Journal of Applied Polymer Science

Effect of Carbon Nanotube Dispersion on the Complex Permittivity and Absorption of Nanocomposites in 2–18 GHz Ranges

Ruey-Bin Yang,[1] Wen-Shyong Kuo,[1] Heng-Chih Lai[2]
[1]Department of Aerospace and Systems Engineering, Feng Chia University, Taichung 407, Taiwan
[2]Graduate Institute of Mechanical and Aeronautical Engineering, Feng Chia University, Taichung 407, Taiwan
Correspondence to: R. B. Yang (E-mail: rbyang@fcu.edu.tw) and W. S. Kuo (E-mail: wskuo@fcu.edu.tw)

ABSTRACT: The effect of dispersion processes on complex permittivity ($\varepsilon' - j\varepsilon''$) and microwave absorption in 2−18 GHz ranges is presented for nanocomposites loaded with different amounts of carbon nanotubes (CNTs) ranging from 1 to 5 wt %. The ultrasonic sonication (US) method and the three-roll mill (TRM) method were performed for the manufacturing of the CNT/epoxy absorbers with different dispersing levels. Microscopic observations revealed that the CNT agglomerates were reduced after the TRM process, and individual CNTs were uniformly dispersed in the epoxy resin. For the same weight content of CNT fillers, the percentage increase of ε' between US and TRM samples varies from 35.5% to 101.7% while the corresponding increment of ε'' (dielectric loss) varies from 79.6% to 248.8%. A minimum reflection loss for the US sample with 2 wt % CNTs is only −7.8 dB at 11.3 GHz while the corresponding TRM sample is greatly improved to reach −37.4 dB at 7.76 GHz for the same matching thickness of 3 mm. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40963.

KEYWORDS: composites; dielectric properties; graphene and fullerenes; nanotubes

Received 29 September 2013; accepted 4 May 2014
DOI: 10.1002/app.40963

INTRODUCTION

In recent years, microwave absorbing materials (2−18 GHz) have been intensively investigated not only for military purposes such as radar cross section reduction and stealth technology, but for electronics and wireless communication industries for reducing electromagnetic interference (EMI). Microwave absorbers can also be used to prevent potential health hazards due to exposure to electromagnetic wave radiation.

Traditional magnetic ferrite absorbers offer good absorption performance at low frequency from 30 MHz to 1 GHz.[1] Recent development in microwave absorber technology has resulted in microwave absorbers with strong absorption, thin thickness, light weight, low cost, and wide absorbing bandwidths. This goal can be achieved by mixing or blending dielectric/magnetic fillers with nonmagnetic polymeric resins to fabricate composite absorbers with various concentration of the filler absorbent.[2] The composite materials filled with magnetic metal particles such as carbonyl iron, nickel particles, and others can be used in EMI suppression and/or microwave absorbers at higher frequencies for their relatively low electrical conductivities, high Curie temperatures, good temperature stabilization, and high specific saturation magnetization intensities.[3] However, the magnetic ferrites and iron powder with a high specific mass are relatively heavy which is a shortcoming possibly hindering their applications in aerodynamic performance of stealth jetfighters.

Because of the relatively light weight, carbon materials are widely used as microwave absorbents because of their corrosion resistance and good conductivity. In the past decade, much attention has been paid to nanocarbon materials, including fullerenes, carbon nanotubes (CNTs), carbon nanofibers, and carbon nanoparticles.[4] CNTs are composed of graphene layers rolled into a cylindrical form. Depending on the rolling direction, the CNTs can be either metallic or semiconductive.[5] CNTs have been studied extensively due to their exceptional mechanical properties and interesting electric behavior.[6] They are usually used as fillers to polymers to make CNT-reinforced nanocomposites which have the potential to revolutionize structural materials for aerospace, electrical and thermal conductors for energy applications, EMI shielding, and microwave absorbing materials.[7] CNTs are naturally entangled in the form of agglomerates which are usually too dense to allow a complete resin impregnation. Without the surrounding matrix for consolidation, most individual CNTs within the agglomerates become ineffective in bearing loads or other functions such as heat and electric transfer.[8] To achieve a nanocomposite able to reflect the advantages of CNTs, properly dispersing CNTs is indispensable. Many techniques have been developed to the homogeneous dispersion of CNTs in the polymer matrix, such as high-speed stirring,[9] ultrasonic sonication (US),[10,11] and three-roll mill (TRM) calendaring method[12–14] to enhance mechanical and electrical properties of the composites.[15]

CNTs have many advantages such as strong microwave absorption properties in a wide gigahertz frequency range, low density, good compatibility, and corrosion resistance which make CNTs ideal materials for EMI shielding and microwave absorption.[16] Although the use of CNTs on the EMI shielding and microwave absorbing materials has been conducted in the literature,[17–21] most of the dispersion methods employed are either US or mechanical stirring methods which may form agglomerates and may not reflect the advantage of CNTs' microwave properties. To the best of authors' knowledge, the effect of dispersion and microstructures of CNTs on the complex permittivity and microwave absorptivity of CNT/epoxy composite absorbers in 2−18 GHz has rarely been revealed and investigated. In this article, we use two different methods, i.e., TRM and US respectively to prepare different CNT/epoxy nanocomposites and study the effect of dispersion processes on their complex permittivity ($\varepsilon' - j\varepsilon''$) and microwave absorption in the frequency range of 2−18 GHz, and the possible mechanism is also discussed.

EXPERIMENTAL

Preparation of CNT/Epoxy Composites

The CNTs employed in the present work was supplied by Advanced Nanopower, Taiwan. The CNTs are multiwalled, with typically 50–60 μm in length and 40–50 nm in diameter. The matrix material was a low-viscosity, room-temperature type epoxy resin, supplied by Te-Wang Chemistry, Taiwan. The weight ratio between the epoxy and the hardener is 3 : 1, and the curing time is 24 h. The first dispersion method was to use a sonicator to disperse CNTs. The sonicator was a probe type with 1500 W in power and working at 20 kHz. The vibration of the probe tip caused rapid agitation to the mixture and the CNT agglomerates were attached by the shock waves. Five sample materials were made with CNT weight fractions 1, 2, 3, 4, and 5 wt % and coded as US-1, US-2, US-3, US-4, and US-5, respectively. The processing of sonication lasted for 15 min. After the mixing process is completed the mixture is placed into a steel mold to fabricate toroid-shaped composite specimens with an inner diameter of 3.04 mm, an outer diameter of 7.00 mm, and 2–4 mm in thickness for transmission/reflection measurements. The mold with the mixture was cured for 24 h at 25°C and post-cured for 2 h at 80°C.

The second dispersion process was the use of three-roll mill which provided a high shear-strain rate to separate the CNTs from the agglomerates. Figure 1 schematically shows the three-roll mill. The first roll (the feed roll) and the third roll (the apron roll) rotate in the same direction, while the second roll (the middle roll) rotates in the opposite direction. The rotational speeds are adjustable, but the speed ratios are fixed ($\omega_1{:}\omega_2{:}\omega_3 = 1 : 3 : 9$). The speed in the apron roll was set at 540 rpm. The gap between rolls is adjustable from 200 μm to 3 μm. The different speeds in adjacent rolls give rise to a shear-strain rate in the gap, which in turn leads to a shear stress in

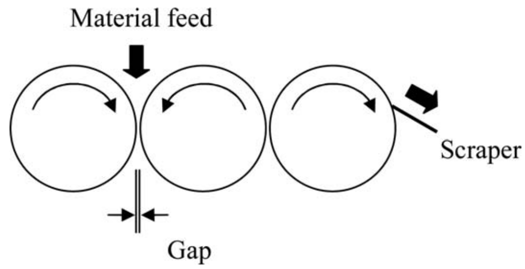

Figure 1. Schematic diagram of the three-roll milling apparatus.

the mixture when it passes through the gap. The shear stress can pull some CNTs from the agglomerate, and the size of the agglomerate is reduced each time it passes the gap. To increase the shear stress, one can achieve it by increasing the rotational speed or by reducing the gap. The former relies on the milling facility and is difficult to accomplish due to high-speed induced vibration problems. The latter depends on the accuracy of the gap control. The collected mixture was sent back to the first roll for another cycle of processing. The gap setting controls the shear stress and the processing time required to complete a cycle. In the present study, the sequential gap settings were 50, 30, 20, 10, and 5 μm, respectively, and five cycles of milling were repeated for each gap setting. The dispersion mechanism is to pull off individual CNTs from the agglomerate through the action of the high shear stress, making the agglomerate smaller or vanishing. Once CNTs are torn apart from the agglomerate, it was soon surrounded and wetted by the epoxy resin to avoid them to be gathered again through van der Waals interactions. The curing process was the same as the sonicated composite. Five sample materials were processed at 1, 2, 3, 4, 5% CNT weight fractions and denoted as TRM-1, TRM-2, TRM-3, TRM-4, and TRM-5, respectively. Beyond 5 wt % of CNT fillers, the processing becomes difficult as the mixture is too viscous to flow properly.

Electromagnetic Characterization of Nanocomposites

After the toroid-shaped specimens were obtained from the US and TRM processing, the effective complex dielectric permittivity ($\varepsilon' - j\varepsilon''$) and complex magnetic permeability ($\mu' - j\mu''$) of the CNT/epoxy composite samples in 2–18 GHz frequency range were measured by using the transmission/reflection method with an Agilent 8510C vector network analyzer and an Agilent coaxial transmission airline (850151-60010) with characteristic impedance of 50 Ω. The measurement system for the electromagnetic properties is shown in Figure 2. The specimen under test was closely inserted between the inner and outer conductor standard of the coaxial airline. Calibration based on Agilent calibration kit (85050C) was performed before the experiments. In this method, the effect of air gap between the specimen and coaxial airlines is considered and corrected to reduce the measuring error with the transmission line. From the measured complex transmission and reflection scattering parameters (S_{11} and S_{21}), the frequency dependence of ε', ε'', μ', and μ'' for each sample was then obtained using Agilent 85071E material measurement software which is based on Nicolson–Ross-Weir's reflection/transmission formulation using scattering parameters of the reflected and transmitted transverse electromagnetic (TEM) waves in the

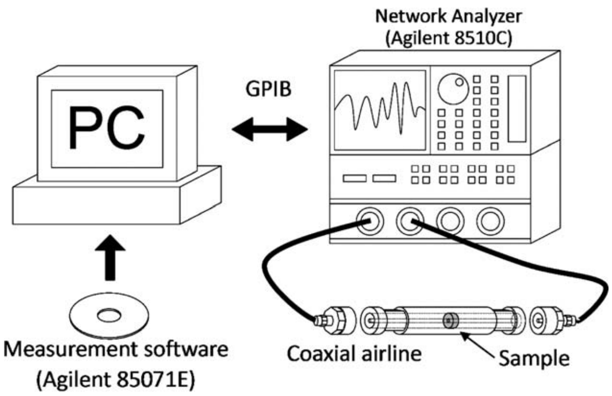

Figure 2. Coaxial airline measurement system.

frequency range 2–18 GHz.[21] The effects of air gap between the sample and holder were automatically corrected by the built-in algorithm of Agilent 85071E when the actual size of the sample was input into the software. In the present study, the CNT/epoxy composite is a nonmagnetic material and the measurement shows that the features of μ' is nearly equal to 1 and μ'' is very close to zero despite the consequences of frequency and concentration of CNTs. It is then suggested that the microwave absorption enhancement of CNT/epoxy composites results mainly from dielectric loss rather than magnetic loss.

Based on a generalized transmission line theory, the reflection loss (R_L) of normal incident electromagnetic waves of CNT/epoxy composites backed by a metal plate can be calculated from the measured complex relative permittivity and permeability data for the given frequency and absorber thickness d by using the following equations:

$$Z_{\text{in}} = \sqrt{\frac{\mu}{\varepsilon}}\tanh\left[j\frac{2\pi f d}{c}\sqrt{\varepsilon\mu}\right] \tag{1}$$

$$R_L\ (\text{dB}) = 20\log\left|\frac{Z_{\text{in}}-1}{Z_{\text{in}}+1}\right| \tag{2}$$

where ε and μ are the relative permittivity and permeability, respectively, of the composite medium. Z_{in} is the input impedance normalized with respect to the impedance of the free space Z_0, f is the frequency of the electromagnetic wave, and c is the velocity of electromagnetic waves in free space. For the metal-backed absorber, the equation for impedance matching (zero reflected condition) is expressed as:

$$\sqrt{\frac{\mu}{\varepsilon}}\tanh\left[j\frac{2\pi f d}{c}\sqrt{\varepsilon\mu}\right] = 1 \tag{3}$$

The perfect matching condition for no reflection requires $Z_{\text{in}} = 1$ which can be determined by the combination of the six characteristic parameter f, d, ε', ε'', μ', and μ''. The matching frequency f_m of the incident wave and matching thickness d_m of the composite material on a metal plate can be determined by using eqs. (1) and (2).

RESULTS AND DISCUSSION

Dispersion of CNTs in Polymer

After the CNT/epoxy composite was cured, the sample material was cut and the condition of CNT dispersion was examined by a scanning electron microscope (SEM). Figure 3(a) shows a typical micrograph of a sonicated composite with 5 wt % of CNT fillers. After 15-min sonication, the agglomerates were significantly reduced in size and the largest ones were reduced from about 100–200 μm in the originally supplied form to about 5–20 μm. Most agglomerates are in the range of 5–10 μm. In theory, the CNTs can be better dispersed and the agglomerate sizes further reduced by extending the sonication time or by adding solvents into the mixture. However, the factors to be considered include the heat generated by sonication and the potential damage to CNTs due to the prolonged high-energy impulses. Adding solvents is an option to reduce the viscosity of the resin which helps CNT dispersion during sonication. However, the solvent must be completely removed after the dispersion process; otherwise the residual solvent can adversely affect the properties of the composites.

Figure 3(b) is a typical section of the three-roll milled sample (TRM-5). The noticeable agglomerates are virtually nonexistent. The high-shear in the milling has been proven effective in pulling individual CNTs from agglomerates. The CNT length is a critical factor affecting the CNT dispersion. Long CNTs, for instance 100 μm in length, can have an aspect ratio higher than 2000, and they can be highly entangled with one another. In that case, the CNTs on the outer agglomerate can be pulled to break before pulled out from the group. In contrast to US process, a higher viscosity of the resin is desirable in the TRM, as it induces a higher shear stress for the CNTs to be pulled out. For the present materials, the experimental results showed that the TRM is the preferred process for CNT dispersion.

Figures 3(c,d) compare the results of resin impregnation by both methods. Within an agglomerate, CNTs are densely entangled and the spaces between CNTs are too narrow for the resin to penetrate into CNTs by using typical resin-transfer methods, such as vacuum-assisted impregnation. Without a complete wetting, voids can be present inside the agglomerate, and some CNTs around the void are dry. Figure 3(c) shows a section of the fractured agglomerate with some voids in the US-5 sample. The incomplete wetting renders CNTs unusable in transferring physical quantities, such as stress, heat, and electricity. Figure 3(d) shows a typical section of the TRM-5 sample. Voids are virtually nonexistent, and the spaces between CNTs are entirely filled by resin. Obviously the complete resin impregnation cannot be achieved merely by offering a vacuum chamber. The TRM processing provides two mechanisms. First, the high shear stress can pullout most CNTs from the agglomerates. These separated CNTs will soon be surrounded and wetted by the resin. Second, when agglomerates pass through the small gap between rollers, the agglomerates are squeezed, and some trapped air can be extruded out while some resin pushed in. While the agglomerate becomes smaller due to the loss of CNTs, the trapped air is even easier to escape. The milling processes repeatedly squeeze-and-deform the agglomerate from various directions, allowing complete removal of the air and voids.

Complex Permittivity of CNT/Epoxy Composites

The reflection and attenuation characteristics of dielectric absorbers can be determined by the effective complex

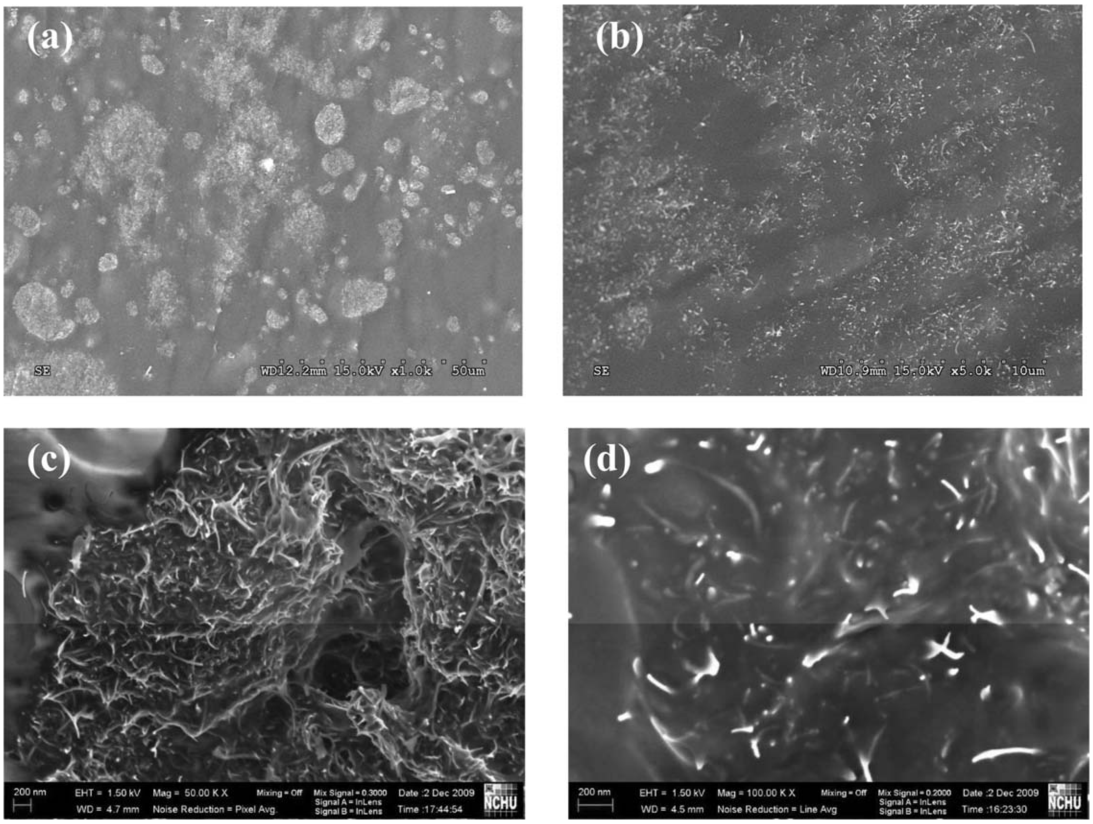

Figure 3. SEM micrographs of lower magnification showing CNT dispersion in (a) US-5 and (b) TRM-5 composites and higher magnification (c) US-5 and (d) TRM-5 composites.

permittivity ($\varepsilon' - j\varepsilon''$) of the composites, where the real part ε' is the effective relative dielectric constant, which is related to the stored energy within the medium. The imaginary part ε'' is related to the dissipation of energy and can be related to the effective electrical conductivity (σ) of the material by

$$\sigma = 2\pi f \varepsilon_0 \varepsilon'' \tag{4}$$

where ε_0 is the permittivity of free space.[22] The complex permittivity of the epoxy resin was measured as 2.8 – j0 while the complex permeability was 1.0 – j0 by the transmission/reflection method in the frequency range of 2–18 GHz.

Figure 4 shows the real (ε') and imaginary (ε'') parts of complex permittivity of the specimens as a function of frequency with CNT concentration 1–5 wt %. Both ε' and ε'' of the composites exhibit a similar trend with respect to frequency and CNT concentrations. All cases show that both ε' and ε'' increase with increasing CNT concentration which can be explained by the effective medium theory developed for an insulating matrix containing conductive fillers.[23] With a suitable effective medium formula, the effective macroscopic electromagnetic properties of the composite absorbers can be determined by the electromagnetic properties of each constituent and volume concentration of the fillers. In a general trend, an increase in conductive fillers, either in term of concentration or intrinsic conductivity, corresponds to an increase of both ε' and ε''. Accordingly, the permittivity of the all composites is higher than that of the pure resin because of the conductive effect of the CNT fillers. However, the measured complex permittivity is different for the composites prepared under different mixing and processing methods even with the same CNT loading contents. On the same filler content, the TRM samples show higher ε' and ε''than those in the US samples. The higher permittivity in the TRM samples can be attributed to a better dispersion of CNTs in the polymer matrix. This can be explained by the fact that after TRM processing, more conducting CNTs are isolated and wetted by insulating thin polymeric resin. As a result, the number of microcapacitors constituted by CNTs is increasing and the distances between CNTs are decreasing which are the main contributions to the significant increase in the permittivity of the CNT/epoxy composites.[24]

For the lowest CNT concentration (i.e., US-1 and TRM-1), both ε' and ε'' are relatively flat over the test frequency range, which shows insignificant variations with frequency for the composites with weight percentage equal to 1 wt %. However, for composites with weight percentage equal to 2 or 3 wt %, ε' and ε'' show a trend of monotonically decreasing with increasing frequency. For CNT concentrations greater than 3 wt %, ε' and ε'' exhibit a small fluctuation as the frequency increases. It should be noted that a dramatic decrease with increasing frequency (especially in the low frequency range) was observed for ε' and ε'' in the TRM-5 samples. Such remarkable frequency dispersion characteristic is advantageous for the impedance matching design of the incident waves and can be utilized in the design of wide-band absorbers.

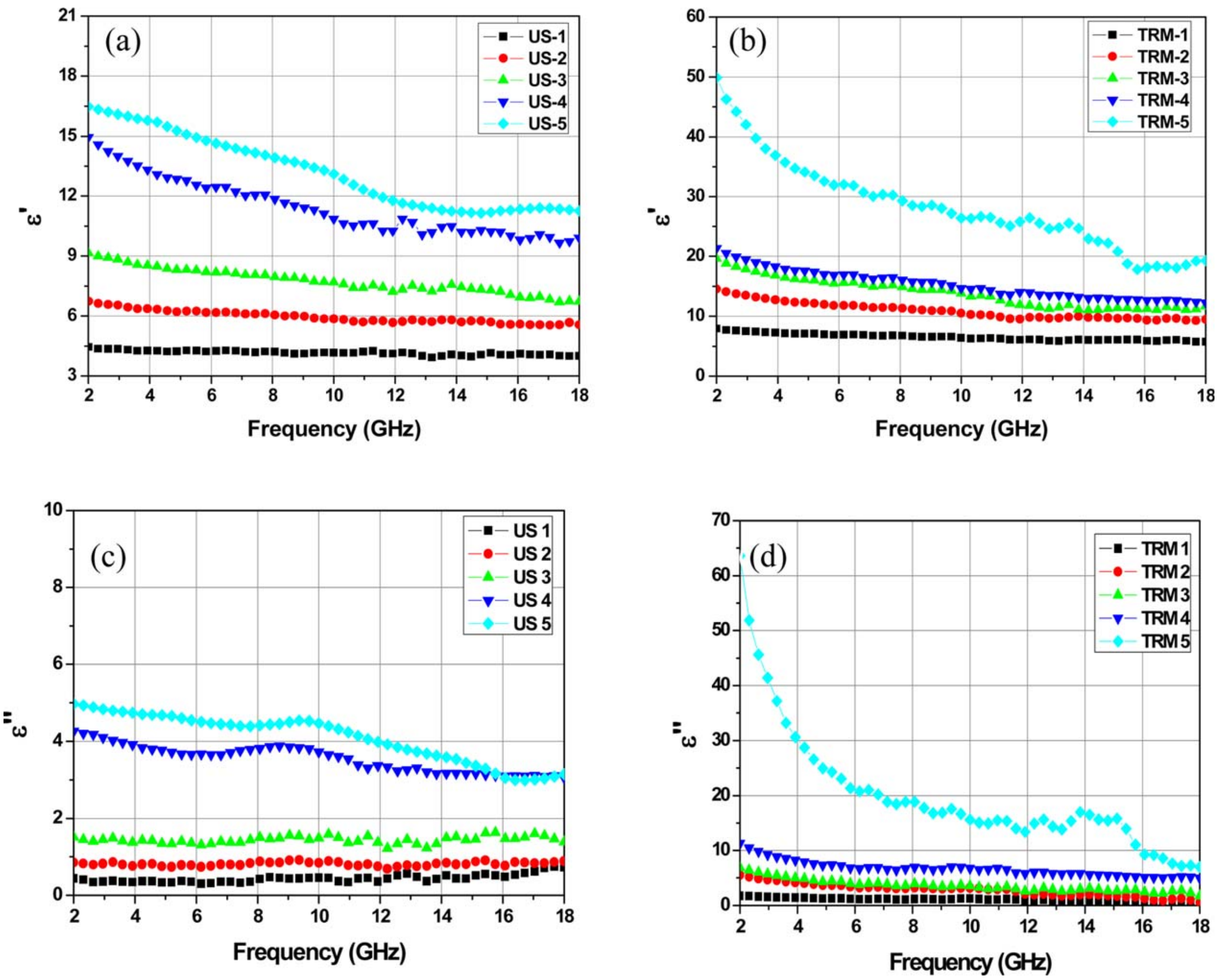

Figure 4. Frequency dependence of the real part of the permittivity by (a) US and (b) TRM, imaginary part of the permittivity by (c) US and (d) TRM with different loadings. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Comparing the US-5, the TRM-5 samples show a considerable increase in both ε' and ε''. This can be attributed to the formation of a continuous path of conductive fillers according to the SEM micrograph shown in Figure 3(c). Such critical filler concentration (5 wt % for TRM composites) can be termed as the percolation threshold. For comparison, Table I lists the values of ε' and ε'' for various US and TRM samples measured at 10 GHz which corresponds to the center frequency within the X-band. It is important to note that on the same filler wt % basis, the TRM samples show higher real and imaginary parts of permittivity than those of US samples. It is observed that for CNT filler from 1 to 5 wt %, the percentage increment of ε' between US and TRM samples varies from 35.5% to 101.7% while the corresponding increment of ε'' between US and TRM samples varies from 79.6% to 278.6%. The value of ε'', relating to the dissipation and conductivity of the medium, is affected more by the dispersion state of the CNT fillers than that of ε' which is related to the capacity of energy storage. The permittivity of the nanocomposites depends on the dispersion of the conductive fillers during composite processing. The results indicate that the TRM processing is an effective method of improving the dispersion of unmodified CNTs in a polymer matrix.

The dielectric loss tangent ($\tan \delta_e = \varepsilon''/\varepsilon'$), which represents the phase lag of the dipole oscillations with respect to the applied electric field, is associated with the magnitude of energy dissipation that can convert the incident microwave energy into thermal energy within the dielectric medium. The frequency dependence of the dielectric loss tangent for US and TRM samples with different loadings can be obtained by $\varepsilon''/\varepsilon'$. It is observed that the values of dielectric loss for TRM samples are much higher than those of US counterpart at 2−18 GHz. In the whole frequency range and for all specimens, the dielectric loss $\tan \delta_e$ increases with filler loading except there is a small intersection in the high frequency range. In particular, the magnitude of $\tan \delta_e$ for the TRM-5 composite is as high as 1.3 at 2 GHz. The dramatic enhancement of complex permittivity is primarily ascribed to the improved dispersion and interfacial bonding of the CNTs. The high dielectric loss is the main element leading to high attenuation of the electromagnetic wave.

Microwave Absorption Characteristics

In the fabrication of CNT/epoxy composites, we can significantly vary the frequency dependent permittivity ε' and dielectric loss ε'' by altering the CNT concentration as well as using

Table I. The Influence of Dispersion Processes on the Complex Permittivity of CNT/Epoxy Absorbers at 10 GHz

Sample	US (ε')	TRM (ε')	$\Delta\varepsilon'/\varepsilon'_{US}$ (%)	US (ε'')	TRM (ε'')	$\Delta\varepsilon''/\varepsilon''_{US}$ (%)
1 wt %	4.15	6.36	53.3	0.45	1.23	173.3
2 wt %	5.85	10.5	79.1	0.84	3.18	278.6
3 wt %	7.69	13.8	79.6	1.49	3.56	138.9
4 wt %	10.9	14.7	35.5	3.73	6.70	79.6
5 wt %	13.1	26.4	101.7	4.47	15.6	248.7

different dispersion processes. These advantages can be employed in the design of single- or multi-layer absorbers based on the transmission-line theory and the optimum matching parameters such as frequency, bandwidth, and the thickness of absorbers can be obtained by using standard numerical optimization techniques.

With the obtained complex permittivity from the coaxial line measurement, the reflection loss of can then be calculated at a given frequency with various absorber thicknesses and CNT concentration according to eqs. (1) and (2). Figure 5(a,b) show the reflection loss of the US and TRM samples with a thickness of 3 mm. A general trend observed is that the minimum reflection loss shifts to lower frequencies with an increasing CNT concentration due to the increase of the permittivity of the absorbers. The US composite with a thickness of 3 mm and 4 wt % shows the minimum reflection loss of −30.6 dB at 7.44 GHz and the frequency bandwidth corresponding to the reflection loss below −10 dB is 1.98 GHz (6.61−8.59 GHz).

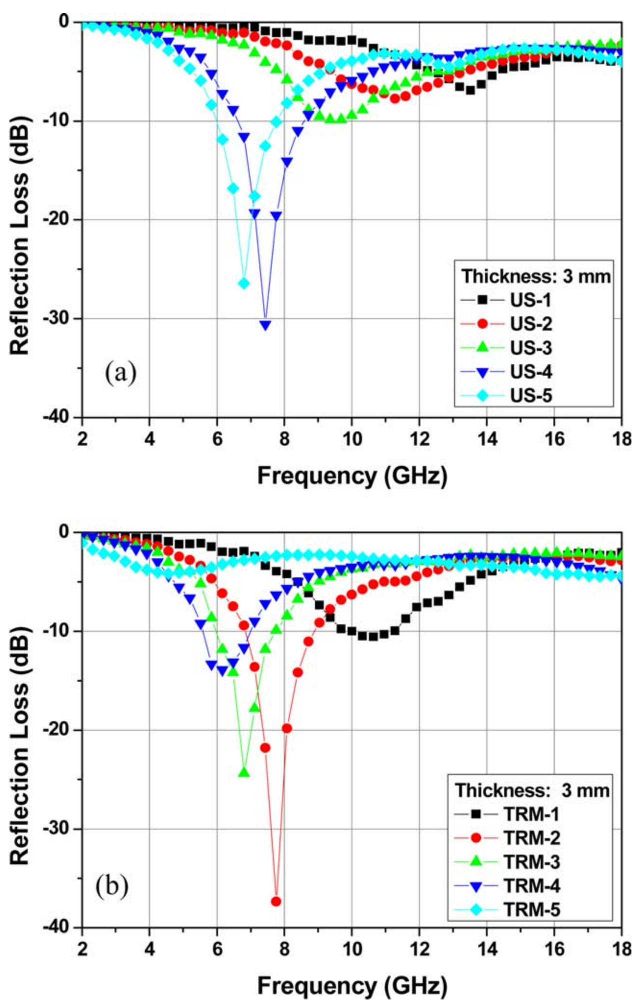

Figure 5. Absorption characteristics of the CNT/epoxy composites by (a) sonication and (b) three-roll milling. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

For the metal-backed absorber with same thickness, the matching frequency (f_m) and matching thickness (d_m) can be determined by using eq. (3). For a composite absorber with same thickness, it is observed that the matching frequency (f_m) shifts to lower frequencies with increasing weight percentage of absorbent fillers. When the three-roll mill is used in the dispersion process, the microwave absorption properties of the TRM composites with CNT concentration from 1 to 5 wt % for 3-mm thickness are shown in Figure 5(b). A superior microwave absorption performance of the TRM sample is observed. The lowest reflection loss of −37.4 dB is found at 7.76 GHz for the 2 wt % TRM sample with a thickness of 3 mm which shows a decrease of 26.1 dB compared with the US-2 counterpart. The frequency bandwidth of TRM-2 absorbers for the reflection loss below −10 dB is 2.06 GHz covering the frequency range from 6.84 to 8.90 GHz. Comparing the US-4, the TRM-2 contains fewer CNTs (only 2 wt %) but provides a lower reflection loss. From impedance matching equation, larger values of ε' and ε'' do not guarantee higher absorption. For 3-mm thickness samples, the complex permittivity of TRM-2 performs better impedance matching than the other samples. This indicates the importance of dispersion and concentration of CNTs in the microwave characteristics of the CNT/epoxy composite absorbers.

CONCLUSIONS

The effect of the dispersion of the unmodified CNTs on the electromagnetic and absorption properties was investigated by using two physical blending processes, i.e., sonication and three-roll milling methods. The results indicate that the high-shear TRM processing is an effective method in pulling individual CNTs from agglomerates. The dispersion of pristine CNTs is greatly improved by using TRM processing and a homogenous dispersion is successfully achieved. Electromagnetic tests show that the complex permittivity and microwave absorbing performance of the composite absorber with fine CNT dispersion by the TRM method are much higher than those of the US counterpart in the frequency range of 2−18 GHz. For a typical

3 mm-thick composite, the matching frequency f_m for both the TRM and US samples shifts to lower frequencies as the CNT concentration increases. The reflection loss reaches its highest value of −30.6 dB at 7.44 GHz for the US composites with 4 wt %. On the other hand, the minimum reflection loss for the TRM composites, taking place at 2 wt % at 7.76 GHz, is −37.4 dB while the corresponding US-2 sample is only 11.3 dB. The results indicate that the complex permittivity and absorbing performance of CNT/epoxy composite absorbers are sensitive to the level of CNT dispersion within the polymer matrix, and the electromagnetic properties can be improved significantly by using the TRM method in the processing of these nanocomposites.

REFERENCES

1. Dosoudil, R.; Usakova, M.; Franek, J.; Slama, J.; Olah, V. *J. Magn. Magn. Mater.* **2006**, *304*, 755.
2. Yang, R. B.; Liang, W. F. *J. Appl. Phys.* **2011**, *109*, 07A311.
3. Liang, W. F.; Yang, R. B.; Lin, W. S.; Jian, Z. J.; Tsay, C. Y. ; Wu, S. H.; Lin, H. M.; Choi, S. T.; Lin, C. K. *J. Appl. Phys.* **2012**, *111*, 07B545.
4. Saib, A.; Bednarz, L.; Daussin, R.; Bailly, C.; Lou, X.; Thomassin J. M.; Pagnoulle, C.; Detrembleur, C.; Jerome, R.; Huynen, I. *IEEE Trans. Microw. Theory Tech.* **2006**, *54*, 2745.
5. Cabria, I.; Mintmire, J. W.; White, C. T. *Phys. Rev. B* **2003**, *67*, 121406.
6. Thostenson, E. T.; Ren, Z. F.; Chou, T. W. *Compos. Sci. Technol.* **2001**, *61*, 1899.
7. Yu, S. Z.; Wong, W. M.; Hu, X.; Juay, Y. K. *J. Appl. Polym. Sci.* **2009**, *113*, 3477.
8. Ma, P. C.; Siddiqui, N. A.; Marom, G.; Kim, J. K. *Compos. A* **2010**, *41*, 1345.
9. Sandler, J.; Shaffer, M. S. P.; Prasse, T.; Bauhofer, W.; Schulte, K.; Windle, A. H. *Polymer* **1999**, *40*, 5967.
10. Lu, K. L.; Lago, R. M.; Chen, Y. K.; Green, M. L. H.; Harris, P. J. F.; Tsang, S. C. *Carbon* **1996**, *34*, 814.
11. Mukhopadhyay, K.; Dwivedi, C. D.; Mathur, G. N. *Carbon* **2002**, *40*, 1373.
12. Thostenson, E. T.; Chou, T. W. *Carbon* **2006**, *44*, 3022.
13. Lo, T. S.; Kuo, W. S.; Cheng, C. C. *Polym. Compos.* **2011**, *32*, 305.
14. Wu, T. L.; Lo, T. S.; Kuo, W. S. *Polym. Compos.* **2010**, *31*, 292.
15. Li, Y.; Shimizu, H. *Polymer* **2007**, *48*, 2203.
16. Zhang, H. Y.; Zeng, G. X.; Ge, Y.; Chen, T. L.; Hu, L. C. *J. Appl. Phys.* **2009**, *105*, 054314.
17. Fan, Z. J.; Luo, G. H.; Zhang, Z. F.; Zhou, L.; Wei, F. *Mater. Sci. Eng. B* **2006**, *132*, 85.
18. Park, S. H.; Thielemann, P.; Asbeck, P.; Bandaru, P. *Appl. Phys. Lett.* **2009**, *94*, 243111.
19. Huang, S.; Zhou, W.; Luo, F.; Zhu, D. *J. Appl. Polym. Sci.* **2013**, *130*, 1392.
20. Theilmann, P.; Yun, D. J.; Asbeck, P.; Park, S. H. *Org. Electron.* **2013**, *14*, 1531.
21. Liu, Z.; Bai, G.; Huang, Y.; Li, F.; Ma, Y.; Guo, T.; He, X.; Lin, X. Gao, H.; Chen, Y. *J. Phys. Chem. C* **2007**, *111*, 13696.
22. Wu, T. L.; Lo, T. S.; Kuo, W. S. *Polym. Compos.* **2010**, *31*, 292.
23. Yang, R. B.; Hsu, S. D.; Lin, C. K. *J. Appl. Phys.* **2009**, *105*, 07A527.
24. Jiang, M. J.; Dang, Z. M.; Bozlar, M.; Miomandre, F.; Bai, J. *J. Appl. Phys.* **2009**, *106*, 084902.