber to Si₃N₄ ceramic matrix with its excellent mechanical properties and physical and chemical compatibility of two phases. When the sintering temperature is 1700°C, the relative density and hardness of Si₃N₄ ceramics decrease with the increase of carbon fiber content. The

relative density of the samples decreased from 85.6%, the hardness from 12.85 GPa to 8.86 GPa, and the fracture toughness increased by 22.7%. The fracture modes of Si₃N₄ ceramics include intergranular fracture and transgranular fracture. X-Ray Diffraction analysis shows that β-Si₃N₄ is the main component of the prepared Si₃N₄ ceramics, α-Si₃N₄ is completely transformed, and when the carbon content increases to a certain extent, SiC is formed. The main toughening mechanisms of carbon fiber toughened silicon nitride ceramics are fiber pulling out mechanism and dispersion toughening of SiC particles, thus improving the toughness of silicon nitride ceramics. The dielectric properties of ceramics are improved with the increase in carbon fiber content.

ACKNOWLEDGEMENT

Authors appreciate the financial supported by the Shandong Province Key Research and Development Plan (2020JMRH0401). The project was supported by the State Key Laboratory of Advanced Technology for Materials Synthesis and Processing (Wuhan University of Technology). This work was financially supported by the National Natural Science Foundation of China (51632003), the Taishan Scholars Program, and the Case-by-Case Project for Top Outstanding Talents of Jinan.

DECLARATION OF INTEREST STATEMENT

No conflict of interest exists in the submission of this manuscript, and the manuscript is approved by all authors for publication. I would like to declare on behalf of my co-authors that the work described was original research that has not been published previously, and is not under consideration for publication elsewhere, in whole or in part. All the authors listed have approved the manuscript that is enclosed.

REFERENCES

1. Yan S, et al. Fabrication and tribological characterization of laser textured engineering ceramics: Si₃N₄, SiC and ZrO₂. *Cera Inter.* 2021;47:13789-13805.
2. Subramanian K, et al. Study on dry sliding wear and friction behaviour of Al₇O₆₈/Si₃N₄/BN hybrid composites. *Mater.* 2021;14:6560.
3. Yola ML, et al. A novel electrochemical lung cancer biomarker cytokeratin 19 fragment antigen 21-1 immunosensor based on Si₃N₄/MoS₂ incorporated MWCNTs and core-shell type magnetic nanoparticles. *Nanoscale.* 2021;13:4660-4669.
4. Zhao D, et al. BN Nanoparticles/Si₃N₄ wave-transparent composites with high strength and low dielectric constant. *J of Nanomater.* 2011.
5. Cheng Z, et al. Mechanical and dielectric properties of porous and wave-transparent Si₃N₄-Si₃N₄ composite ceramics fabricated by 3D printing combined with chemical vapor infiltration. *J of Adva Cera.* 2019:399-407.
6. Liu Y, et al. Fabrication and properties of carbon fiber-Si₃N₄ nanowires-hydroxyapatite/phenolic resin composites for biological applications. *Cera Inter.* 2020;46:16397-16404.
7. Lenka K, et al. Fracture toughness and toughening mechanisms in graphene platelet reinforced Si₃N₄ composites. *Scri Mater.* 2012;66:793-796.
8. Swift GA, et al. High-temperature elastic properties of in situ-reinforced Si₃N₄. *App Phys Lett.* 2003;82:1039-1041.
9. Tatami J, et al. Fracture resistance and contact damage of TiN particle reinforced Si₃N₄ ceramics. *J of the Cera Soc of Japan.* 2006;114:1049-1053.

10. Wang X, et al. Influence of short carbon fiber content on mechanical and dielectric properties of C-fiber/Si₃N₄ composites. *Scrip Mater.* 2007;57:309-312.
11. Bin L, et al. Effects of fiber surface treatments on mechanical properties of T700 carbon fiber reinforced BN-Si₃N₄ composites. *Mater Scie & Engin A.* 2007;471:169-173.
12. Wang HY, et al. Preparation and properties of carbon fiber/Si₃N₄ composites. *J of Inorg Mater.* 2014;29:1003-1008.