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## Enhanced microwave absorption performance of $\text{Fe}_3\text{Al}$ flakes by optimizing the carbon nanotube coatings†

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$\text{Fe}_3\text{Al}$  is a good magnetic loss absorber for microwave absorption. However, due to the relatively high density and poor impedance matching ratio, the potential of  $\text{Fe}_3\text{Al}$  cannot be fully released. Herein, a dielectric loss absorber of carbon nanotubes (CNTs) is coupled with  $\text{Fe}_3\text{Al}$  to form  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers. CNTs are randomly tangled and coated on the surface of the  $\text{Fe}_3\text{Al}$  flakes, forming a connecting conductive network. By carefully tuning the content of CNTs, the optimized  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorber with 1.5% of CNTs can combine both magnetic loss and dielectric loss mechanisms, thus achieving an impedance matching ratio close to 1 while keeping strong attenuation for enhanced microwave absorption. As a result, an effective absorption bandwidth ( $RL \leq -10$  dB) of 4.73 GHz at a thickness of 2 mm is achieved.

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### 1. Introduction

The development of efficient electromagnetic (EM) wave absorption materials is of significance in both military and civil applications.<sup>1–4</sup> Magnetic loss absorbers such as metals, alloys, and spinel ferrites have shown great potential as efficient EM wave absorption materials.<sup>5–10</sup> In particular,  $\text{Fe}_3\text{Al}$ -based intermetallic compounds have attracted increasing attention as good magnetic loss absorbers due to their high permeability, low electrical conductivity, excellent oxidation resistance, and good corrosion resistance.<sup>11–15</sup> Tuning the morphology and order degree of  $\text{Fe}_3\text{Al}$  can significantly affect its EM wave absorption performance. For example, annealing  $\text{Fe}_3\text{Al}$  flakes at 500 °C generated the most  $\text{DO}_3$  superlattice structure, exhibiting an effective absorption bandwidth ( $RL \leq -10$  dB) of 7.2 GHz with a thickness of 1.35 mm.<sup>16</sup> Nevertheless, some issues such as relatively high density and unsatisfied EM wave absorption properties should be properly addressed for possible scale-up applications. To achieve excellent EM wave absorption performance, effective impedance matching and strong attenuation are important. Generally, magnetic loss absorbers exhibit good attenuation but poor impedance matching.<sup>17–19</sup> A proper combination of magnetic loss absorbers and dielectric loss

absorbers is an efficient approach to increase the EM wave absorption performance.

However, light weight is also important for practical applications specially in the aeronautic and astronautic fields.<sup>20–23</sup> In this regard, carbon nanotubes (CNTs) are one of the good candidates as dielectric loss absorbers to be coupled with magnetic loss absorbers due to the low density, high aspect ratio, and interesting electrical properties.<sup>24</sup> For example, a composite absorber composed of mesoporous carbon hollow sphere (MCHS)@Co@carbon nanotubes (CNTs) was fabricated, and the CNTs functioned as electron transportation channels to reinforce conductive loss, leading to a strong absorption intensity of  $-37.3$  dB and a wide effective absorption bandwidth of 8.0 GHz.<sup>25</sup> Coiled carbon nanotubes (CCNTs) were coated using  $\text{CoFe}_2\text{O}_4$  (CFO) nanoparticles to form CCNTs/CFO composites, which exhibited a minimum reflection loss of  $-14$  dB in the frequency range of 2–18 GHz, and an effective absorption bandwidth of 4.0 GHz.<sup>26</sup> In another example, nano- $\text{Fe}_3\text{O}_4$  compact-coated CNTs (FCCs) and  $\text{Fe}_3\text{O}_4$  loose-coated CNTs (FLCs) were fabricated by a solvothermal method, and the FCCs with the optimized CNT to  $\text{Fe}^{3+}$  ratio exhibited a minimum reflection loss of  $-28.7$  dB with a thickness of 1 mm and an effective absorption bandwidth of 8.3 GHz.<sup>27</sup> CoNi nanoparticles encapsulated within the nitrogen-doped CNT (NCNT) arrays were grown on ultrathin rGO sheets (3D CoNi/NGCT), which showed excellent microwave absorption properties.<sup>28</sup> N-doped carbon hollow spheres supported on N-doped carbon nanotubes (Ni@NCNT/NCHSs) with a thickness of 1.6 mm, showed an  $RL_{\min}$  of  $-64.75$  dB and an effective absorption bandwidth of 4.17 GHz.<sup>29</sup> Similarly, N-doped carbon fibers (NCF) with N-doped carbon nanotubes (NCNTs) vertically grown

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on the surface of NCF exhibited an absorption bandwidth of 4.0 GHz and a minimum reflection loss of  $-49.56$  dB at a thickness of 1.5 mm.<sup>30</sup> Hierarchical nitrogen-doped carbon hollow microspheres assembled with loose and porous magnetic carbon sheets were prepared by a facile two-step method, exhibiting an effective absorption bandwidth of  $\sim 4.00$  GHz at a thickness of 2.5 mm and an  $RL_{min}$  of  $-39.87$  dB.<sup>31</sup> Inspired by these previous studies, the incorporation of CNTs may be an effective strategy to address the high density issue of  $\text{Fe}_3\text{Al}$  while enhancing the EM wave absorption performance.

Herein, we report a facile mechanical blending method for the fabrication of  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers with different CNT contents. The CNTs can form a connecting conductive network coated on the surface of  $\text{Fe}_3\text{Al}$  flakes. With the optimized mass content of 1.5% of CNTs, the  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  composite absorber can take advantage of both dielectric-loss and magnetic-loss mechanisms to promote microwave absorption, demonstrating an effective absorption bandwidth ( $RL \leq -10$  dB) of 4.73 GHz at a thickness of 2 mm. The excellent EM wave absorption performance is attributed to the proper impedance matching ratio and strong attenuation. The low absorber thickness, high reflection loss and relatively broad absorption bandwidth of the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorber demonstrate great potential for practical applications.

## 2. Experimental section

### 2.1 Synthesis of $\text{Fe}_3\text{Al}/\text{CNT}$ composite absorbers

$\text{Fe}_3\text{Al}$  flakes were prepared according to our previous study.<sup>16</sup> A mechanical alloying method was used to prepare a  $\text{Fe}(\text{Al})$  solid solution. More specifically, Fe and Al metal powders with an atomic ratio of 7 : 3 were mixed in a 500 mL zirconium oxide ball milling tank, which was then located in a QM-3SP2 high-energy ball milling machine (Nanjing University). Zirconium oxide grinding balls with sizes of 1–10 mm were used, and the ball-to-powder weight ratio was 15 : 1. The ball milling time was 50 h, and the rotation speed was 400 rpm. To avoid the oxidation of the samples during the ball milling process, the ball milling tank was vacuumed and filled with Ar as the protective gas. When the ball milling process was finished and cooled to room temperature, the ball milling tank was transferred to a glove box filled with Ar. Then, the ball milling tank was opened in the glove box and the obtained powder was taken out to avoid any possible oxidation.

A vacuum tube furnace (OTF-1200X, Hefei Kejing) was used to anneal the above-obtained powder. Before the annealing process, the tube furnace was firstly vacuumed to  $10^{-3}$  Pa and then filled with Ar as the protective gas. The annealing temperature was 500 °C, and maintained for 2 h. After naturally cooling to room temperature, the ordered  $\text{Fe}_3\text{Al}$  powder was obtained.

Then, a mechanical blending method was used to prepare the  $\text{Fe}_3\text{Al}/\text{carbon nanotubes}$  (CNTs) composite powders, and the contents of CNTs were controlled as 0%, 0.5%, 1% and 1.5% (mass ratio) of  $\text{Fe}_3\text{Al}$ . According to the amount of CNTs added, the obtained four  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite powders were denoted as  $\text{Fe}_3\text{Al}$ ,  $\text{Fe}_3\text{Al}-0.5\%\text{CNT}$ ,  $\text{Fe}_3\text{Al}-1\%\text{CNT}$ , and  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$ .

To ensure the homogeneous dispersion, sodium dodecylbenzene sulfonate (SDBS) was selected as the dispersant and dissolved in a water bath at 80 °C.  $\text{Fe}_3\text{Al}$  and CNT suspensions were prepared with the ratios of SDBS :  $\text{Fe}_3\text{Al} = 1 : 10$  and SDBS : CNTs = 10 : 1. The above suspensions were mixed under ultrasonication and mechanical stirring together for 2 h. The obtained powders were centrifuged and dried in a vacuum oven, and  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite powders were obtained.

All the synthesis samples were repeated three times and scanning electron microscopy was conducted to confirm the morphology of the samples.

### 2.2 Characterization

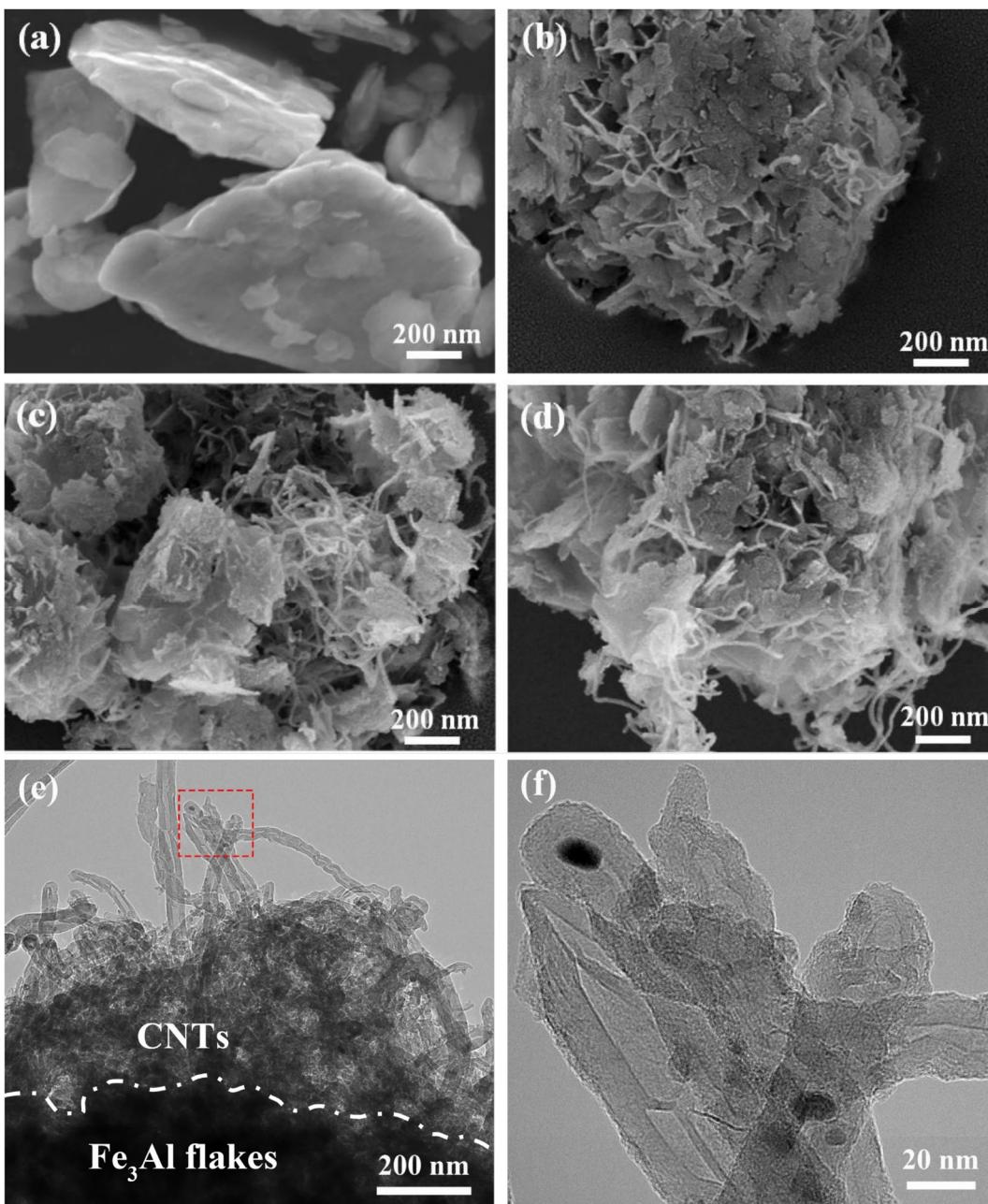
The morphology of the powders was characterized by scanning electron microscopy (TESCAN MIRA LMS). The crystal structure and phase composition of the powders were analyzed by a Bruker D8-Advance X-ray electron diffractometer (Cu  $K\alpha$  radiation with an accelerating voltage of 40 kV and a current of 35 mA). The CNT signals in the composite powders were characterized by a Thermo Scientific Nicolet iS5 infrared spectrometer. For measurement of the microwave absorption properties, the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite powder was mixed in paraffin wax at the mass ratio of 4 : 1, which was then pressed into a ring specimen with an inner diameter of  $\varphi$  3.04 mm and an outer diameter of  $\varphi$  7.00 mm.<sup>32</sup> For comparison, other samples were prepared with the  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  composite/paraffin wax mass ratios of 3 : 2, 2 : 3, and 1 : 4, respectively. The electromagnetic parameters in the range of 2–18 GHz were determined using a vector network analyzer (N5225B) and a coaxial line test method. To collect more reliable results, three samples were prepared for each  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite with the same CNT content. Data would be selected only if all 3 samples showed similar results.

## 3. Results and discussion

### 3.1 Morphology analysis

The morphologies of  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers at different CNT additions are shown in Fig. 1. It can be clearly seen that  $\text{Fe}_3\text{Al}$  powder obtained after high-energy ball milling and ordering treatment is a lamellar-like structure with a thickness of about 100 nm, and the size of each flake is around 600 nm (Fig. 1a). CNTs have a tubular structure with smooth walls, which are randomly distributed on the surfaces of the  $\text{Fe}_3\text{Al}$  flakes. As shown in Fig. 1b, when the addition amount of CNTs is 0.5%, CNTs are coated on the surface of the  $\text{Fe}_3\text{Al}$  flakes. In addition, CNTs are tangled together to form a relatively loose connecting conductive network. When the CNT content is 1%, more CNTs can be observed, the intermolecular van der Waals forces cause the aggregation of the CNTs, as shown in Fig. 1c. When the CNT content is increased to 1.5%, a thicker CNT coating can be observed, and the  $\text{Fe}_3\text{Al}$  flake is fully covered by CNTs (Fig. 1d). To understand the interfacial interaction between  $\text{Fe}_3\text{Al}$  flakes and CNTs, the  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  sample was characterized by transmission electron microscopy (TEM). As shown in Fig. 1e, CNTs are tangled together and well-





**Fig. 1** Microstructure of  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers obtained at different levels of CNT additions. (a)  $\text{Fe}_3\text{Al}$ , (b)  $\text{Fe}_3\text{Al}-0.5\%\text{CNT}$ , (c)  $\text{Fe}_3\text{Al}-1\%\text{CNT}$ , and (d)  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$ . (e) TEM image of  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$ . (f) Enlargement of (e) in the selective area (red square).

coated on the  $\text{Fe}_3\text{Al}$  flakes. The tube structures of the surface CNTs can be clearly observed in Fig. 1f.

### 3.2 Phase analysis

Crystal structures of the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers were characterized by X-ray diffraction (XRD). As can be seen from Fig. 2, the XRD patterns of the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers with four different additions of CNTs were very similar. The characteristic diffraction peaks appeared at  $2\theta$  of  $47.16^\circ$ ,  $67.53^\circ$  and  $84.81^\circ$ , which can be assigned to the  $\text{Fe}_3\text{Al}$  phase.<sup>33</sup> It should be mentioned that the CNT signal cannot be observed in the XRD patterns, possibly due to the very small content that is

out of the detection limit of the instrument. However, the one-dimensional morphology of CNTs can be clearly observed in the SEM images (Fig. 1b-d), indicating the successful loading of CNTs in the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers.

### 3.3 Fourier transform infrared spectroscopy analysis

Since CNTs cannot be accurately determined by XRD, Fourier transform infrared (FTIR) spectra of the samples were collected for further characterization of CNTs. As shown in Fig. 3, the FTIR spectra of the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers show significant changes in the peaks at the wavenumber ranges of  $1500-1700\text{ cm}^{-1}$ ,  $2800-3000\text{ cm}^{-1}$ , and  $3400-3500\text{ cm}^{-1}$ ,



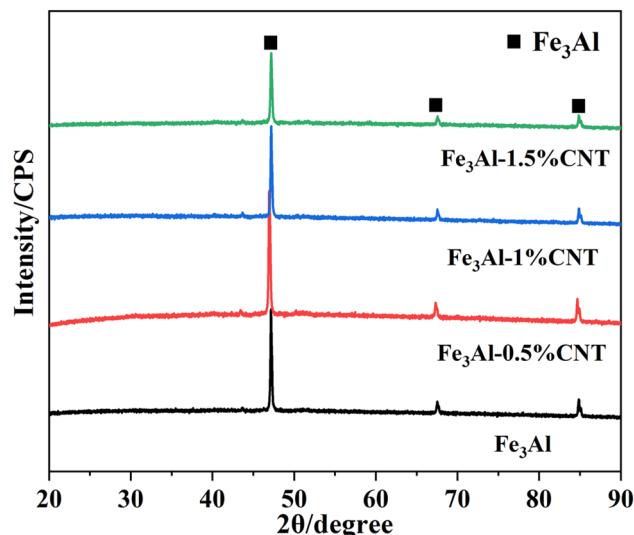


Fig. 2 XRD patterns of the Fe<sub>3</sub>Al/CNTs composite absorbers obtained with different CNT additions.

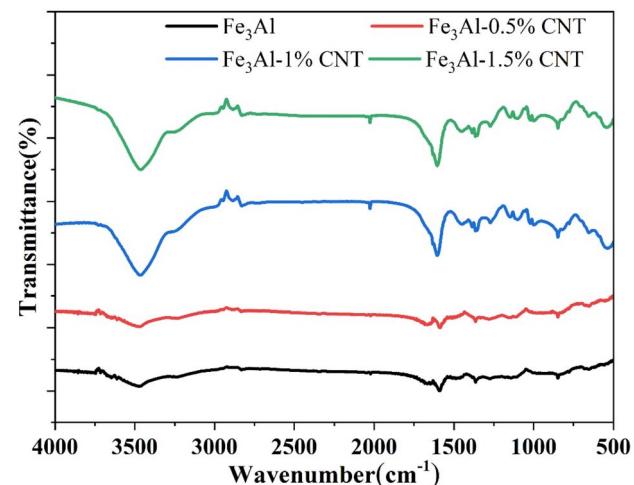


Fig. 3 FTIR spectra of the Fe<sub>3</sub>Al/CNTs composite absorbers.

compared to the pristine Fe<sub>3</sub>Al powder. With the increased content of CNTs, these signals become stronger. Compared to the standard FTIR spectrum of CNTs,<sup>34–36</sup> it was found that the positions of the enhanced peaks are those of carbon nanotubes, which proves the presence of carbon nanotubes in the Fe<sub>3</sub>Al/CNTs composite absorbers.

### 3.4 The complex permittivity and permeability spectra

Fig. 4 shows the curves of four electromagnetic parameters of the Fe<sub>3</sub>Al/CNTs composite absorbers obtained at different levels of CNT additions with respect to frequency. As shown in Fig. 4a, the real part of the dielectric constant ( $\epsilon'$ ) of Fe<sub>3</sub>Al starts at 2 GHz with a value of 9.4, then slightly increases to 9.8 at 8.8 GHz with some fluctuations, and decreases to 9 at 10.8 GHz, then slightly increases to 9.9 at around 17.5 GHz with some fluctuations. While for the Fe<sub>3</sub>Al/CNTs composite absorbers, the real part of

the dielectric constant ( $\epsilon'$ ) shows a decreasing trend with the increase of frequency. In particular, the  $\epsilon'$  values of the Fe<sub>3</sub>Al-0.5%CNT and Fe<sub>3</sub>Al-1%CNT samples significantly decrease in the frequency range of 2–18 GHz compared to that in their Fe<sub>3</sub>Al counterpart. The addition of highly conductive CNTs will increase the conductivity of the absorber, and thus reduce the dielectric properties of the absorber, leading to a significant decrease of the  $\epsilon'$  values.<sup>23,24</sup> However, when the CNT content is further increased to 1.5%, the CNTs will form a conductive network in the Fe<sub>3</sub>Al-1.5%CNT sample, thus inducing strong interfacial polarization, leading to the increase of the  $\epsilon'$  values.

As revealed in Fig. 4b, the imaginary part ( $\epsilon''$ ) of the dielectric constant of the Fe<sub>3</sub>Al/CNTs composite absorber increases with the increase of CNTs from 2 to 18 GHz, compared with that of the pristine Fe<sub>3</sub>Al. Since the imaginary part reflects the ability to lose electromagnetic waves,<sup>37</sup> the addition of CNTs improves the overall dielectric loss capability of the composite absorbers. The imaginary part of the dielectric constant of the Fe<sub>3</sub>Al-1.5%CNT composite absorber is the largest, and its value is in the range of 2.4–3.8. The significant increase of the  $\epsilon''$  values in Fe<sub>3</sub>Al-1.5% CNT is attributed to the strong interfacial polarization induced by the conductive network formed by the surface CNTs. Therefore, the addition of 1.5% of CNTs can exhibit the largest dielectric loss capability for microwave absorption.

Fig. 4c and d are the magnetic permeability real part  $\mu'$  and magnetic permeability imaginary part  $\mu''$  versus frequency curves of the Fe<sub>3</sub>Al/CNTs composite absorbers. It can be observed that the real part of the complex permeability ( $\mu'$ ) of Fe<sub>3</sub>Al is around 2.02 at 2 GHz, and then gradually decreases to 1.04 at 18 GHz. With the addition of CNTs, the Fe<sub>3</sub>Al-0.5%CNT, Fe<sub>3</sub>Al-1%CNT, and Fe<sub>3</sub>Al-1.5%CNT samples show substantially decreased  $\mu'$  values of 1.58, 1.49, and 1.47, respectively. Since CNTs are not magnetic materials, the addition of CNTs will decrease the magnetic properties of the composite absorber, leading to a decreased capacity to store and dissipate magnetic energy.<sup>38–40</sup> Combined with the morphology characteristics shown in Fig. 1, the tubular CNT-coated Fe<sub>3</sub>Al flake structure weakens the magnetic properties of Fe<sub>3</sub>Al to some extent. Thus, with the increase of the CNT content, both the real part of magnetic permeability  $\mu'$  and the imaginary part of magnetic permeability  $\mu''$  decrease. In addition, both  $\mu'$  and  $\mu''$  change with the change of the frequency is caused by the frequency dispersion effect.<sup>41</sup>

Fig. 5 shows the curves of dielectric loss tangent ( $\tan \delta_\epsilon$ ) and magnetic loss tangent ( $\tan \delta_\mu$ ) versus the frequency of the CNTs/Fe<sub>3</sub>Al composite absorbers obtained after different levels of CNT additions. It is commonly accepted that  $\tan \delta_\epsilon = \epsilon''/\epsilon'$  and  $\tan \delta_\mu = \mu''/\mu'$  represent the dielectric loss capability and magnetic loss capability of an absorber, respectively. The larger value indicates the stronger attenuation capability of the material for electromagnetic waves. As demonstrated in Fig. 5a, the dielectric loss tangent ( $\tan \delta_\epsilon$ ) of the samples shows a wave-like increase with the increase in frequency. To summarize, the addition of CNTs makes the  $\tan \delta_\epsilon$  value of the composite absorbers increase. The main dielectric loss mechanisms of the Fe<sub>3</sub>Al/CNTs composite absorbers include the polarization effect generated by the unsaturated dangling bonds on the surface of the carbon nanotubes, and the conductive network formed by



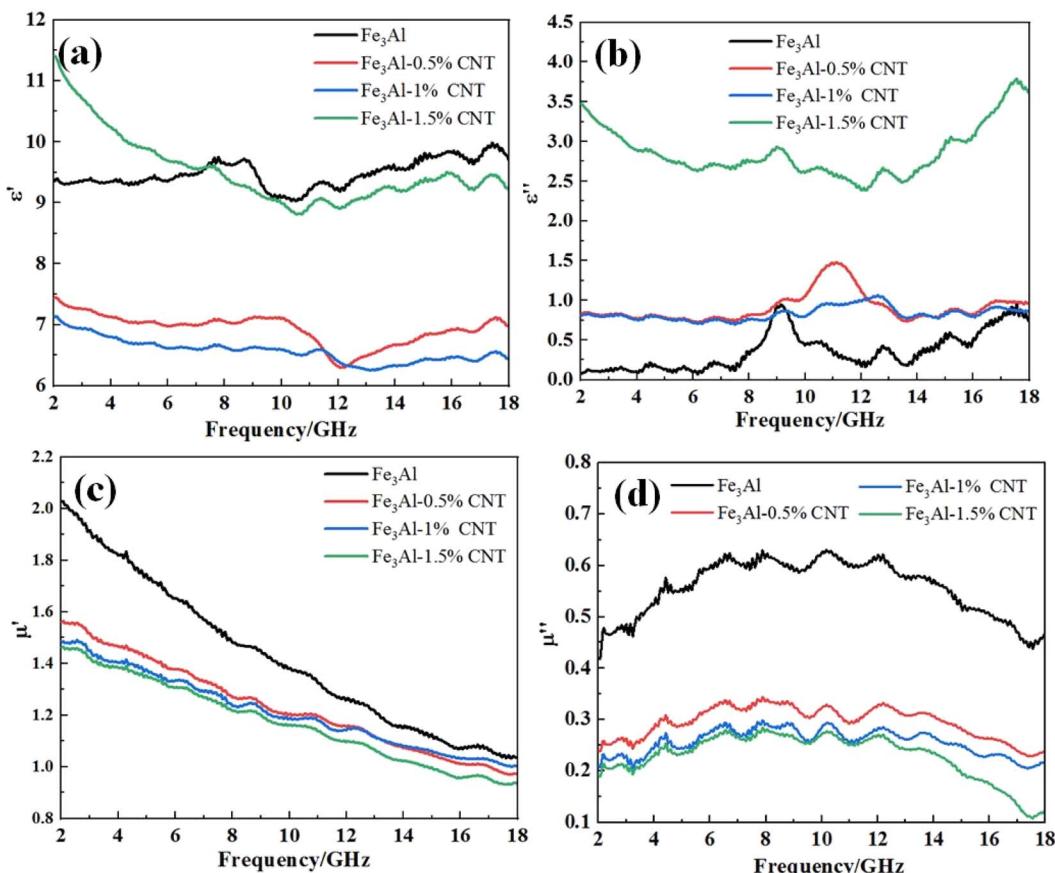


Fig. 4 Frequency dependence of (a) real and (b) imaginary parts of complex permittivity, frequency of dependence of (c) real and (d) imaginary parts of complex permeability curves of the Fe<sub>3</sub>Al/CNTs composite absorbers.

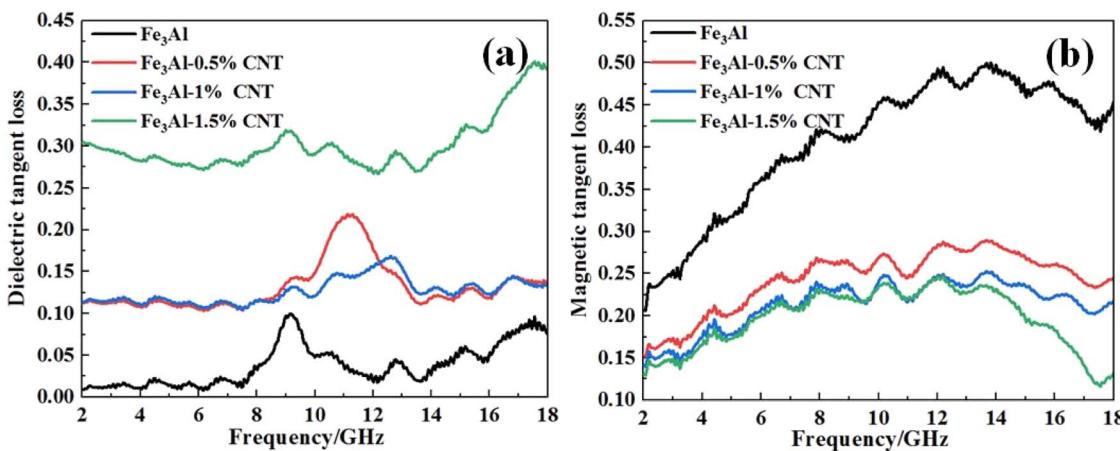


Fig. 5 Frequency dependence of (a) dielectric loss tangent and (b) magnetic loss tangent curves of the Fe<sub>3</sub>Al/CNTs composite absorbers.

the connection of the CNTs as well, which improves the migration rate of the carriers, and thus increasing the dielectric loss of the material.

Fig. 5b shows the curves of the magnetic loss tangent ( $\tan \delta_\mu$ ) versus the frequency of four composite absorbers. More specifically, the values of  $\tan \delta_\mu$  increase with the increase of the frequency, reaching a maximum value at 12–14 GHz. With the

further increase of the frequency,  $\tan \delta_\mu$  shows a decreasing trend. The reason for this phenomenon is due to the internal domain wall resonance and eddy current loss of the Fe<sub>3</sub>Al magnetic loss absorber. Note that the introduction of CNTs does make Fe<sub>3</sub>Al less capable for the magnetic loss of electromagnetic waves. Thus, with the increased content of CNTs, the  $\tan \delta_\mu$  decreases.

Generally, for the composite absorbers system containing two absorbers with completely different loss mechanisms, the comparable values of  $\tan \delta_e$  and  $\tan \delta_\mu$  can achieve excellent electromagnetic wave absorption performance. If the values of  $\tan \delta_e$  and  $\tan \delta_\mu$  are greatly different, the impedance mismatch of the materials will occur. As can be observed from Fig. 5,  $\tan \delta_e$  and  $\tan \delta_\mu$  of  $\text{Fe}_3\text{Al}$  are significantly different. With the addition of CNTs, the difference between  $\tan \delta_e$  and  $\tan \delta_\mu$  of all  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers is smaller, indicating that CNTs can improve the impedance matching for microwave absorption.

### 3.5 Absorbing properties

Fig. 6 shows the three-dimensional reflection loss diagrams of the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers with different levels of CNT additions at frequencies from 2 to 18 GHz with a thickness range of 1–5 mm, which is calculated according to the simulation of the electromagnetic parameters. It can be observed that the minimum reflection loss values of the four composite absorbers are less than  $-10$  dB, which means that up to 90% of the incident electromagnetic wave can be absorbed, exhibiting excellent electromagnetic wave absorption performance. For the pristine  $\text{Fe}_3\text{Al}$ , the minimum reflection loss peak of  $-52.42$  dB occurs at a thickness of 1.45 mm and a frequency of 14.45 GHz. However, with the addition of CNTs, the minimum reflection loss peaks of  $-21.22$ ,  $-18.53$  and  $-50.59$  dB are observed for  $\text{Fe}_3\text{Al}-0.5\%$ CNT,  $\text{Fe}_3\text{Al}-1\%$ CNT, and  $\text{Fe}_3\text{Al}-1.5\%$ CNT with the thicknesses of 2.45, 2.3 and 3.35 mm, respectively.

Fig. 7 shows the two-dimensional projection of the reflection loss of the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers. It can be observed that the maximum absorption bandwidth of  $\text{Fe}_3\text{Al}$  is 5.67 GHz (12.33–18.00 GHz) when the thickness is 1.4 mm.  $\text{Fe}_3\text{Al}-0.5\%$ CNT shows the maximum absorption bandwidth of 5.20 (11.40–16.60 GHz) when the thickness is 2.0 mm.  $\text{Fe}_3\text{Al}-1\%$ CNT exhibits the maximum absorption bandwidth of 5.02 (12.40–17.42 GHz) when the thickness is 1.8 mm. As for  $\text{Fe}_3\text{Al}-1.5\%$ CNT, the maximum absorption bandwidth is 5.91 (12.09–18.00 GHz) when the thickness is 1.7 mm. In addition,  $\text{Fe}_3\text{Al}$  exhibits strong absorption at high frequencies of 8–18 GHz, while  $\text{Fe}_3\text{Al}-1.5\%$ CNT exhibits strong absorption in a wider frequency range of 4–18 GHz. This phenomenon is attributed to the coupling effect between the dielectric absorber of CNTs and the magnetic absorber of  $\text{Fe}_3\text{Al}$ .<sup>42</sup>

The RL- $f$  curves of the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers at the same thickness of 2 mm calculated based on the transmission line theory equation are shown in Fig. 8. The effective absorption bandwidths ( $f_E$ ) of the four absorbers are 4.27, 5.20, 4.77 and 4.73 GHz, respectively, which indicates that with the addition of the appropriate amount of CNTs, the composite structure of the CNT-coated  $\text{Fe}_3\text{Al}$  flakes can achieve the expansion of the frequency bandwidth and improvement of the wave absorbing efficiency through the double absorption mechanisms of the dielectric loss and magnetic loss. The pristine  $\text{Fe}_3\text{Al}$  sample exhibits an  $RL_{\min}$  value of  $-23$  dB at a frequency of 10 GHz. When the CNT contents are 0.5% and 1%, the RL peaks shift to a higher frequency of 14 GHz and the  $RL_{\min}$  values for the  $\text{Fe}_3\text{Al}-0.5\%$ CNT and  $\text{Fe}_3\text{Al}-1\%$ CNT samples

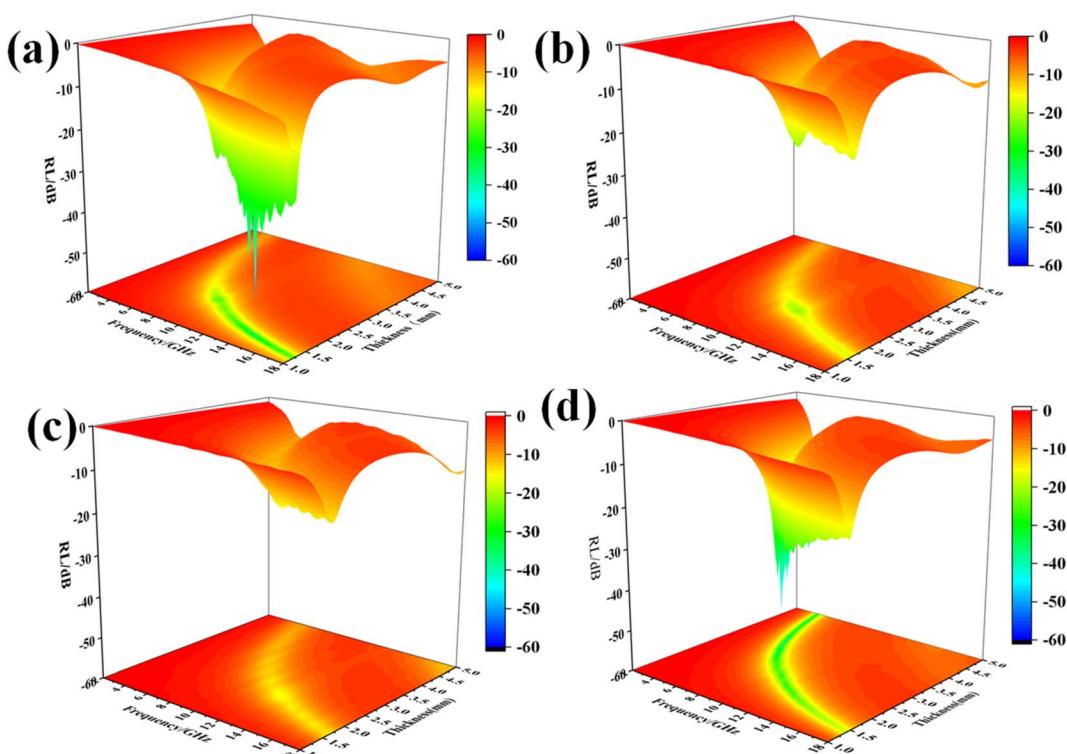


Fig. 6 Three-dimensional representation of the microwave reflection losses of the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers. (a)  $\text{Fe}_3\text{Al}$ , (b)  $\text{Fe}_3\text{Al}-0.5\%$ CNT, (c)  $\text{Fe}_3\text{Al}-1\%$ CNT, and (d)  $\text{Fe}_3\text{Al}-1.5\%$ CNT.



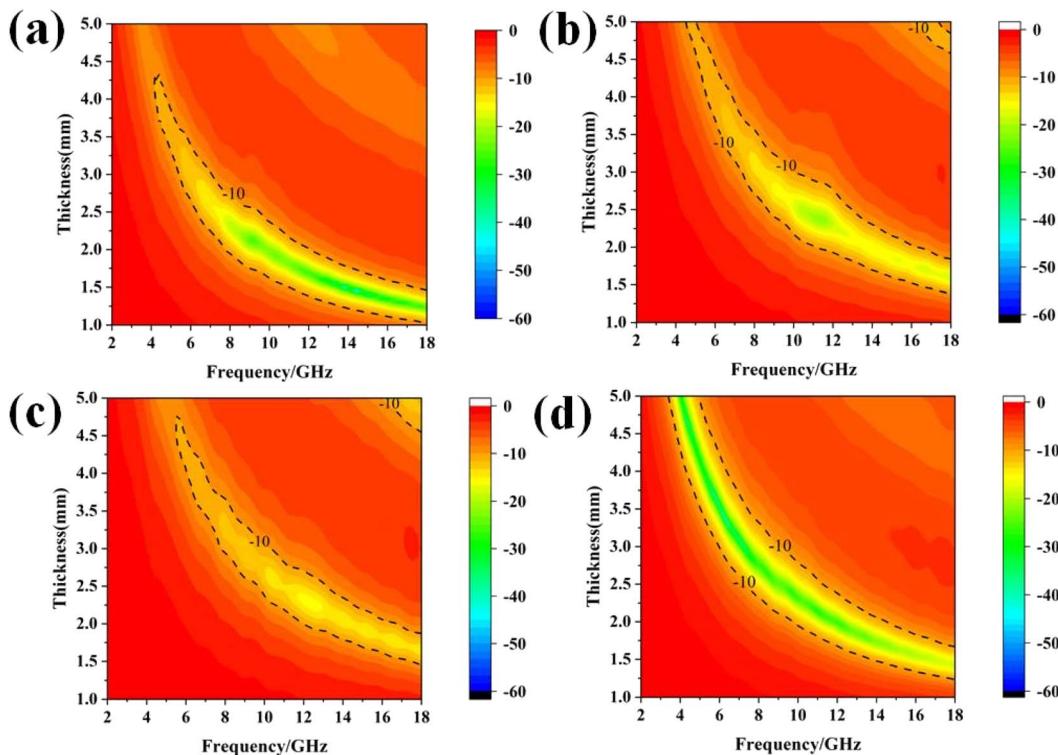


Fig. 7 Two-dimensional representation of the microwave reflection losses of  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers. (a)  $\text{Fe}_3\text{Al}$ , (b)  $\text{Fe}_3\text{Al}-0.5\%\text{CNT}$ , (c)  $\text{Fe}_3\text{Al}-1\%\text{CNT}$ , and (d)  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$ .

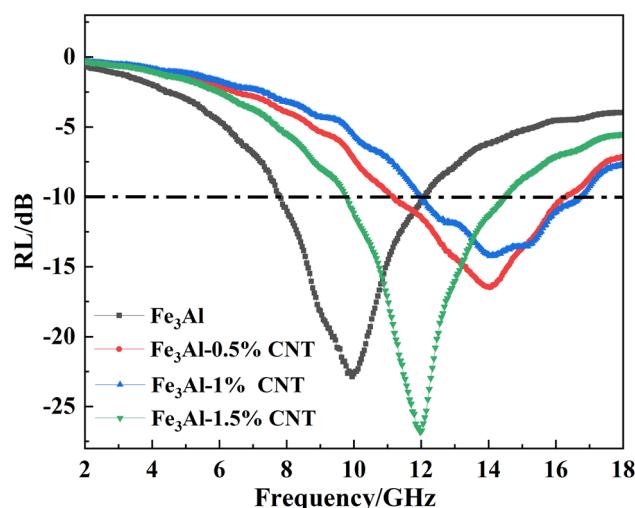


Fig. 8 Microwave reflection loss of the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers with a thickness of 2 mm.

are significantly decreased to  $-17$  dB and  $-14$  dB, respectively. However, when the CNT content is 1.5%, the RL peak negatively shifts to 12 GHz with the  $\text{RL}_{\min}$  value significantly reduced to  $-27$  dB.

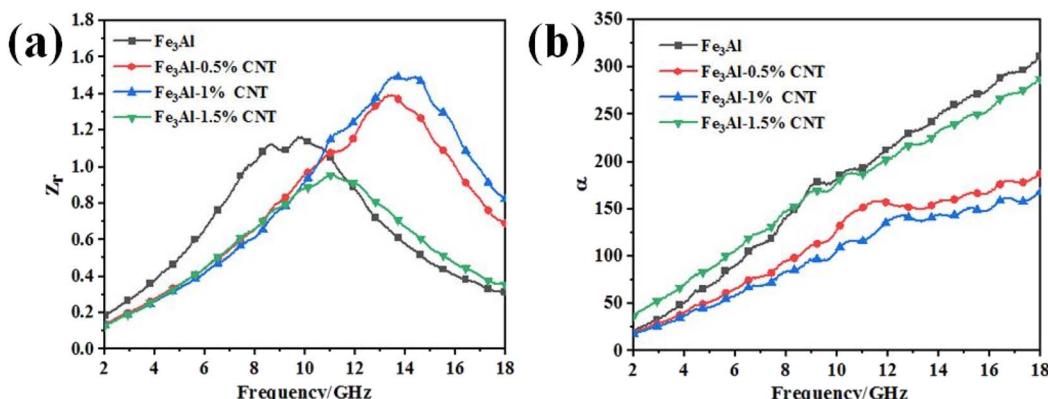
To evaluate the microwave absorption performance of the as-synthesized  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  composite, the minimum reflection loss, the corresponding thickness and the effective bandwidth at a certain thickness of the metal, alloy and oxide-based

composites reported recently are listed in Table 1. It can be observed that the comprehensive performance of the  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  composite is relatively good. It should be mentioned that the mass ratio of 4 : 1 (equal to a filler ratio of 80 wt%) for the as-synthesized  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  composite mixed with paraffin wax is necessary to obtain satisfactory performance. The microwave absorption performance of the  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  composite decreases significantly with other filler ratios of 60 wt%, 40 wt%, and 20 wt% (Fig. S1, ESI†).

Effective impedance matching and strong attenuation are the two key factors in achieving excellent microwave absorption properties of an absorber.<sup>52–54</sup> The synergistic effect between the magnetic loss of  $\text{Fe}_3\text{Al}$  and dielectric loss of CNTs in the  $\text{Fe}_3\text{Al}/\text{CNTs}$  composite absorbers can effectively increase the attenuation of the incident microwaves. Good impedance matching can ensure the incident microwave is absorbed in the absorber without reflection, while strong attenuation can convert the absorbed microwave into thermal energy or be dissipated through interference, resulting in enhanced microwave absorption performance. Generally, the impedance matching ratio is defined as  $Z = |Z_{\text{in}}/Z_0|$  (where  $Z_{\text{in}}$  is the impedance of the absorber, and  $Z_0$  is the impedance of the free space).<sup>55–57</sup> When the impedance matching ratio ( $Z$ ) is close to 1, the absorber can absorb the maximum incident microwaves.<sup>58</sup> Fig. 9a shows the impedance matching ratios of the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers with a thickness of 2 mm, it can be observed that the peak of the  $Z$  value of  $\text{Fe}_3\text{Al}-1.5\%\text{CNT}$  is the closest to 1 in all samples, indicating the best impedance matching to absorb more incident microwaves.

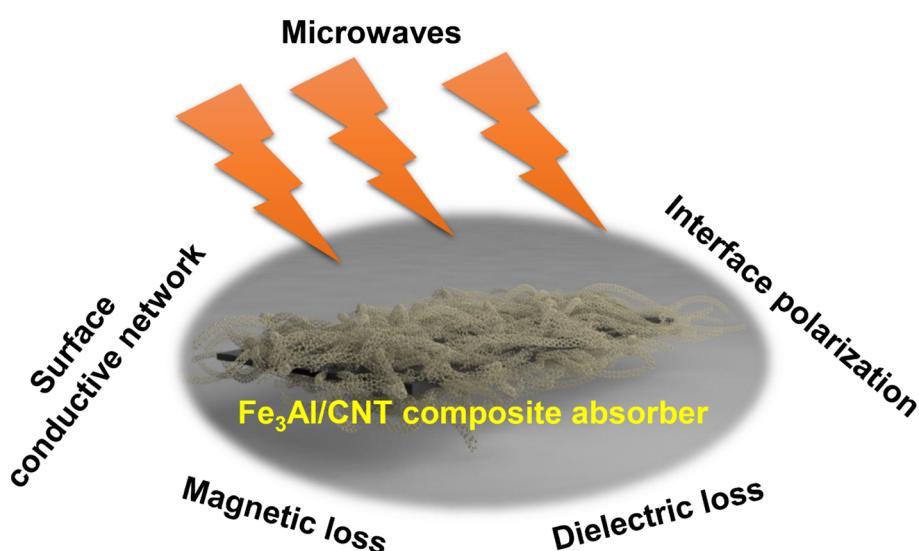
Table 1 Microwave absorption properties of metal, alloy and oxide-based composites in the frequency range of 2–18 GHz in recent years

Absorber	RL <sub>min</sub> (dB)	Thickness (mm)	Effective bandwidth (GHz) RL < -10 dB	Ref.
Fe <sub>3</sub> Al-1.5%CNT	<b>-50.59</b>	3.35	<b>5.91 (1.7 mm)</b>	This work
C@Ni <sub>x</sub> Co <sub>1-x</sub> Fe <sub>2</sub> O <sub>4</sub>	-51	1.9	3.3 (1.9 mm)	42
Doped, conductive SiO <sub>2</sub> nanoparticles	-55.09	4.2	~5 (1.3 mm)	43
Ni/CNTs	-49.3	3.0	2.96 (3.0 mm)	44
Al/H <sub>2</sub> treated TiO <sub>2</sub>	-58.02	3.1	6.13 (1.61 mm)	45
Fe <sub>3</sub> O <sub>4</sub> @C	-64.92	1.75	4.64 (1.75 mm)	46
Mg/H <sub>2</sub> treated TiO <sub>2</sub>	-34.69	1.7	3.96 (0.92 mm)	47
Co@C	-53.99	2.28	6.00 (2.73 mm)	48
CoNi/C@PPy	-68.78	2.43	5.62 (2.10 mm)	49
Co@NC/Ni	-47.10	2.5	6.84 (2.5 mm)	50
C@Ni <sub>0.5</sub> Zn <sub>0.5</sub> Fe <sub>2</sub> O <sub>4</sub>	-26.87	1.8	4.05 (1.8 mm)	51

Fig. 9 (a) Impedance matching ratio and (b) attenuation constant  $\alpha$  of the Fe<sub>3</sub>Al/CNTs composite absorbers with a thickness of 2 mm.

The attenuation constant ( $\alpha$ ) *versus* the frequency curves of the Fe<sub>3</sub>Al/CNT composite absorbers with a thickness of 2 mm are shown in Fig. 9b. It can be observed that the  $\alpha$  values of all samples increase with the increase of frequency, suggesting that

the absorbed microwave at a higher frequency is easier to be attenuated.<sup>59</sup> In addition, with the addition of CNTs, the  $\alpha$  value decreases compared to pristine Fe<sub>3</sub>Al, indicating that CNTs have some negative effect on the attenuation of the absorbed

Fig. 10 Schematic of microwave absorption in a Fe<sub>3</sub>Al/CNT composite absorber.

microwaves for  $\text{Fe}_3\text{Al}$ . However, when the CNT content is 1.5%, the  $\alpha$  *versus* frequency curve of  $\text{Fe}_3\text{Al}$ -1.5%CNT is almost overlapped with that of its pristine  $\text{Fe}_3\text{Al}$  counterpart, suggesting the strongest attenuation constant among all  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers.

Based on the above analysis, the main microwave absorption mechanisms of a  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorber are shown in Fig. 10. The conductive network formed by the surface CNTs can significantly increase the conductive loss, which can convert the microwave energy into heat. In addition, the interfaces between  $\text{Fe}_3\text{Al}$  and CNTs generate interface polarization, which can also dissipate the microwave energy. Furthermore, the magnetic loss and dielectric loss also contribute to the absorption and attenuation of the microwaves. Therefore, the combination of CNTs with  $\text{Fe}_3\text{Al}$  can effectively increase the microwave absorption performance.

## 4. Conclusion

$\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers with different CNT contents were synthesized by a mechanical blending method, and the CNTs were attached to the surface of the  $\text{Fe}_3\text{Al}$  flakes. Since CNT is a dielectric-loss absorber while  $\text{Fe}_3\text{Al}$  is a magnetic-loss absorber, the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorber can combine both dielectric-loss and magnetic-loss mechanisms to promote microwave absorption. In particular, the  $\text{Fe}_3\text{Al}$ -1.5%CNT composite absorber has the best impedance matching ratio and the strongest attenuation constant among all the  $\text{Fe}_3\text{Al}/\text{CNT}$  composite absorbers, thus exhibiting excellent microwave absorption properties, with an effective absorption bandwidth ( $\text{RL} \leq -10$  dB) of 4.73 GHz at a thickness of 2 mm. The results are only applicable to several grams of samples prepared in the lab. Uniformity and consistency should be considered for possible scale-up applications in future.

## Consent for publication

All authors agree to the submission and publication of article materials.

## Author contributions

All authors contributed meaningfully to all aspects of experimentation, analysis and composition.

## Conflicts of interest

All the authors have no competing interests to declare.

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