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ORIGINAL RESEARCH PAPER

Effect of carbon black content on the microwave absorbing properties of CB/epoxy composites

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ABSTRACT

To prevent serious electromagnetic interference, a single-layer and double layer wave-absorbing coating employing complex absorbents composed of carbon black with epoxy resin as matrix was prepared. The morphologies of carbon black /epoxy composites were characterized by scanning electron microscope and atomic force microscope, respectively. The carbon black particles exhibit obvious polyaromatic were characterized by X-ray diffraction. The electromagnetic parameters of carbon black were measured in the frequency range of 8–12 GHz by transmission/reflection technology, and the electromagnetic loss mechanisms of the two particles were discussed, respectively. The microwave absorption properties of the coatings were investigated by measuring reflection loss using arch method. The effects of carbon black mass ratio, thickness and double-layer on the microwave absorption properties were discussed, respectively. The results showed that the higher thickness, higher ratio and double-layer of carbon black /epoxy content could make the absorption band shift towards the lower frequency range. Significantly, the wave-absorbing coating could be applied in different frequency ranges according to actual demand by controlling the content of carbon black in composites.

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INTRODUCTION

Recently, electromagnetic wave in GHz range is increasingly used in wireless telecommunication systems and high frequency circuit devices, such as mobile phone, local area network, satellite broadcast systems, and so on. [1-6]. Electromagnetic absorbers (EMA) are currently gaining much attention especially in the field of microwave frequencies applications. These materials, historically introduced as radar absorbing materials (RAMs). Adding pure dielectric or magnetic fillers to a polymer matrix is a possible way to change the material electromagnetic properties and performance [7].

The alteration of the material is based on the following considerations: (i) in the GHz range, electrical properties of activated carbon-fiber felt are similar to that of metal; (ii) activated carbon-fiber felt has the structure of fabrics and the fibers composed of it has the irregular-shaped cross-sections, which are very advantageous for the absorption of electromagnetic wave; (iii) activated carbon-fiber felt is lighter than metal [8-9].Carbon black is also used in electromagnetic interference

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shielding applications, mainly as conductive fillers in composite materials, due to their electrical conductivity, chemical resistance and low density. Percolation takes place at a critical CB loading, called percolation concentration, where the first three-dimensional continuous CB network is built throughout the polymer matrix. The percolation concentration of such mixtures depends on the CB structure (particle size, aggregate shape and structure, porosity and surface chemistry), on the polymer characteristics (chemical structure and crystalloid) and on the processing methods and processing conditions. Generally, higher conductivities of the polymer composites can be obtained by using CBs of smaller particle size (larger surface area), lower particle density (higher particle porosity), higher structure (better aggregation) and low volatility (fewer chemisorbed oxygen groups) [10-17].

The electromagnetic (e.m.) properties and absorption performance were evaluated by performing wave guide measurements in the X-band (8.2–12.4 GHz). This e.m. band, in fact, is increasing its importance in a variety of engineering applications such as polarimetric radar [18], medical accelerators [19], space radars [20], stealth purposes, etc. Carbon nanofibres (CNFs) were selected as lossy media due to their peculiar electromagnetic properties. Compared to common micrometer conductive fillers, in fact, CNFs present higher aspect ratio and electrical conductivity [21-23] that allow to obtain good em losses at low filler concentration, with a considerable reduction of thickness and weight [7]. A two-layered absorber consisting of the carbon fiber composite substrate can be proposed as a light-weighted structural absorber having low microwave reflection.

In this study, a single-layer and double-layer wave absorbing coating employing CB as dielectric loss absorbent with epoxy resin as matrix were prepared. Electromagnetic properties and microwave absorption mechanisms of CB were analyzed, respectively. The effects of CB ratio, thickness and double-layer on the microwave absorption properties were discussed.

MATERIALS AND METHODS

All of the reagent for prepared of CB/resin epoxy such as acetone, LY5052 resin epoxy and CH 5052 hardner were kindly provided by Huntsman.

Characterisations

XRD patterns were recorded by a Philips X-ray diffractometer using Ni-filtered Cu Ká radiation. Scanning electron microscopy (SEM) images were obtained on LEO. Prior to taking images, the samples were coated by a very thin layer of Pt to make the sample surface conducting and prevent charge accumulation, and obtaining a better contrast. An atomic force microscopy (AFM) model NT-MDT Solver P47 was used in the tapping mode for morphological characterization using ultrasharp Si cantilevers. The reflection loss (RL) versus frequency of the wave-absorbing coatings was tested by a HP8720B vector network analyzer in the 8-12 GHz range using the arch method.

Fabrication of CB/epoxy composites

The wave-absorbing coatings were prepared by adding CB into epoxy resin composites including hardener, aceton and resin acceptor. A calculated amount of CB was suspended in certain of acetone on a mechanical stirrer was stirred at 500 rpm for 20 minutes. Then, with respect to CB, a certain amount of resin added to the solution and was stirred for 2 hours on a mechanical stirring at 500 rpm was continued. Continue stirring for 1 hour at a speed of over 1100 rpm put up more mixed is full uniform. Then, to make a stiff mixture and remove acetone in solution agitation was continued. Then, the resulting mixture stayed in the darkness for 24 hours then in the vacuum oven at 50 °C with a pressure of 300 mbar was set to remove bubbles for 45 minutes until leave acetone completely after that, consider the ratio of hardener was added and stirred for 20 minutes. The uniform mixtures were coated on a rectangular steel substrate (80mm×50mm). Meanwhile, the thicknesses of coatings were controlled by perspex dies. After the hardening of epoxy resin, the as-prepared coatings were ready for microwave properties test. The mixture proportions and the thicknesses of samples were shown in Table 1.

RESULTS AND DISCUSSION

Morphologies of CB/epoxy composites

Fig. 1 shows the SEM images of A2, A3, A4 and A5 samples. Fig. 1 (a) shows the SEM image surface of sample A2 indicates that the unevenness is less than smooth surface. According to Fig. 1 (b) SEM image surface of sample A3 indicates that the surface is

Table 1. The mix proportions and thicknesses of samples.

The samples	Layer no.	CB (wt%)	Thickness (mm)
A1	1	2	2
A2	1	5	2
A3	1	7	2
A4	1	10	2
A5	1	7	3
A6	1	10	3
A7	1	5	1
	2	10	2
A8	1	7	1
	2	10	2

almost flat but uneven. Fig. 1 (c) and (d) show the SEM images surface of samples A5 and A4 indicate that the surface smoother than their predecessor and notice the good uniformity of the composite indicating the good degree of dispersion of the nanofiller.

Fig. 2 (a,b) show the SEM images surface of samples A7 and A8 indicate that the surface smoother and notice the good uniformity of the composite.

Figs. 3 (a,b) are AFM images of A2, Figs. 3 (c) and (d) are AFM images of A4 and Figs. 4 (a,b) are AFM images of A7 and A8, which roughness in the surface of composite approve presence of CB in epoxy matrix.

X-ray diffraction of CB/epoxy composites

Fig. 5 shows the X-ray diffraction patterns of the CB/epoxy composites. The CB particles exhibit obvious polyaromatic, turbostatic structural features with (002) peak, despite the fact that some amorphous structures may coexist due to the large half-peak width and low intensity of the related (002) reflection [24].

Microwave absorption properties of coatings

Theory

According to the transmission line theory, the RL (dB) of electromagnetic radiation under normal incident wave at the surface of single-layer material backed by a perfect conductor can be defined by [25]:

$$RL(dB) = 20\log\left(\frac{Z_{in} - 1}{Z_{in} + 1}\right) \quad (1)$$

while the normalized input impedance (Z_{in}) was calculated by:

$$Z_{in} = \sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left[j\left(\frac{2\pi f d}{c}\right)\sqrt{\mu_r.\varepsilon_r}\right] \quad (2)$$

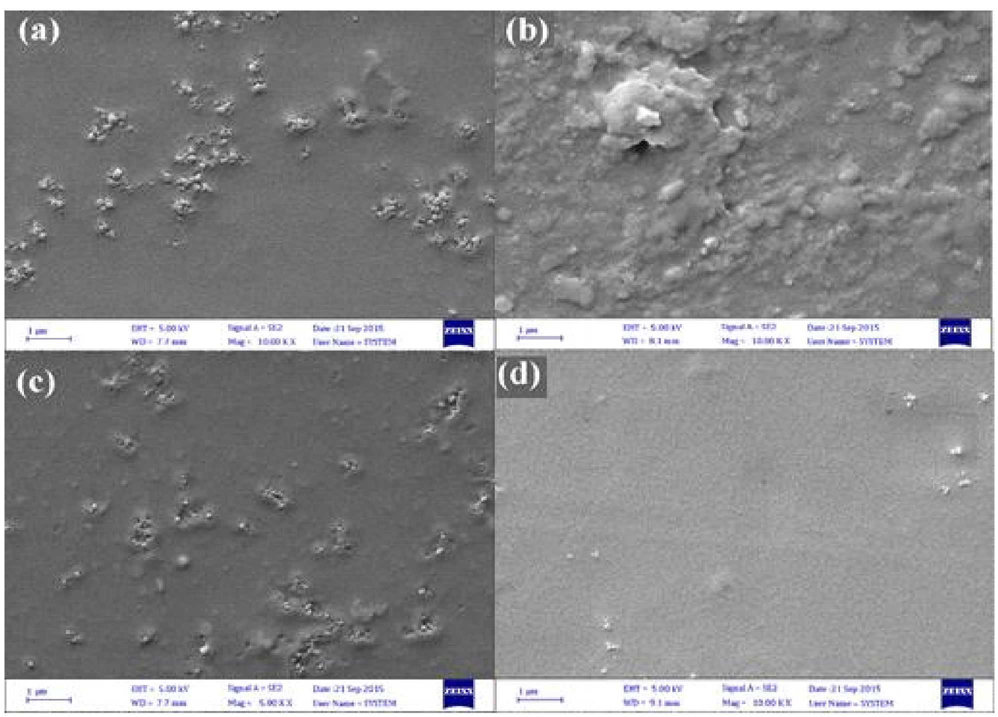

Fig. 1. SEM images of A2, A3, A4 and A5 samples.

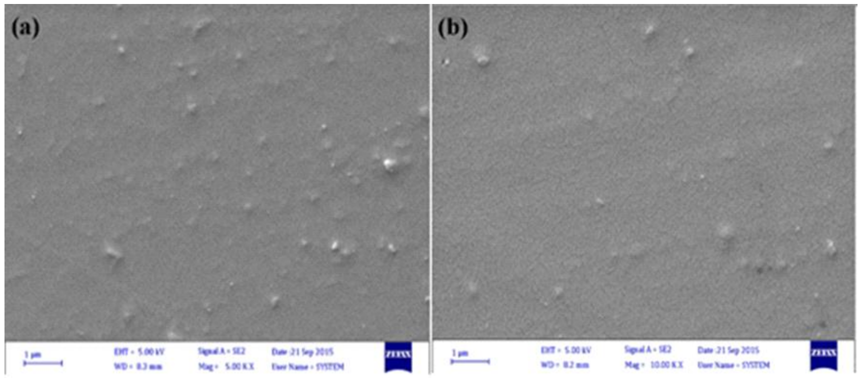

Fig. 2. SEM images surface of (a) A7 and (b) A8 samples.

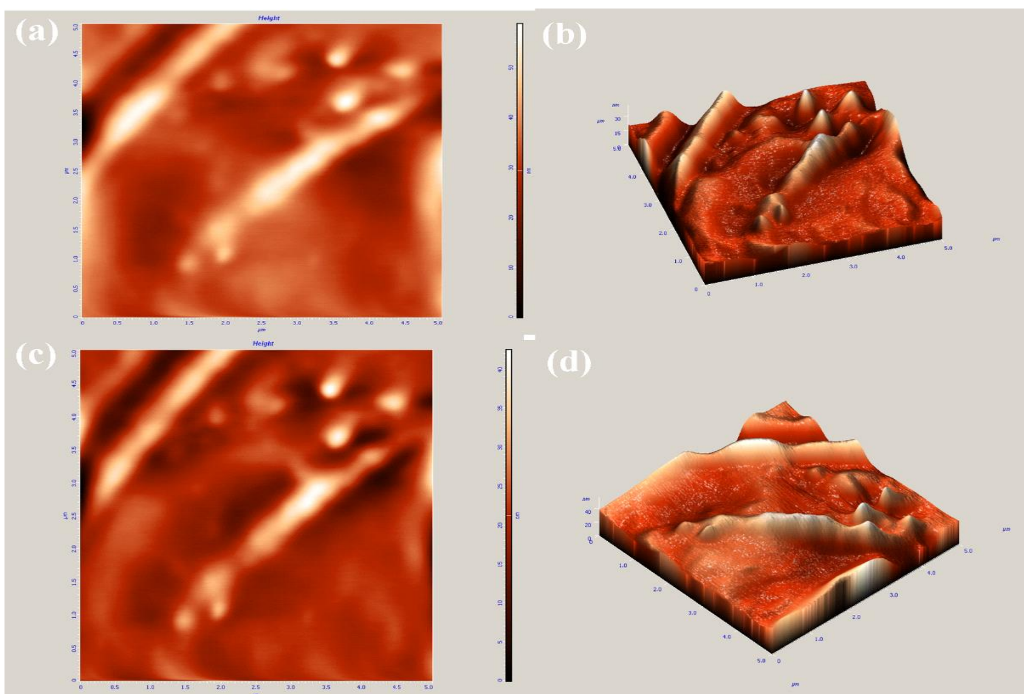

Fig. 3. AFM images of: (a,b) A2 and (c,d) A4 samples.

where f is the microwave frequency, d is the thickness of the absorb layer, c is the velocity of electromagnetic wave in vacuum, and ε_r and μ_r are the complex relative permittivity and permeability, respectively [26].

Effect of CB ratio on the microwave absorption properties

Fig. 6 shows the variations of RL versus frequency in the range of 8-12 GHz for the samples with the thickness of 2mm (A1, A2, A3 and A4) the coatings are composed of different CB mass ratios (2 wt.%, 5 wt.%, 7 wt.% and 10 wt.%). The microwave absorption properties of the four samples are shown in Table 2. From the data in Table 2 and curves in Fig. 6, as increasing the CB ratio, the absorption bands of the samples shift towards the lower frequency range, the bandwidth of A4 sample achieves -29.6 GHz (8.2-8.6 GHz).

The results may be from the following factors: firstly, the dielectric polarization is increased because of higher CB content. Secondly, the more CB particles in matrix per unit volume, the more surface areas are provided to attenuate electromagnetic wave by multi-scatter and reflection. Finally, the increasing CB content is helpful to form conductive net, the electromagnetic wave is attenuated by eddy current loss.

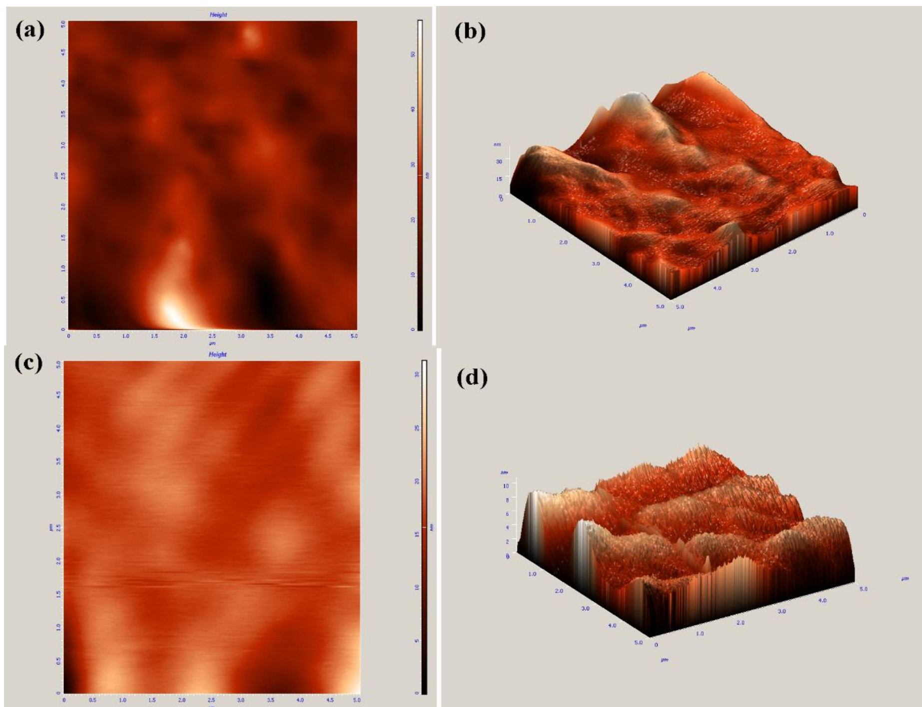

Fig. 4. AFM images of: (a,b) A7 and (c,d) A8 samples.

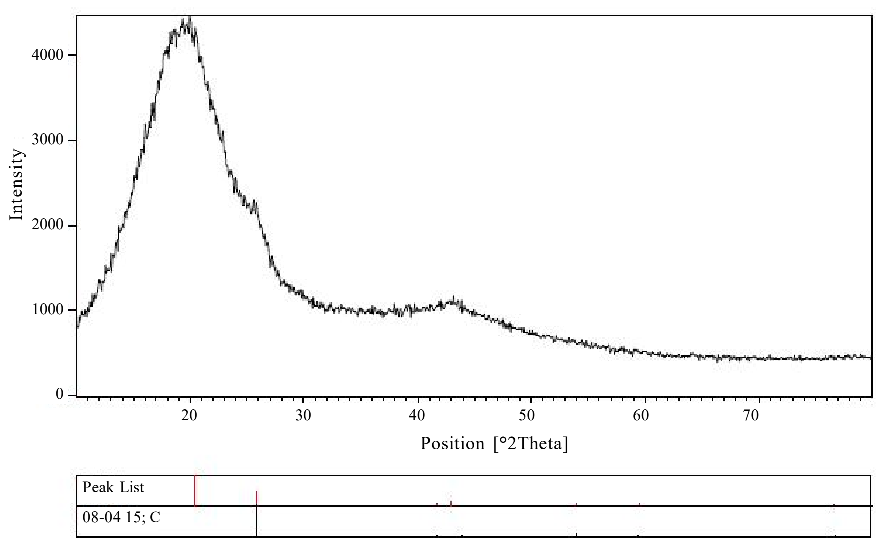

Fig. 5. XRD patterns of the CB/epoxy composites.

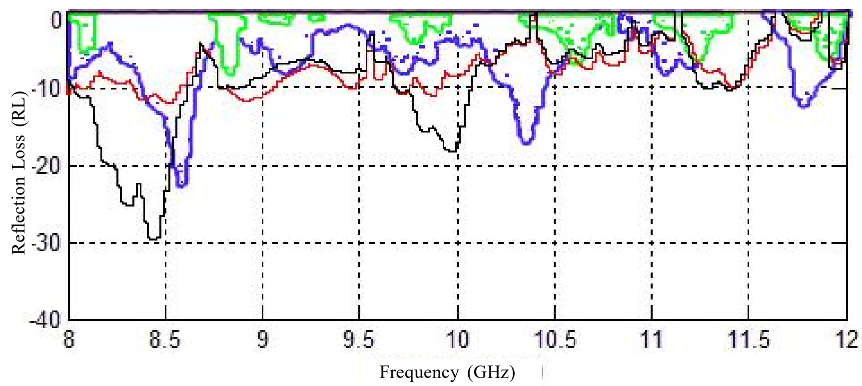

Fig. 6. The variations of RL versus frequency in the range of 8-12 GHz for A1 (green), A2

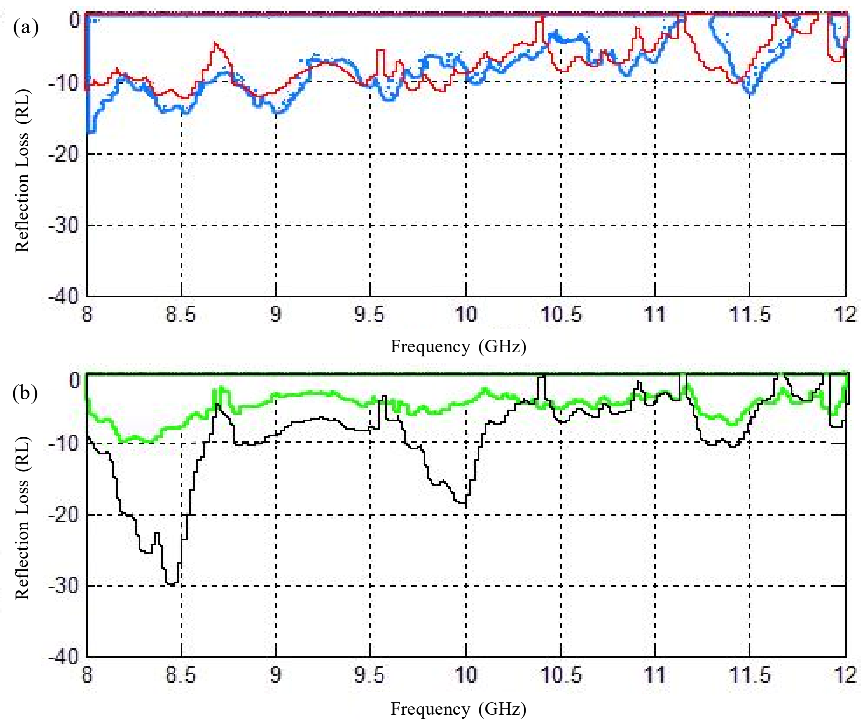

Fig. 7. The RL curves of: (a) A3 (red) and A5 (blue) and (b) A4 (black) and A6 (green) samples.

Table 2. Microwave absorption properties of the samples with different CB mass ratios.

The samples	Frequency of absorption peak (GHz)	Minimum RL value (dB)
A1	10.5	-7.8
A2	8.5, 10.1	-23.2
A3	8.5	-11.8
A4	8.2-8.6	-29.6

Table 3. Microwave absorption properties of the samples with different thicknesses.

The samples	Frequency of absorption peak (GHz)	Minimum RL value (dB)
A3	8.5	-11.8
A5	8.5, 9.5	-20.3
A4	9.1	-29.6
A6	8.5	-9.5

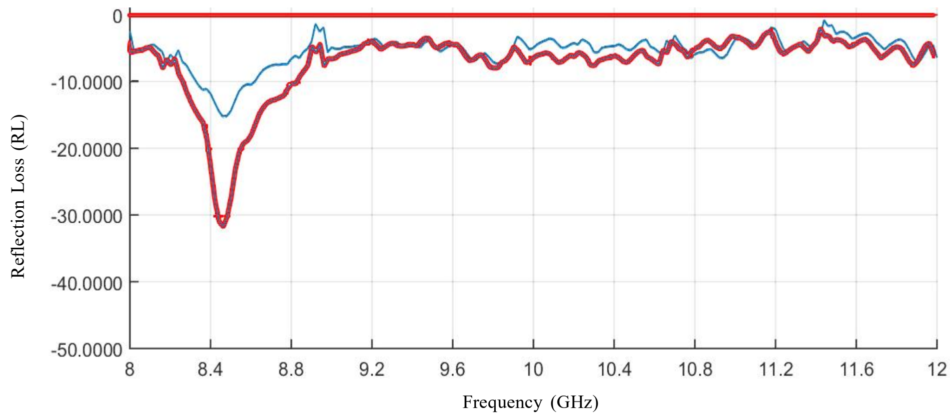

Fig. 8. The RL curve of A7 (blue) and A8 (red) samples.

Effect of thickness on the microwave absorption properties

Fig. 7 (a) and (b) shows the RL curves of the samples with the same CB ratio 7 wt.% (A3 and A5) and 10 wt.% (A4 and A6), different thicknesses (2 mm and 3 mm). Some key data for the microwave absorption properties of the samples are shown in Table 3. According to Fig. 7 and Table 3, obviously, the absorption peaks of samples shift towards the lower frequency range with increasing thickness of coating.

From the above results, it is found that increasing thickness of absorber could improve the microwave absorption properties of coating in the lower frequency range. The phenomenon can be explained by the equation [27].

$$f_m = \frac{c}{2\pi\mu'' d} \quad (3)$$

where f_m, c, and d are the matching frequency (the frequency of absorption peak), the velocity of light, and the sample thickness, respectively. This equation indicates that the matching frequency f_m shifts towards lower frequency with increasing sample thickness. So the microwave absorption properties of the samples can be affected by thickness.

Effect of two absorb layers on the microwave absorption properties

The RL of two absorb layers were calculated as follows:

$$R = \left[\frac{Z_{ino} - \eta_0}{Z_{ino} + \eta_0}\right] \quad (4)$$

Table 4. Microwave absorption properties of the samples with two absorb layers.

The samples	Frequency of absorption peak (GHz)	Minimum RL value (dB)
A7	8.5	-15.4
A8	8.2-8.8	-32.1

where η_0 is the free-space impedance and Z_{in0} is the input impedance of the panel for $z = 0$. Expressing Z_{in0} as function of the coefficient of the bi-layer transmission matrix, beside, The input-to-output field transfer matrix of the two-layer panel is computed as the cascade of the transmission matrices of the lossy sheet, $[\phi_L]$, and of the spacer, $[\phi_S]$:

$$[\phi] = [\phi_s][\phi_L] \quad (5)$$

where the coefficients of the (2×2) matrices $[\phi_S]$ and $[\phi_L]$ assumes the well know expressions reported in [28-29]. Finally, it yields:

$$R = \left[\frac{\phi_{L12}\,(\emptyset_{S11} + \eta_0\,\phi_{S21}) + \phi_{L11}\,(\emptyset_{S12} + \eta_0\,\phi_{S11})}{\phi_{L12}\,(\emptyset_{S11} - \eta_0\,\phi_{S21}) + \phi_{L11}\,(\emptyset_{S12} - \eta_0\,\phi_{S11})}\right] \quad (6)$$

Fig. 8 shows the RL curve of the samples with the same thickness (3mm) and CB ratio 5 wt.% + 10 wt.% (A7) and 7 wt.% + 10 wt.% (A8). Some key data for the microwave absorption properties of the samples are shown in Table 4. According to Fig. 8 and Table 4, obviously, the absorption peaks of samples shift towards the lower frequency range with increasing ratio of coating. The bandwidth of A8 sample achieves -32.1 GHz (8.2-8.8 GHz).

CONCLUSION

In this paper, the microwave absorption properties of single-layer and double layer coatings composed of CB are studied in the 8–12 GHz range. The frequency of absorption bands shift towards the lower frequency range with increasing CB ratio or thickness and two layers. So the wave-absorbing coating could be applied in desired frequency range through adjusting the content of CB in composites. Besides, the absorption bandwidth (RL <"4 dB) can be enhanced by increasing CB content for the samples with the thickness of 3mm, especially, for the sample (double-layer) with CB ratio (7 wt.% + 10 wt.%), the absorption bandwidth reaches -32.1GHz (8.2-8.8 GHz).

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CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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