MWCNTs and nanoparticles composite as a high efficient and lightweight electromagnetic

wave absorber in 4-18 GHz

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SI-1: The selection of the absorber loading for MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄

With an absorber content of 5%, illustrated in Figure S1, the strong RL of -16.1 dB was observed at 17 GHz with an absorber thickness of 5.5 mm. Thus, at this absorber loading, the EM wave attenuation ability is limited.



Figure S1. The relationship between RL-F for MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄ with the absorber content of 5%.

However, when the absorber loading is about 10% (Figure S2), the EM wave absorption was strongly enhanced. The minimum RL value of -45.6 dB (more than 99.99% absorption) can be gained at 14.4 GHz with a matching thickness of 5.5 mm. Also with a thickness of 5.0 mm, a -35.8 dB loss can be obtained. At this loading, the nanocomposite absorber has an excellent EM wave attenuation performance with strong EM wave absorbency especially in K_u band (12-18GHz). However, it still cannot meet the strict requirement for an excellent absorber.



Figure S2. The relationship between RL-F for MWCNTs/Ag/ $Co_{0.2}Fe_{2.8}O_4$ with the absorber content of 10%.

At a 15% absorber content as shown in Figure S3, RL value below -10 dB can be obtained in the EM wave C band (4-8 GHz), X band (8-12 GHz) and K_u band (12-18 GHz), which means 90% of the incident EM wave energy is absorbed. Hence, at this loading, the synthesized nanocomposite can serve as a broad band EM wave absorber. Though there are two absorption peaks for absorber thickness of 4.0 mm, 4.5 mm, 5.0 mm and 5.5 mm, respectively, the minimum RL values are all around -15 dB at different absorber thicknesses. Thus, at this content, the nanocomposite cannot satisfactorily meet the strict requirement for an excellent absorber.



Figure S3. The relationship between RL-F for MWCNTs/Ag/ $Co_{0.2}Fe_{2.8}O_4$ with the absorber content of 15%.

When the absorber content increased to 20% (Figure S4), the EM wave attenuation performance was greatly improved. At EM wave frequency of about 12.8 GHz and 9.7 GHz, RL of -34.8 dB (more than 99.9% attenuation) and -42.1 dB (more than 99.99% attenuation) were achieved with relatively thin absorber thickness of 1.5 and 2.0 mm, respectively. With absorber thickness 1.5-5.5 mm, RL value below -10 dB can be achieved from 3.0 to 14.4 GHz, which covers the C band, the X band and the K_u band. Undoubtedly, with absorber content of 20%, the nanocomposite absorber has the potential to meet the strict requirements (wide absorption band width, thin matching thickness, strong absorbency) for EM wave absorber. According to the above EM wave RL data, the synthesized MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄ nanocomposite absorber exhibits a tunable EM wave absorption characteristic by adjusting the absorber loading to achieve wide absorption band width, strong EM wave absorption ability, and relatively thin matching thickness.



Figure S4. The relationship between RL-F for MWCNTs/Ag/ $Co_{0.2}Fe_{2.8}O_4$ with the absorber content of 20%.

SI-2: The Microwave absorption properties for MWCNTs

In the nanocomposite absorber MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄, the content of MWCNTs is about 61.8% as detected by EDS shown in Figure 3. Thus, for comparison, a sample containing MWCNTs and paraffin wax with 12.4% loading ($20\% \times 61.8\% = 12.4\%$) was used to determine the EM parameters, and the obtained EM parameters were used to simulate the EM wave reflection loss of MWCNTs by the transmission line theory as shown in Figure S5.



Figure S5. The relationship between RL-F for MWCNTs with the absorber content of 12.4%.

SI-3: The Microwave absorption properties for MWCNTs/Ag

In the nanocomposite absorber MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄, the total content of MWCNTs and Ag nanoparticles is about 66% as detected by EDS shown in Figure 3. Thus, for comparison, a sample containing MWCNTs/Ag and paraffin wax with 13.2% loading ($20\% \times 66\% = 13.2\%$) was used to determine the EM parameters, and the obtained EM parameters were used to simulate the EM wave reflection loss of MWCNTs/Ag by the transmission line theory as shown in Figure S6.



Figure S6. The relationship between RL-F for MWCNTs/Ag with the absorber content of 13.2%.

SI-4: The dielectric loss tangent of MWCNTs, NWCNTs/Ag and MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄

The nanocomposite absorber (MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄) has a better EM wave attenuation ability for the decoration of Ag nanoparticles and Co_{0.2}Fe_{2.8}O₄ nanoparticles. The dielectric loss ability (dielectric loss tangent $\tan \delta_{\epsilon} = \epsilon''/\epsilon'$) of MWCNTs/Ag is higher than that of MWCNTs for the introduction of Ag nanoparticles. When Co_{0.2}Fe_{2.8}O₄ nanoparticles were used to decorate MWCNTs, the dielectric loss ability of the nanocomposite absorber (MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄) was strongly enhanced as sketched in Figured S7.



Figure S7. The dielectric loss tangent of MWCNTs, NWCNTs/Ag and MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄.

SI-5: The Cole-Cole semicircles of MWCNTS, MWCNTs/Ag, and MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄

According to the electromagnetic theory, it is known that the dielectric loss of MWCNTs may be attributed to natural resonance, electron polarization relaxation and Debye dipolar relaxation and so on. As for the Debye dipolar relaxation, the relative complex permittivity (ε_r) can be expressed by the following equation (1):

$$\varepsilon_{r} = \varepsilon_{\infty} + \frac{\varepsilon_{s} - \varepsilon_{\infty}}{1 + j2\pi f\tau} = \varepsilon' - j\varepsilon''$$
⁽¹⁾

where f, ε_s , ε_{∞} and τ are frequency, static permittivity, relative dielectric permittivity at the high-frequency limit, and polarization relaxation time, respectively. Consequently, ε' and ε'' can be described by the following equation (2) and (3):

$$\varepsilon' = \varepsilon_{\infty} + \frac{\varepsilon_{s} - \varepsilon_{\infty}}{1 + (2\pi f)^{2} \tau^{2}}$$
⁽²⁾

$$\varepsilon'' = \frac{2\pi f \tau (\varepsilon_s - \varepsilon_\infty)}{1 + (2\pi f)^2 \tau^2}$$
(3)

According to the above equation (2) and (3), the relationship between ε' and ε'' can be deduced as:

$$\left(\varepsilon^{'} - \frac{\varepsilon_{s}^{} + \varepsilon_{\infty}}{2}\right)^{2} + \left(\varepsilon^{''}\right)^{2} = \left(\frac{\varepsilon_{s}^{} - \varepsilon_{\infty}}{2}\right)^{2} \tag{4}$$

Thus, the plot of $\varepsilon' vs. \varepsilon''$ would be a single semicircle, generally denoted as the Cole-Cole semicircle. Each semicircle corresponds to one Debye relaxation process. The Cole-Cole semicircles of MWCNTs, MWCNTs/Ag, and MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄ are shown in Figure S8.



Figure S8. The relationship between the real part and the imaginary part of the relative permittivity for (a) MWCNTs; (b) MWCNTs/Ag; (c) MWCNTs/Ag/Co_{0.2}Fe_{2.8}O₄.