Carbon Nanotube/Fused Silica Composite with Gradient Structure and Carbon Fiber/ Fused Silica Back Reflection Layers for Microwave Absorbing Application

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ABSTRACT

A microwave absorbing material with laminated structure was designed and fabricated by hot-pressed sintering. Pure fused silica (SiO_2) was applied as upper face layer of the laminated composite in order to decrease the reflection to incident microwave. Two layers of SiO_2 matrix composites with 2.5 and 5 wt% carbon nanotubes (CNTs) were designed as microwave absorbing layers. Continuous carbon fiber (C_f)/SiO_2 was used as bottom layer to reflect the penetrating microwave. The results showed that the laminated composite was fabricated and all interfaces were clear. This unique four-layer composite had an improved microwave absorbing efficiency in X-band compared with the single-layer and two-layer composites of CNT/SiO_2 containing considerable content of CNTs.

Keywords: carbon nanotube, carbon fiber, layered composite, microwave absorbing properties

1 INTRODUCTION

The unique physical and mechanical properties of carbon nanotubes (CNTs) [1-4] combining with the high electrical resistivity and low density of pure silica (SiO_2) [5-7] make it attractive to prepare CNT/SiO_2 composite and investigate its corresponding properties. Attempts have been made to develop dense SiO_2 composites with varied contents of CNTs and the results show that the composites have improved mechanical properties [8, 9] and excellent microwave attenuation properties [5]. Further investigation reveals that microwave attenuation properties of the composite increase rapidly as the increasing content of CNTs [5]. However, the incorporation of CNTs will change the insulating properties of SiO_2 into metallic conductivity [5, 10], which in turn results in the increase of microwave reflection on the surface, and it is disadvantageous for the absorbing material. Fortunately, our previous work of calculation with transmission-line theory demonstrates the decrease of reflection at interface and the improvement of absorbing efficiency in the two-layer composite of CNT/SiO_2 [11]. Motojima et al. also reported multilayer absorbing composite of polyurethane based carbon material with a higher microwave absorptivity than that of single-layer composite [12]. So in order to achieve further absorbing efficiency, it is available to design and prepare CNT/SiO_2 composite with laminated gradient structure.

In addition, metal sheet is commonly used as a perfect reflection layer in the test of absorbing efficiency, which reminds us of the excellent reflection property of continuous carbon fiber (C_f) with 0^o cross angle to the electric field of incident microwave [13]. And it is coincidental that C_f has good physical and chemical compatibility with the SiO_2 matrix [14, 15]. Thus it is feasible to prepare an integral structure of multilayer CNT/SiO_2 composite with C_f/SiO_2 as back reflection layer and explore its corresponding properties. Hiza et al. reported similar multilayer microwave absorber based on organic materials, whose absorber was ferroelectric and ferromagnetic materials and the reflecting layer was composed of a metal or a C_f reinforced composite material [16]. However, the layers of their microwave absorber were bonded together into an integral structure by a silicone-type adhesive compound, which would increase the complexity and influence the properties of the multilayer microwave absorber. In our experiment, as all the laminated layers were based on SiO_2, it is possible to prepare the composite by one-step hot-pressed sintering at a proper temperature.

In this paper, we first described the design and preparation of multilayer absorbing composite composed of pure SiO_2, CNT/SiO_2 and C_f/SiO_2, and then investigated the laminated structure and microwave absorbing efficiency.

2 EXPERIMENTAL DETAILS

The multi-walled carbon nanotubes (MWCNTs, 20 to 40 nanometer in diameter, and 5 to 15 micrometer in length) provided by Shenzhen Nanoport Co. Ltd. of China were fabricated by catalytic pyrolysis of hydrocarbon and used as received. C_f (HTA 3K, Toray, Tokyo, Japan) was used to fabricate C_f/SiO_2 back reflection layer. Commercial SiO_2 powders were provided by Dahan Minerals (Xinyi) Co. Ltd., China. The aqueous solution of polyvinyl alcohol 124 prepared at 95 ^oC isothermal bath was selected as binder.

The SiO_2 slurry was prepared by a conventional method. Commercial SiO_2 powders were mixed with deionized water and binder followed by ball-milling for 24 h using agate balls as the grinding media. C_f was then infiltrated into the as-prepared SiO_2 slurry and dried naturally. After that, the obtained C_f/SiO_2 tapes were cut into 30×35 mm^2 green sheets (marked as GS-C_f-SiO_2).

A parallel experimental procedure was conducted as following: commercial SiO_2 powders with different content of MWCNTs were mixed with absolute alcohol (EtOH) and ball-milled for 24 h using agate balls as the grinding media. After dried at 100 ^oC, the powders were sieved through a 200 mesh screen and marked as EtOH-CNTs-SiO_2.

Finally, the GS-C_f-SiO_2 and EtOH-CNTs-SiO_2 with different content of MWCNTs were added into a graphite die orderly and hot-pressed at 1300 ^oC under an applied stress of 30MPa in N_2 atmosphere for 0.5 h. The obtained laminated composite was marked as C_1. The total thickness of the C_1 was about 5.8 mm and that of the laminated gradient CNT/SiO_2 layers with different content of MWCNTs was about 5 mm.

For comparison, a two-layer composite (marked as C_2) of CNT/SiO_2 was prepared as the same procedure as C_1, without the preparation of GS-C_f-SiO_2. The composite C_2 had one layer of pure SiO_2 and the other layer of CNT/SiO_2 with 10 wt% CNTs. The total thickness of composite C_2 was about 5 mm, and that of CNT/SiO_2 layer with 10 wt% CNTs was about 2.5 mm. Another reference composite (marked as C_3) appeared in our previous paper [5] was a single layer composite of CNT/SiO_2 with 5.8 wt% (6.25 vol%) CNTs and a thickness of 5 mm.

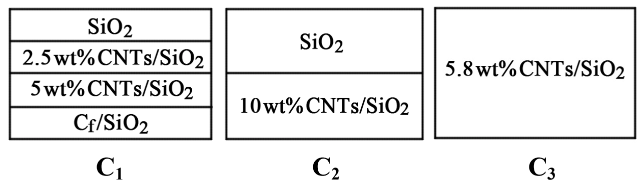

Fig. 1. Scheme of the composites C_1, C_2 and C_3.

The microstructure of the laminated absorbing composite (C_1) was observed by a MHO-2 Optical Microscope. The sample was cut into the size of 10 (length) ×4 (width) mm^2 and the optical photos of the cross-section were obtained in regular sequence along the direction of thickness. For microwave attenuation property measurements, the specimens were cut into the size of 22.86 (length) ×10.16 (width) mm^2 and conducted on a WILTRON 54169A Scalar Measurement System in X-band (8-12GHz) at room temperature.

3 RESULTS AND DISCUSSION

As shown in Fig. 1(C_1), a four-layer composite was designed and prepared in this paper. The first layer was composed of pure SiO_2, which was a good candidate for high-performance microwave penetrating material as SiO_2 has close impedance with air [5]. The second and third layer formed a laminated gradient structure for microwave absorbing, in which the content of CNTs was 2.5 and 5 wt%, respectively. The last layer was a back reflection layer of C_f/SiO_2.

The optical microstructure of the laminated microwave absorbing composite (C_1) could be seen from Fig. 2. The white thread-like C_fs, almost all of which ranked perpendicularly to the orientation of applied stress, were marked by arrows, as shown in Fig. 2(a). In the absorbing

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layers, different content of CNTs resulted in different optical microstructure. The interface of SiO_2 layer and 2.5 wt% CNT/SiO_2 layer, and that of 2.5 wt% CNT/SiO_2 layer and 5 wt% CNT/SiO_2 layer were clear.

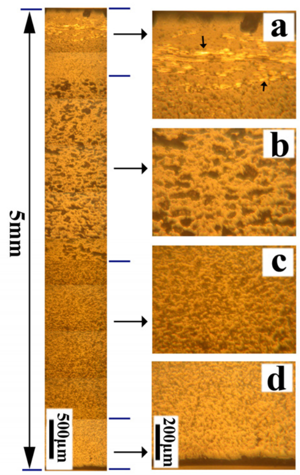

Fig.2. Representative optical microstructures of C_1 in different layers: (a) C_f/SiO_2, (b) 5wt% CNT/SiO_2, (c) 2.5 wt% CNT/SiO_2, (d) pure SiO_2.

The insertion loss (IL), a physical parameter expressing the total loss of microwave radiating on the samples, is usually considered as three parts: reflection, absorption, and multiple reflections [5, 17]. It is defined as:

$$IL=10\log (P_T/P_I), \quad (1)$$

where P_T and P_I are the power of the transmitted and incident microwave, respectively. The unit of the IL is decibel (dB). From the definition, one can conclude that the lower the IL value is, the better the microwave attenuation properties of the material are. Fig. 3 showed the microwave IL value of C_1 and C_2, where microwave was introduced from the side of pure SiO_2 layer entirely. As seen from Fig. 3, although the microwave IL value fluctuated to some extent with the increase of microwave frequency, all the values of C_1 were much lower than those of C_2. When the frequency was 10 GHz, the IL value of C_1 was -34.98 dB, much lower than that of C_2 (-24.34 dB) and that of C_3 (-25.76 dB) [5]. All these results demonstrated that the four-layer composite (C_1) had better microwave attenuation properties than the two-layer composite (C_2) and the single layer composite (C_3).

As the IL includes return loss(R) and absorption loss, in

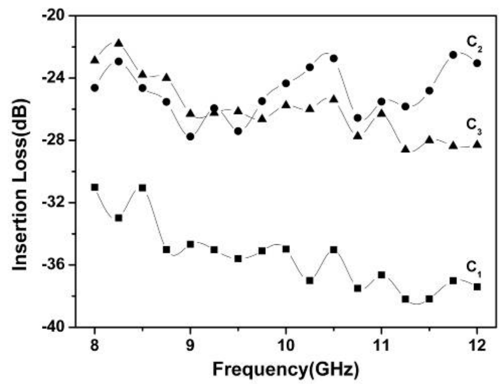

Fig.3. Microwave insertion loss of C_1, C_2 and C_3 as a function of frequency in X-band at room temperature.

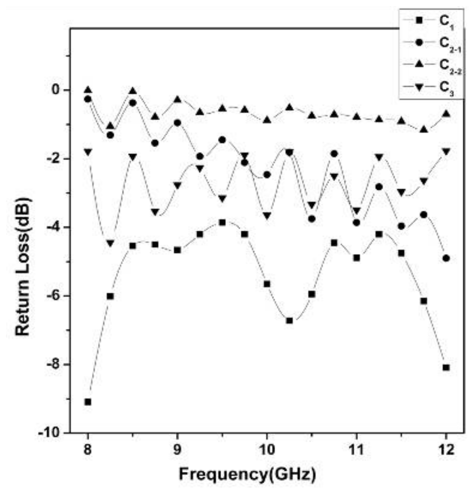

Fig.4. Microwave return loss of C_1, C_2 and C_3 with considerable content of CNTs as a function of frequency in X-band at room temperature.

order to compare the absorbing efficiency, the physical parameter R is necessarily introduced. It is defined as:

$$R=10\log (P_R/P_I), \quad (2)$$

where P_R and P_I are the power of the reflected and incident microwave, respectively. The unit of R is also dB. From this definition, one can conclude that the lower the R value is, the less the reflected microwave from the surface of sample is, and this is beneficial for the microwave absorption properties of the material. Fig. 4 represented the microwave R value of C_1, C_2 and C_3 as a function of frequency in X-band at room temperature. The microwave was introduced from the side of pure SiO_2 layer for the curve C_1, C_{2-1} and C_3. While for the curve C_{2-2}, the microwave was introduced from the side of CNT/SiO_2 layer with 10 wt% CNTs. As shown in Fig. 4, compared with C_2 and C_3, all the R values of C_1 were smaller in X-band, which indicated that less electromagnetic wave would be reflected from the surface of composite C_1. According to the impedance-matching principle, C_1 had the closer impedance with air. As to composite C_2, different incident layers for microwave resulted in different R values. When the microwave was introduced from the side of pure SiO_2 layer (curve C_{2-1}), the R values were smaller than those of the curve C_{2-2}, for which the microwave was introduced from the side of CNT/SiO_2 layer with 10 wt% CNTs. This result indicated two points: one was that the pure SiO_2 layer had closer impedance with air than that of the CNT/SiO_2 layer containing 10 wt% CNTs; the other was that the increase of CNT content would change the insulating properties of SiO_2 into metallic conductivity, which in turn resulted in the increase of microwave reflection on the surface. Furthermore, the gap between curve C_{2-1} and C_{2-2} increased as the frequency increased, which illuminated that the effect of impedance-matching increased with the increase of frequency.

From above analysis, it is clear that the laminated gradient absorbing layers with the back reflection layer of C_f/SiO_2 finally resulted in the high microwave attenuation properties and low reflection on the surface, thus this new structural composite had high microwave absorbing properties.

4 CONCLUSIONS

A multilayer absorbing composite was designed and fabricated by hot-pressed sintering. The four-layer composite had three gradient layers of CNT/SiO_2 with 0, 2.5 and 5 wt% CNTs and a last layer of C_f/SiO_2 as a back reflection layer. The unique composite exhibited high microwave attenuation properties and low reflection on the surface, i.e. high microwave absorbing properties. This kind of laminated composite might be a promising material for high microwave absorbing application.

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