Available online at www.sciencedirect.com
SciVerse ScienceDirect
Ceramics International 39 (2013) 3135–3140
CERAMICS INTERNATIONAL
www.elsevier.com/locate/ceramint

Influences of SiC_f content and length on the strength, toughness and dielectric properties of SiC_f/LAS glass-ceramic composites

Zhibin Huang*, Wenbo Kang, Yuchang Qing, Fa Luo, Wancheng Zhou, Dongmei Zhu

State Key Laboratory of Solidification Processing, School of Materials Science and Engineering, Northwestern Polytechnical University, Xi'an 710072, China

Received 3 August 2012; received in revised form 27 September 2012; accepted 27 September 2012
Available online 3 October 2012

Abstract

SiC_f/LAS glass-ceramic composites were prepared by hot-press sintering using SiC fibers, SiO_2, Al_2O_3 and Li_2CO_3 powders as starting materials. Mechanical properties, such as strength, toughness, and dielectric properties at X-band microwave frequency of these composites were evaluated. Results showed that both the content and the length of SiC_f had significant influences on the dielectric and the mechanical properties of SiC_f/LAS composites. The complex permittivity of the composites rose from 12 to 35 for ε' and 18 to 120 for ε'' at 8.2 GHz with the increasing SiC_f content. In addition, the reinforcement and toughening effects of SiC_f on the composites were also significant. Our results indicate that the SiC_f/LAS composite is a promising material with both the electromagnetic wave absorbing and the load-bearing abilities suitable as structural microwave absorbing and microwave shielding materials.

Keywords: A. Hot pressing; B. Composites; C. Dielectric properties; D. Glass ceramics

1. Introduction

Silicon carbide fibers (SiC_f) have been extensively studied in past years for their excellent properties of oxidation resistance, thermal resistance, chemical stabilization, high tension strength, low density, etc. There have been a great number of researches about preparing high-temperature ceramic matrix composites with SiC fibers as reinforcement and toughening phase to enhance overall performances of the composites, e.g: SiC_f/SiC [1], SiC_f/β-SiAlON [2], SiC_f/ZrB_2 [3], SiC_f/$Al(PO_3)_3$ [4], SiC_f/metal [5], etc.

However, dielectric properties of SiC are also interesting, which is correlated with their potential applications in the field of microwave absorption and shielding [6–8].

The excellent properties of SiC_f mentioned above, especially the semiconductivity and relatively stable dielectric characteristics at high-temperature, make them highly promising absorbents which could be employed in microwave absorbing materials and shielding materials. Therefore it is possible to fabricate SiC_f-reinforced composites with both microwave absorbing and load-bearing abilities suitable as structural radar absorbing materials or structural radar shielding materials.

The lithium aluminum silicate (LAS) glass ceramic is an excellent candidate matrix for composites with SiC_f due to its distinctive advantages, such as low coefficient of thermal expansion (CTE), low density, high temperature resistance, chemical stability and transparency [9]. So, not only the LAS glass ceramic can bear high temperature but also matches well with the thermal expansion characteristics of SiC_f. Previously, we studied the hot-pressed sintered Al_2O_3-matrix composites including $MoSi_2$/Al_2O_3 [10] and C_{sf}/Al_2O_3 [11]. While comparing with Al_2O_3, the greatest advantage of LAS matrix is the excellent thermal shock resistance because its CTE is 10 times lower. In addition, the light-weight advantage of LAS is also significant, which is two-fifth lighter than Al_2O_3.

In the current paper, chopping SiC fibers perform reinforcing, toughening and microwave-absorbing functions in SiC_f/Al_2O_3 composites. Effects of the content and the fiber length of SiC_f on microstructure, mechanical properties and dielectric properties of SiC_f/Al_2O_3 composites were investigated.

*Corresponding author. Tel./fax: +86 29 88494574.
E-mail address: huangzhibin83@163.com (Z. Huang).

0272-8842/$ - see front matter © 2012 Elsevier Ltd and Techna Group S.r.l. All rights reserved.
http://dx.doi.org/10.1016/j.ceramint.2012.09.094

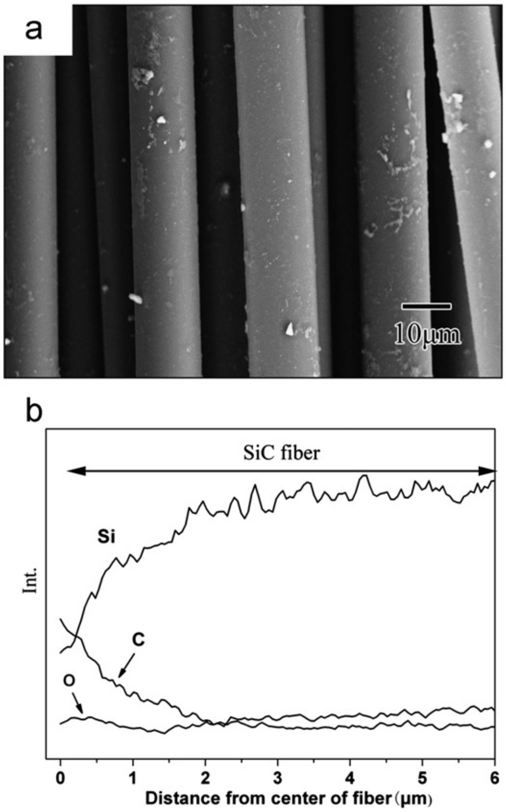

Fig. 1. (a) SEM photographs of KD-1 SiC_f and (b) chemical element intensity profile along fibers diameter.

Table 1
Properties of KD-1 SiC fiber and LAS glass ceramic.

	SiC fiber	LAS
Diameter (μm)	10	–
Density (g/cm^3)	2.4–2.45	2.5
Tensile-strength (MPa)	2200	–
Elastic modulus (GPa)	200	9.3×10^4
Thermal expansivity (K^{-1})	30×10^{-7}	$9–15\times10^{-7}$
Electric conductivity (S/cm)	2–3	2×10^{-11}

2. Experimental

2.1. Experimental materials and preparation method

The LAS glass-ceramic composites reinforced with chopping SiC fibers were prepared by hot-press sintering. Commercial KD-1 SiC fibers were provided by National University of Defense Technology, Changsha, China (see Fig. 1a), whose characteristics are given in Table 1. Volume fraction and length of the SiC_f investigated in this study were 0–35% and 2–4 mm respectively. Starting materials of LAS matrix were analytical grade SiO_2, Al_2O_3 and Li_2CO_3 powders, which were mixed in mol ratio of 4:1:1. The mixtures were melted at 1560–1620 °C for 2 h and quenched to get LAS glass. Then, the glass was crushed and milled into LAS powder with the size of 5–20 μm.

SiC_f and LAS powders were proportionally weighed and ultrasonic dispersed for 30 min in the water solution of polyvinyl alcohol and glycol. The mixtures were then mechanically blended for 45 min and spread on a thin sheets to dry. The dried mixtures were cut into round flakes (about 46 mm in diameter) and put into a graphite die layer by layer before sintering. Finally, the hot-press sintering was carried out at 1200 °C in nitrogen ambient with the heating rate of 10 °C/min. A 12.5 MPa load was applied to the graphite die before heating, and when the temperature reached 1200 °C, the load was increased to 20 MPa and maintained at that level for 10 min.

2.2. Mechanical properties measurement

Relative densities of the sintered samples were measured with the Archimedes method. The hot-pressed billets were ground and polished into 3 mm × 5 mm × 36 mm bars for flexural strength measurements, and 2 mm × 4 mm × 30 mm notched samples for fracture toughness measurement, respectively. The notch length was half of the sample height (2 mm) and cut by a thin diamond saw. Both tests were carried out using an Instron universal testing machine (2–20 kN) with a cross-head speed of 0.5 mm/min.

2.3. Dielectric measurement

The permittivity of the specimens was measured with the method, which was based on the measurements of the reflection and transmission modules, in the fundamental wave-guide mode TE10 by Agilent E8326B PNA series network analyzer (Palo Alto, CA), using standard samples (10.16 mm × 22.86 mm × 2.00 mm) for 8.2–12.4 GHz. After calibrated with short circuit and blank holder, reflection and transmission coefficients were obtained with the help of an automated measuring system. Both the real and the imaginary parts of the permittivity and permeability were then calculated after measurement. For a dielectric material ($\mu'=1$, $\mu''=0$) the relative error varies between 1% (pure dielectric) and 10% (highly conductive materials).

3. Results and discussion

3.1. Mechanical properties and microstructure

Fig. 1b shows the chemical element intensity profile along SiC_f diameter characterized by EDS. It can be observed that there is an excess carbon outer layer on the fiber surface, which is common for commercial SiC fibers and most likely formed during the fabrication process [12–14].

Considering the electrical conductivity of carbon is much higher than Si–C–O or Si–C one, the electric behavior of the KD-1 SiC_f is mainly dominated by the outer carbon-rich layer [15]. Fig. 2 shows images of the

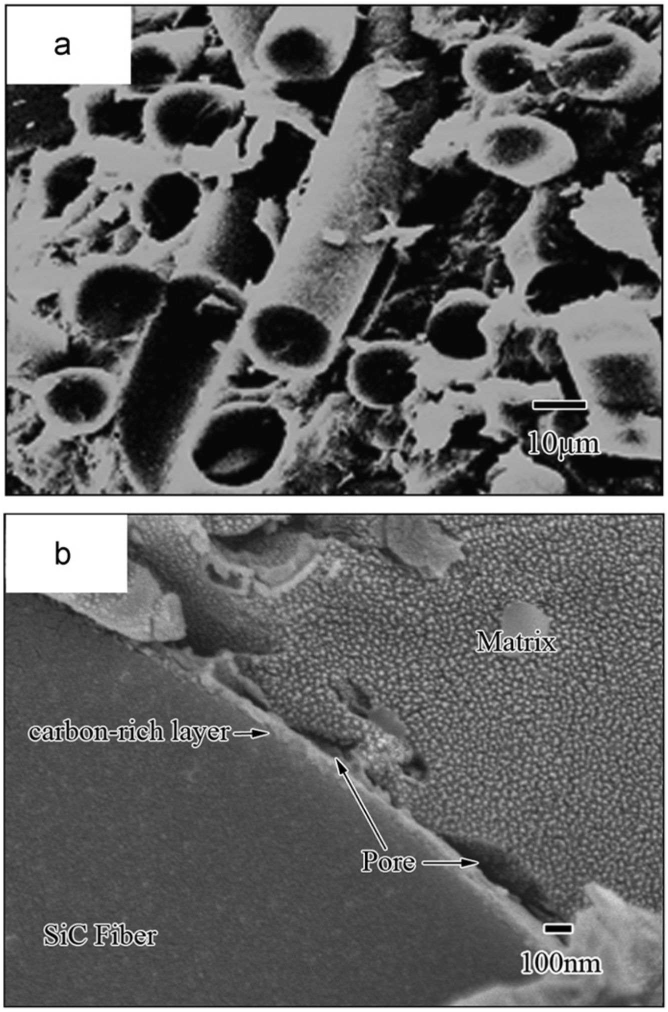

Fig. 2. Fractographs of SiC_f/LAS composites: (a) lower magnification and (b) higher magnification.

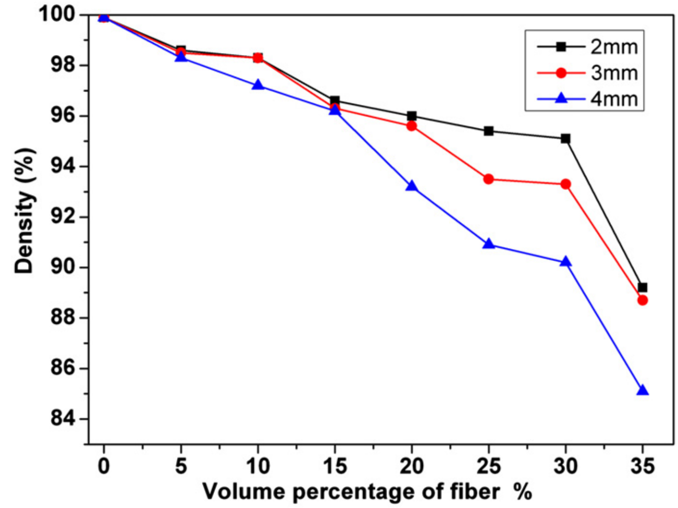

Fig. 3. Relative density of the composites versus SiC_f content.

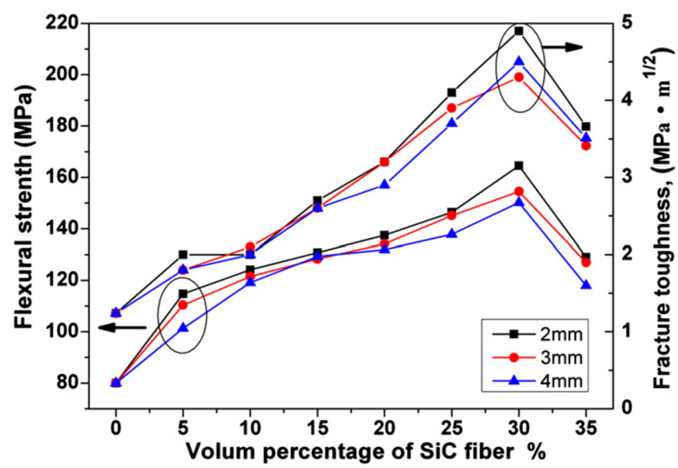

Fig. 4. Mechanical properties of the composites versus SiC_f content.

fracture surface of an as-received sample broken by bending test. As can be seen in Fig. 2a, the shape of SiC_f has little change after undergoing the process of hot-press sintering, which is very important for them to keep original dielectric properties in the composite. In addition, phenomena of fiber pull-out and breakage are observed, which are beneficial to improve the mechanical properties of the composites.

As shown in Fig. 2b, undesirable micro pores are observed on the interface along matrix side, although the interface between SiC_f and LAS are relatively clean and compact. The influences of the mismatch in thermal expansion are expected to be small because the CTEs between the fibers and the matrix are very close. We consider that the formation of these pores may be attributed to the gases released from the active oxidation of SiC_f, which usually occur in low-oxygen atmosphere [16]:

$$SiC + O_2 = SiO(g) + CO(g) \quad (1)$$

Relative densities of the composites with different contents of SiC_f are shown in Fig. 3. The relative density of pure LAS sintered at 1200 °C achieves 99.85%, while it decreases when SiC_f are added. The formation of micro pores (see Fig. 2b) on the fiber/matrix interface may be mainly responsible for the decrease of density. It is worthy of note, that the density of composites also drop fast with the increasing content and length of SiC_f, as shown in Fig. 3. This density-drop phenomenon can be attributed to the worse dispersion of SiC_f as more and longer SiC_f are added into the matrix.

Fig. 4 shows the mechanical properties of SiC_f/LAS composites. As expected, the addition of SiC_f significantly improves the flexural strength and fracture toughness of LAS matrix. The composites containing 30% SiC_f shows the best flexural strength and fracture toughness, which are almost two times and five times higher than pure LAS glass ceramic respectively. It is also important to note that the mechanical properties of the composites suddenly decrease when the fiber content increases from 30% to 35%. Worse dispersion of high-content SiC_f in the matrix may be the main reason responsible for this phenomenon.

3.2. Dielectric properties

Dielectric performances of the composites with 2–4 mm SiC_f are shown in Figs. 5–7. It can be observed that the complex permittivities of the composites are closely correlated with the volume fraction of SiC_f. Both the real part (ε') and imaginary part (ε'') of the composites rise across the whole frequency range with the increasing SiC_f content.

The ε' of permittivity is known as an expression of the polarization ability of materials. In this case, the increase of ε' could be mainly attributed to the interface polarization as a result of the formation of conductor/dielectric interface. As discussed above, the SiC_f used in this research are covered by a semiconductive carbon-rich layer [15]. When subjected to an alternating electrical field, the moveable electrons such as free electrons and weakly bound electrons in the semiconductive carbon-rich layer rapidly respond and gather along the SiC_f/LAS interface, contributing to the rise of ε'.

On the other hand, the ε'', which represents capacity of dielectric loss in the microwave frequency, is also associated with the increase of SiC_f content. When the excited electrons shift along the SiC_f, the microwave energy will be dissipated and converted to thermal energy in order to overcome the electrical resistance, which is characterized by ε''. Generally, the shifting electrons in the composites fall into two categories: one is polarized electrons constrained in single SiC_f, the other is hopping electrons moving among SiC_f. Thereby, the ε'' can be correspondingly expressed as follows:

$$\varepsilon'' = \varepsilon_p'' + \varepsilon_h'' \tag{2}$$

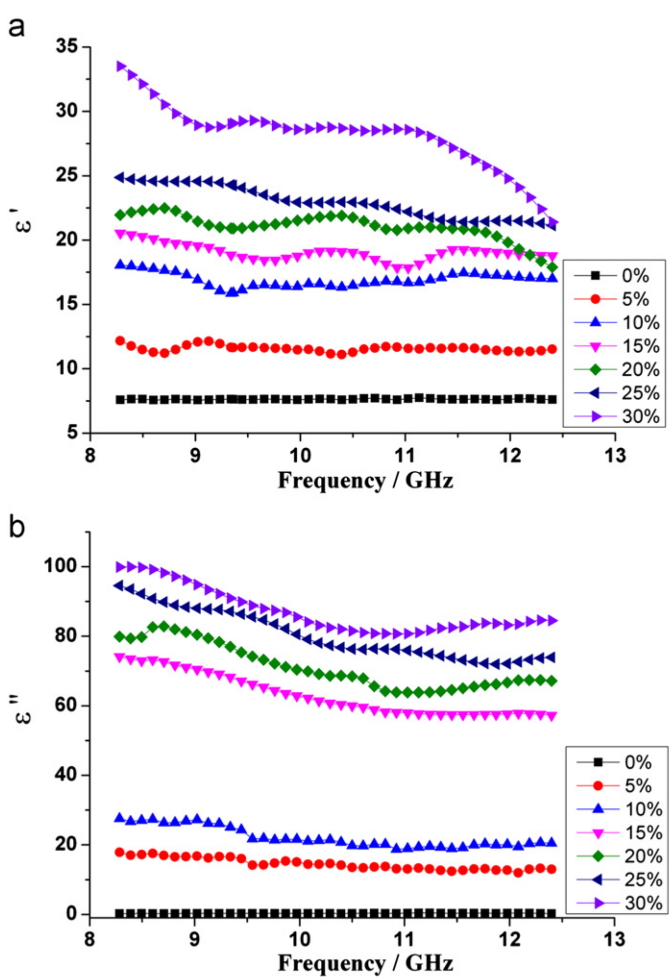

Fig. 5. (a) ε' and (b) ε'' of the composites filled with 2 mm SiC_f versus frequency.

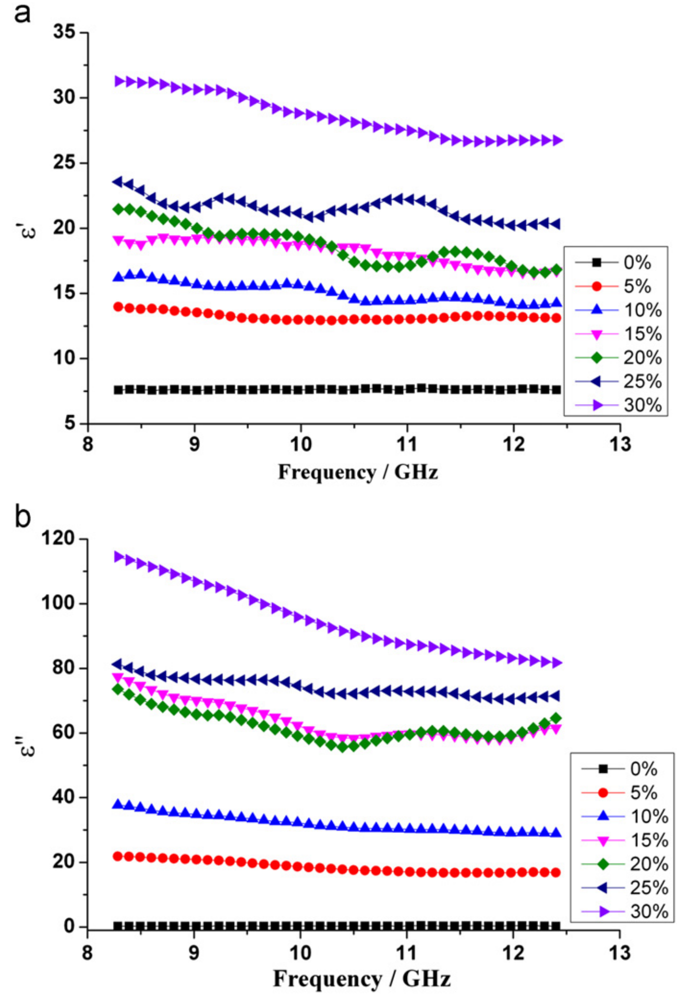

Fig. 6. (a) ε' and (b) ε'' of the composites filled with 3 mm SiC_f versus frequency.

For the composites with low-content SiC_f, the ε_h'' is small due to the well dispersion and low probability of contact between SiC_f. However, with the increase of SiC_f content, the average distances among the SiC_f are gradually shortened with consequent rise of contact probability between the SiC_f. Also, more energy conversion from microwave to thermal energy when the hopping electrons pass through the joining points of SiC_f. As a result, a rapidly increase of ε_h'' occurs when the SiC_f content is up to a critical value i.e. percolation threshold, which is 15–20% in the current research as presented in Figs. 5–7.

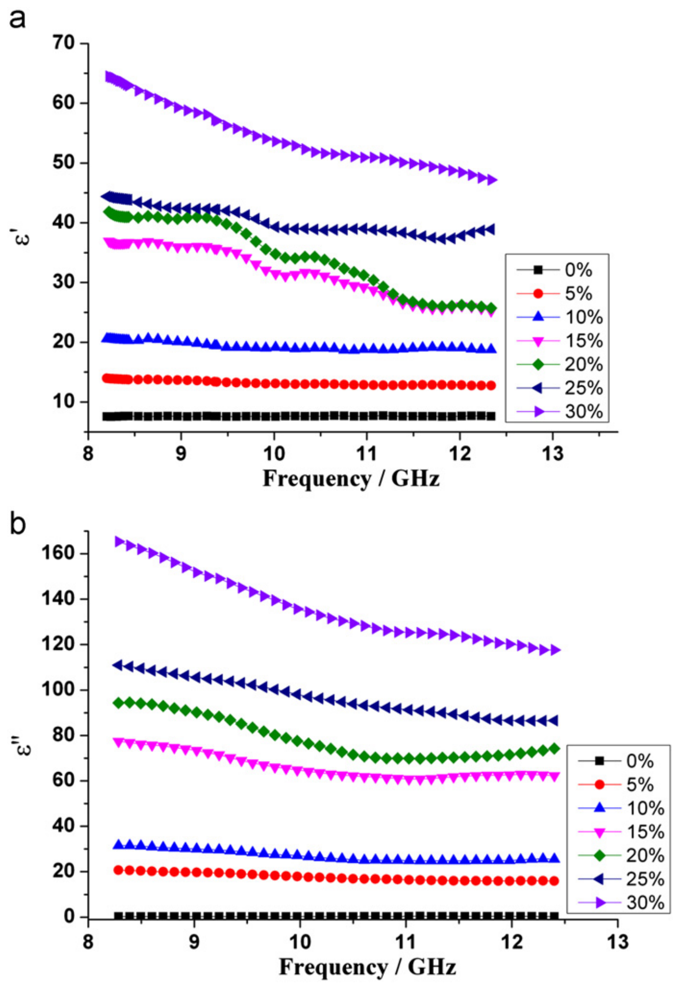

Fig. 7. (a) ε′ and (b) ε″ of the composites filled with 4 mm SiC_f versus frequency.

The ε″ at 8.2 GHz increase nearly two times as the SiC_f content increase from 15% to 20%.

Furthermore, effects of the fiber length on the complex permittivity are also significant for the composites filled with high-content SiC_f. As can be seen from Figs. 5–7, as the fiber length increases from 2 mm to 4 mm, both the ε′ and the ε″ of the composites with 20–30% SiC_f are remarkably enhanced. This can be explained by the higher probability of contact for longer fibers. When the moveable electrons pass through the joining points of the SiC fibers, more energy is dissipated than which shift within a single SiC fiber [17].

4. Conclusions

In summary, the hot-pressed sintered SiC_f/LAS composites exhibit flexible complex permittivity which vary from 12 to 35 for ε′ and 18 to 120 for ε″ at 8.2 GHz. The complex permittivity of the composites can be adjusted by simply choosing appropriate volume and length of SiC_f. In addition, mechanical properties of LAS glass ceramic are also enhanced by the addition of SiC_f. All these results indicated that SiC_f/LAS composite is a good candidate as structural microwave absorbing and microwave shielding materials.

Acknowledgments

This work was financially supported by the fund of the National Nature Science Foundation of China (No. 51072165) and the NPU Foundation for Fundamental Research (No.NPU-FFR-JC20110213).

References

[1] H. Liu, H. Cheng, J. Wang, G. Tang, R. Che, Q. Ma, Effects of the fiber surface characteristics on the interfacial microstructure and mechanical properties of the KD SiC fiber reinforced SiC matrix composites, Materials Science and Engineering A: Structure 525 (2009) 121–127.

[2] A. Demir, Effect of Nicalon SiC fibre heat treatment on short fibre reinforced β-sialon ceramics, Journal of the European Ceramic Society 32 (2012) 1405–1411.

[3] D. Sciti, R. Savino, L. Silvestroni, Aerothermal behaviour of a SiC fibre-reinforced ZrB_2 sharp component in supersonic regime, Journal of the European Ceramic Society 32 (2012) 1837–1845.

[4] D. Ding, W. Zhou, F. Luo, Y. Mu, D. Zhu, The effects of CVD SiC interphase on mechanical properties of KD-1 SiC fiber reinforced aluminum phosphate composites, Materials Science and Engineering A: Structure 534 (2012) 347–352.

[5] Y. Yang, B.E. Stucker, G.D. Janaki Ram, Mechanical properties and microstructures of SiC fiber-reinforced metal matrix composites made using ultrasonic consolidation, Journal of Composite Materials 44 (2010) 3179–3194.

[6] F. Luo, D.-m. Zhu, W.-c. Zhou, A two-layer dielectric absorber covering a wide frequency range, Ceramics International 33 (2007) 197–200.

[7] D. Ding, W. Zhou, B. Zhang, F. Luo, D. Zhu, Complex permittivity and microwave absorbing properties of SiC fiber woven fabrics, Journal of Materials Science 46 (2011) 2709–2714.

[8] H. Liu, H. Tian, Mechanical and microwave dielectric properties of SiC_f/SiC composites with BN interphase prepared by dip-coating process, Journal of the European Ceramic Society 32 (2012) 2505–2512.

[9] Q.-G. Fu, B.-L. Jia, H.-J. Li, K.-Z. Li, Y.-H. Chu, SiC nanowires reinforced MAS joint of SiC coated carbon/carbon composites to LAS glass ceramics, Materials Science and Engineering A: Structure 532 (2012) 255–259.

[10] Z. Huang, W. Zhou, X. Tang, P. Li, J. Zhu, Dielectric and mechanical properties of MoSi2/Al2O3 composites prepared by hot pressing, Journal of the American Ceramic Society 93 (2010) 3569–3572.

[11] Z. Huang, W. Zhou, R. Ma, X. Tang, F. Luo, J. Zhu, Dielectric and mechanical properties of hot-pressed sintered Csf/Al2O3 ceramic composites, International Journal of Applied Ceramic Technology 9 (2011) 413–420.

[12] T.-j. Hu, X.-d. Li, Y.-h. Li, H. Wang, J. Wang, Axial graded silicon carbide fibers with fluctuating carbon layer and sinusoidal electrical resistivity, Materials Letters 65 (2011) 2562–2564.

[13] T.-J. Hu, X.-D. Li, G.-Y. Li, Y.-H. Li, H. Wang, J. Wang, Axial graded carbon fiber and silicon carbide fiber with sinusoidal electrical resistivity, Journal of the American Ceramic Society 94 (2011) 2808–2811.

[14] E. Bouillon, D. Mocaer, J.F. Villeneuve, R. Pailler, R. Naslain, M. Monthioux, A. Oberlin, C. Guimon, G. Pfister, Composition–microstructure–property relationships in ceramic monofilaments

resulting from the pyrolysis of a polycarbosilane precursor at 800–400 °C, Journal of Materials Science 26 (1991) 1517–1530.
[15] T.-j. Hu, X.-d. Li, G.-y. Li, Y.-d. Wang, J. Wang, SiC fibers with controllable thickness of carbon layer prepared directly by preceramic polymer pyrolysis routes, Materials Science and Engineering B: Advanced 176 (2011) 706–710.
[16] N.S. Jacobson, Corrosion of silicon-based ceramics in combustion environments, Journal of the American Ceramic Society 76 (1993) 3–28.
[17] B. Chen, K. Wu, W. Yao, Conductivity of carbon fiber reinforced cement-based composites, Cement and Concrete Composites 26 (2004) 291–297.