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MICROWAVE PERMITTIVITY OF 1D-C/C/SIC COMPOSITES PREPARED BY FORCED-FLOW CHEMICAL VAPOR INFILTRATION

Donglin Zhao [1,2], Wancheng Zhou [1] , and Hongfeng Yin [1]

[1] *State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an 710072, China*

[2] *Institute of Nuclear Energy Technology, Tsinghua University, Beijing 100084, China*

SUMMARY: The carbon fiber is conductive material, and the SiC exhibits semiconducting properties. And the composites (C/PyC/SiC) with pyrolytic carbon interfacial layer exhibit good mechanical properties. So the C/PyC/SiC composites appear to be a good system for the study of problems associated with the preparation of ceramic matrix composites with tailored microwave properties. The composites (1D-C/PyC/SiC) with pyrolytic carbon interfacial layer were fabricated by forced-flow chemical vapor infiltration (FCVI), and the microstructure and microwave permittivity at the frequency range of 8.2~12.4GHz were investigated. A thin pyrolysis carbon layer (0.2± μm) was firstly deposited on the surface of carbon fiber as the interfacial layer with C_3H_6 at 850℃ and 0.1MPa. Methyltrichlorosilane (CH_3SiCl_3 or MTS) was used for the deposition of the silicon carbide matrix. The conditions used for SiC deposition are 1100℃, a hydrogen to MTS ratio of 10 and a pressure of 0.1MPa. The real part (ε′) and imaginary part (ε″) of the complex permittivity of the 1D-C/PyC/SiC composites are 11.53~12.44 and 1.18~2.08 respectively at the frequency range of 8.2~12.4GHz. The C/C/SiC composites would be a good candidate for microwave absorbent.

KEYWORDS : Microwave permittivity, Forced-flow chemical vapor infiltration, 1D-C/C/SiC

INTRODUCTION

Ceramic matrix composites (CMCs) is one of the most promising candidates for high temperature structural material, in which SiC matrix exhibits an excellent oxidation resistance because of the formation of a protective layer of silica which limits further oxidation [1]. CMCs with continuous fibers exhibit attractive properties such as low densities, high elastic moduli, and high strengths at elevated temperatures. They are considered as materials with a high potential in various fields of applications including engines and reentry thermal protection for space crafts. Chemical vapor infiltration (CVI) processed SiC matrix composites reinforced with carbon or SiC fiber have been designed and developed to improve the damage tolerance of inherently brittle SiC monolithic ceramics while retaining their high

stiffness, high-temperature strength, and oxidation resistance [1]. The composites (C/PyC/SiC) with pyrolytic carbon interfacial layer exhibit good mechanical properties and a typical failure behavior involving fiber pull-out and brittle fracture of sub-bundle [2]. The carbon fibers reinforced silicon carbide matrix composites have been studied in detail, but little data regarding the microwave permittivity of this kind of composites exists in literature. The carbon fiber is conductive material, and the SiC exhibits semiconducting properties. And the composites (C/PyC/SiC) with pyrolytic carbon interfacial layer exhibit good mechanical properties. So the C/PyC/SiC composites appear to be a good system for the study of problems associated with the preparation of ceramic matrix composites with tailored microwave properties. In this paper the microwave permittivity of 1D-C/PyC/SiC composites prepared by forced-flow chemical vapor infiltration has been investigated.

EXPERIMENTAL PROCEDURE

The 1D-C/PyC/SiC composites was fabricated by forced-flow chemical vapor infiltration. Forced-flow chemical vapor infiltration is a good route for the rapid densification of high-temperature composites primarily ceramic-matrix composites [3]. It is well known that the mechanical properties of CMCs depend upon the fiber/matrix bonding. So the interposition of a thin layer of a compliant material such as carbon or BN is deposited on the fibers [1]. In this study the carbon layer was employed as the interphase. An isothermal FCVI furnace (shown in Fig.1) was used to deposit carbon (interphase) and silicon carbide (matrix) on the carbon fibers. The carbon fiber utilized was T300 carbon fiber from the Nippon Toray Corporation. The carbon fibers (T300, 2K) were fixed on a graphite holder, which was located at the center of the hot zone of the graphite resistance furnace. A thin pyrolysis carbon layer (0.2± μm) was firstly deposited on the surface of carbon fiber as the interfacial layer with C_3H_6 at 850℃ and 0.1MPa. Methyltrichlorosilane (CH_3SiCl_3 or MTS) was used for the deposition of the silicon carbide matrix because it contains the same number of silicon and carbon atoms in one MTS molecule and thus can easily prepare stoichiometric silicon carbide. MTS, used as the precursor, is a liquid at room temperature (boiling point at 1 atm, 66.4℃). It was kept in a temperature-regulated container through which hydrogen was bubbled, carrying the MTS into a mixing chamber and then into the FCVI furnace. The conditions used for SiC deposition are 1100℃, a hydrogen to MTS ratio of 10 and a pressure of 0.1MPa. Argon was used as dilute gas to slow the reaction rate.

X-ray diffraction (XRD) measurements were made with a Rigaku D/Max-B diffractometer unit using Ni-filtered CuK_α radiation at a scanning rate of $0.5°\cdot sec^{-1}$ and scanning from 15° to 80° of 2θ. The surface morphology and cross-sectional microstructure of the 1D-C/PyC/SiC composites were observed using scanning electronic microscopy (SEM, Model JEOL 840).

The microwave permittivity of the 1D-C/PyC/SiC composites was measured by the method, which is based on measurements of the reflection and transmission moduli between 8.2 GHz and 12.4GHz, in the fundamental waveguide mode TE_{10}. The 1D-C/PyC/SiC composites was firstly chopped up. The resulting short C/PyC/SiC fibers had average length of 2~3 mm. And then they were pressed into a brass holder ($10.16\times22.86\times2mm^3$) which fills the rectangular waveguide. After calibrated with an intermediate of a short circuit and blank holder, reflection and transmission coefficients were obtained with the help of an automated measuring system

(HP8510B network analyzer). Both the real and imaginary parts of the permittivity were calculated.

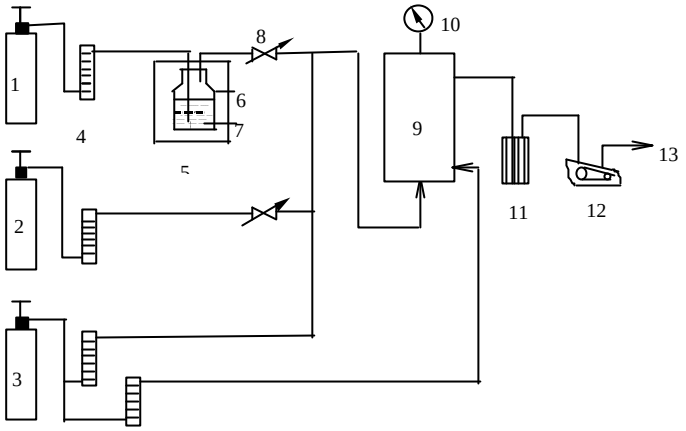


Fig. 1 Schematic diagram of forced-flow chemical vapor infiltration furnace
1. H_2; 2. C_3H_6; 3. Ar; 4. flow meter; 5. thermostatic box; 6. container of MTS; 7. MTS; 8. valve; 9. FCVI furnace; 10.pressure/vacuum meter; 11. cooling box; 12. vacuum pump; 13. waste gas

RESULTS AND DISCUSSION

XRD pattern of the 1D-C/PyC/SiC composites surface is shown in Fig. 2. Detailed analysis of the X-ray results indicated that the deposition are pyrolysis carbon and silicon carbide composed mainly of cubic (3C) type β-SiC with a small amount of 4H type α-SiC. It is clear that the diffraction angles of 35.6°, 60.1°, 72.1° and 75.5° correspond to β-SiC with cubic crystal structure and the diffraction angles of 33.7° corresponds to α-SiC with the hexagonal crystal structure [4]. According to the breadth of diffraction peaks, the crystallite sizes of silicon carbide was calculated from the Scherrer equation,

$$D = 0.89\lambda/\beta COS(\theta)$$

Where λ is the wavelength of characteristic X-ray, θ is the Bragg angle, and β is the calibrated width of the half-height of diffraction peaks. The crystallite sizes of silicon carbide is 10~15nm.

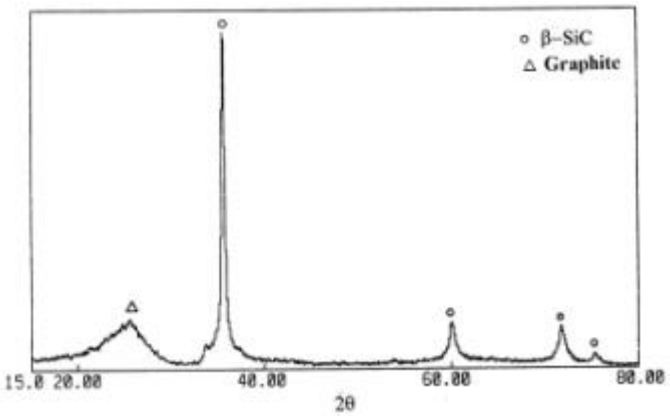
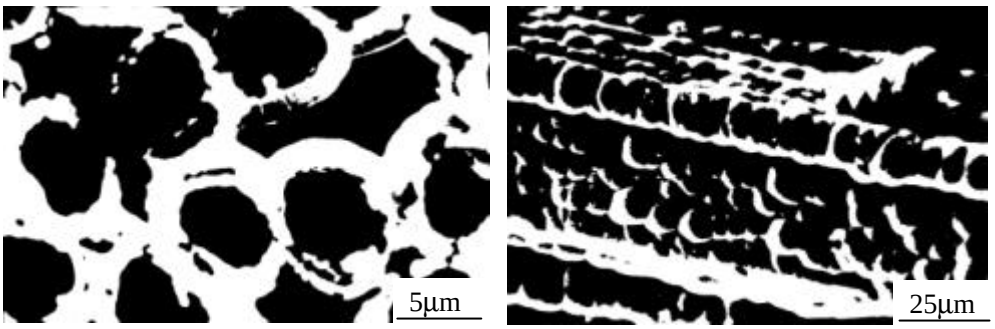


Fig. 2 XRD pattern of the 1D-C/PyC/SiC compsites surface

The morphology of the 1D-C/PyC/SiC composites is shown in Fig.3. The surface morphology of the 1D-C/PyC/SiC composites is shown in Fig.3 (a). It is observed that the SiC matrix is composed of a large number of spherical particles with a cloud-cluster shape. From the XRD pattern, it is easy to determine that each particle is an aggregate consisting of a large number of nanometric SiC crystallites. And it is observed from the transverse section of the 1D-C/PyC/SiC composites (Fig.3 (b)) that the thickness of the SiC deposition is 2~3.5μm.



(a) (b)

Fig. 3 SEM micrographs of 1D-C/PyC/SiC composites
(a) transverse section, (b) longitudinal surface

Electrical properties can be determined at various frequencies [5]. The interaction between electromagnetic waves and condensed matter can be described by using complex permittivity, ε_r ($\varepsilon_r=\varepsilon'+i\varepsilon''$, where ε' is the real part, ε'' the imaginary part), and conductivity, σ_r. The relation between the real part of the (polarization) conductivity $\sigma'(\omega)$ and the imaginary part of the permittivity $\varepsilon''(\omega)$ is $\sigma'(\omega)=\omega\varepsilon''(\omega)$, where ω is the angular frequency.

Fig.4 shows the real part (ε') and imaginary part (ε'') of the complex permittivity and loss tangent ($tg\delta=\varepsilon''/\varepsilon'$) of the 1D-C/PyC/SiC composites versus frequency. The ε' and ε'' are 11.53~12.44 and 1.18~2.08 respectively at the frequency range of 8.2~12.4GHz. The density of the sample is 1.96g/cm^3, The volume filling factor of carbon fiber is 27.68%. Carbon fibers are conductive materials that reflect the microwave directly. But it can be seen that the C/PyC/SiC composites can absorb microwave from Fig.4. So the C/PyC/SiC composites would be a good candidate for microwave absorbent.

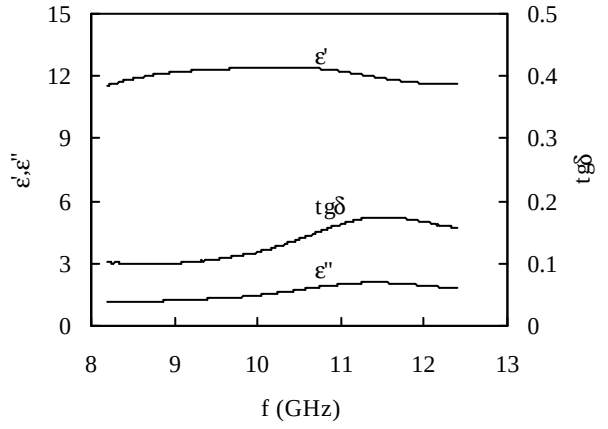


Fig. 4 The ε', ε'' and $tg\delta$ of 1-D C/PyC/SiC composites versus frequency

CONCLUSION

(1) The carbon fiber is conductive material, and the SiC exhibits semiconducting properties. And the composites (C/PyC/SiC) with pyrolytic carbon interfacial layer exhibit good mechanical properties. So the C/PyC/SiC composites appear to be a good system for the study of problems associated with the preparation of ceramic matrix composites with tailored microwave properties.

(2) The composites (1D-C/PyC/SiC) with pyrolytic carbon interfacial layer were fabricated by forced-flow chemical vapor infiltration. A thin pyrolysis carbon layer was firstly deposited on the surface of carbon fiber as the interfacial layer with C_3H_6 at 850℃ and 0.1MPa. Methyltrichlorosilane (CH_3SiCl_3 or MTS) was used for the deposition of the silicon carbide matrix. The conditions used for SiC deposition are 1100℃, a hydrogen to MTS ratio of 10 and a pressure of 0.1MPa.

(3) In the 1D-C/PyC/SiC composites, the thickness of carbon interphase is 0.2± μm, and the thickness of SiC deposition is 2~3.5μm. The SiC matrix is composed of β-SiC with cubic crystal structure and α-SiC with the hexagonal crystal structure. The crystallite sizes of silicon carbide is 10~15 nm.

(4) The real part (ε′) and imaginary part (ε″) of the complex permittivity of the 1D-C/PyC/SiC composites are 11.53~12.44 and 1.18~2.08 respectively at the frequency range of 8.2~12.4GHz. So the C/PyC/SiC composites would be a good candidate for microwave absorbent.

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