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Electromagnetic and Microwave Absorption Properties of the Carbonyl Iron/Tic Hybrid Powders in the X Band

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Abstract

The electromagnetic and microwave absorption properties of the carbonyl iron/TiC hybrid powders are studied in the X band. A favorable microwave absorption property can be obtained by changing the ratio of carbonyl iron (CI) and TiC. When the TiC content was 25%, the hybrid powders have the optimum microwave absorption property. The bandwidth below -20 dB can be obtained in 9.4-10.5 GHz and the minimum reflection loss value was -25.1 dB. The results indicated that the CI/TiC hybrid powders with appropriate ratio are favorable for application in the X band as microwave absorption absorbent owing to integrating the dielectric absorbent and magnetic absorbent.

Keywords

Carbonyl iron, TiC, Dielectric properties, Magnetic properties, Microwave absorption property

Introduction

Electromagnetic wave absorption materials have attracted much interest due to the increase in electromagnetic pollution, which caused by the drastic development of gigahertz (GHz) electronic systems and telecommunication [1,2]. According to the wave absorbing mechanism, the electromagnetic wave absorption materials could be divided into magnetic and dielectric absorbers. Generally speaking, though thin thickness can have excellent absorption ability in low frequency range, magnetic absorbers are restricted by poor characteristics in GHz frequency due to the Snoek's limit and heavy density; The dielectric absorbers are used in high frequency range, and thick matching thickness are required [3-5]. Thus, it is eager to explore absorber with width-frequency band, low density, high compatibility and thin thickness to overcome the shortcomings of using single absorber. By combing the properties of magnetic absorbers and dielectric absorbers are a good choice to solve this problem.

Previously, R.C. Che *et al.* [6] studied the microwave absorption property of carbon nanotubes/$CoFe_2O_4$ spinel nanocomposite and the reflection loss value of pure CNTs and $CoFe_2O_4$ was rather small for all frequencies range between 2-18 GHz when compared with CNTs/$CoFe_2O_4$ nanocomposites. The improvement of microwave absorption obviously originated from the combination of CNTs and $CoFe_2O_4$. Liu *et al.* [7] prepared the Fe_2B/C (a) absorbent using a melt-spun method, the reflection loss below -20 dB could be obtained in the frequencies range of 7.5-16GHz with the thickness of 1.2-2.2 mm. However, their practical applications were restricted due to the complex preparation process of these composites. Zhang *et al.* [3] fabricated the carbonyl iron/MnO_2 composite by a mechanically milling method and the experimental results showed that the composite was a promising candidate for microwave absorbing in S-band (2-4 GHz) and C-band (4-8 GHz). Zhou *et al.* [8] explored the microwave absorption properties of ZnO/carbonyl iron composites, and the reflection loss below -8 dB was obtained in the frequency range from 9.8-14.9 GHz with thick thinness of 3 mm. However, the absorbing materials either with low absorption ability in the X band or with thick thickness, thus it is needed to fabricate a kind of absorbent with good absorption properties in the X band at the same time with thin thickness.

Carbonyl iron (CI) particles with relatively low electrical conductivity, high Curie temperature, and high specific saturation magnetization intensity in comparison with other metallic particles are often used as conventional magnetic fillers [9]. Also, in order to suppress the eddy current losses induced by the EM wave and enhance the effective interaction with EM wave absorbers, it is better to use metallic particles with a size smaller than the skin depth [10]. Titanium carbide (TiC) due to its high melting point, good thermal conductivity, excellent chemical stability, electrical and especially its high conductivity made it a good candidate as microwave absorbent for electromagnetic wave absorption [11,12]. However, seldom reports have been studied the electromagnetic properties of carbonyl iron/TiC in the X band. In this paper, the electromagnetic and microwave absorption properties of carbonyl iron/TiC were

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studied in the X band and expected a kind of thin thickness and good absorption ability absorbent can be fabricated by integrating the advantages of magnetic absorbent and dielectric absorbent.

Experimental

Materials

Carbonyl iron particles with average diameter of 1-5 μm and thickness below 1 um were obtained from Xinghua Chemical CO. Ltd, Shaanxi province, China, which was produced by decomposition of $Fe(CO)_5$, the content of α-iron > 99.5 wt%. The TiC powder was purchased from Guang Yuan Material Co. Ltd, Hunan province, China and the diameter of TiC was about 1-6 μm with the purity of 99%. All materials were used as received without any further purification.

Carbonyl iron powders and TiC powders were mixed at different weight fractions, and the TiC weight fractions are 0, 25%, 50%, 75% and 100% and then the mixed powders were mechanically milled for 3 h on a high energy ball mill, The rotation speed was kept for 300 rpm, the ball to power mass ratio was 10:1. For convenience, the hybrid powders with different TiC weight fractions were denoted as TiC 0%, TiC 25%, TiC 50%, TiC 75%, and TiC 100%, respectively. The hybrid powders were homogeneously dispersed in paraffin and pressed into rectangle with the size of 10.16 × 22.86 × 2.5 mm, the mass ratio of hybrid powders to paraffin wax was 3:1.

Characterization

X-ray diffraction (XRD) of the materials was carried out by Mac Science M18X-ray diffraction spectrometer using Cu Ka radiation (50 Kv, 100 mA). The evaluation of the microwave absorption properties was tested by network analyzer (Agilent technologies E8362B:10MHz-20GHz) based on the measurements of the reflection and transmission module between 8.2 and 12.4 GHz in the fundamental wave-guide mode TE10. In addition, the morphology of the raw CI particles and TiC powder were examined under a scanning electron microscope (SEM, JEOL JSM-5800 LV SKANNING).

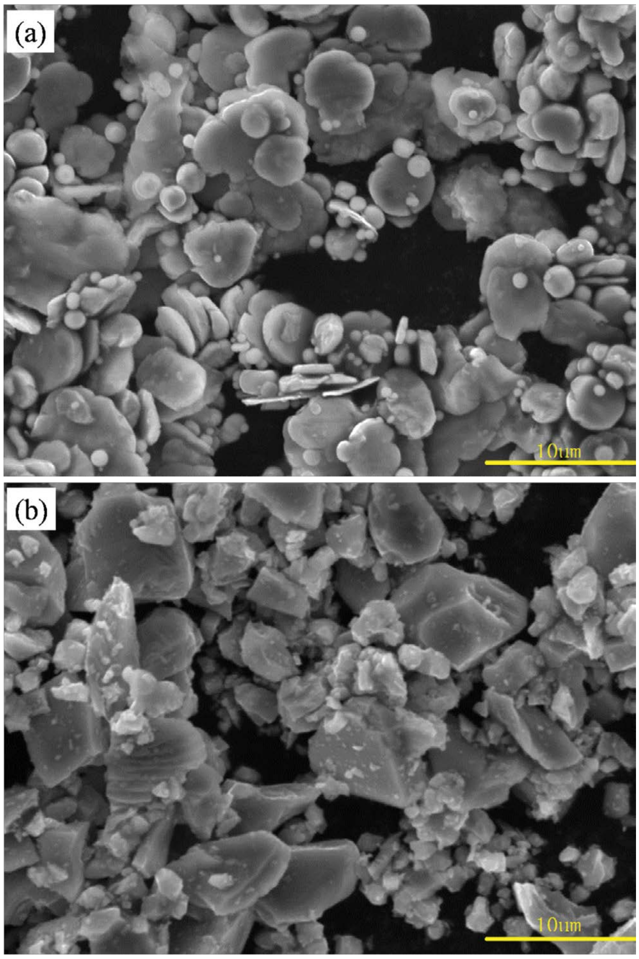

Figure 1: The size and surface morphology of (a) CI particles and (b) TiC particles.

Results and Discussion

The morphology and XRD patterns of carbonyl iron and TiC

Figure 1 shows the size and surface morphology of CI particles and TiC particles. It could be found that the CI particles are flaky with the average diameter 1~5μm and thickness below 1um, and also exhibit a relatively smooth surface. The average size of TiC particles is about 1-6 μm with laminated appearance.

Figure 2 shows the XRD patterns of CI and TiC particles. As can be seen, three main diffraction peaks associated with the (110), (200) and (211) planes of cubic α-Fe are located at 2θ = 44.6°, 64.9° and 82.3°. No other peaks of impurities are detected. The main diffraction peaks of TiC are consistent with the cubic TiC (JCPDS# 32-1383).

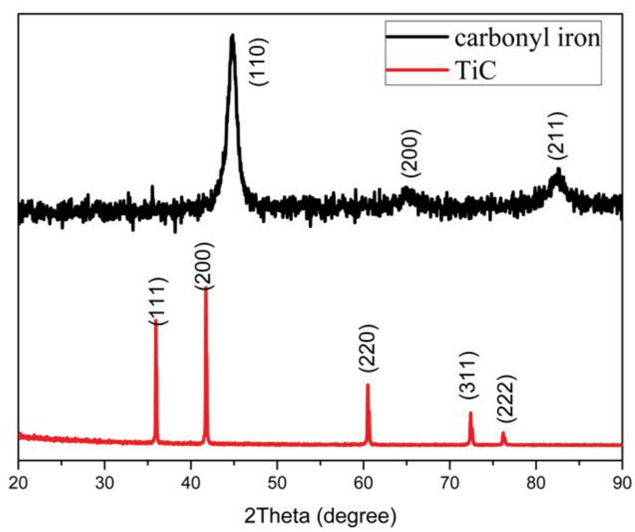

Figure 2: The XRD patterns of the carbonyl iron and TiC particles.

The electromagnetic property of the carbonyl iron/TiC hybrid powders

The complex permittivity and complex permeability of the composites with different weight fraction of TiC are presented in figure 3. It can be observed that the complex permittivity of the hybrid powders is closely correlated with the weight fraction of TiC. Both the real part (ε´) and the imaginary part (ε´´) of permittivity rise across the whole frequency range with increasing of the content of TiC, and also from TiC 0% to TiC 75%, the values of ε´ and ε´´ are almost constant in the X band. For TiC 0%, the values of ε´ and ε´´ are 10.66 and 0.07 at 8.2GHz respectively. Compared with TiC 0%, the values are 11.36 and 0.14 for TiC 25%, 11.81 and 0.21 for TiC 50%, 12.38 and 0.51 for TiC 75% and 12.95 and 1.64 for TiC 100%, respectively. Obviously, the increase of ε´ and ε´´ is attributed to increase the TiC content. The ε´ is known as an expression of the polarization ability of materials. In this case, the ε´ could be mainly attributed to the space charge polarization. The space charge polarization is associated with the heterogeneity presents at the interfaces between the components of the hybrid powders [13], the interfaces of TiC and CI, TiC and paraffin and CI and paraffin. The difference in the conductivity between CI and TiC is responsible for the generation of space charge and also its polarization. TiC is a kind of dielectric loss absorber and the electrical conductivity is 3.0×10^8 s/m, while the electrical

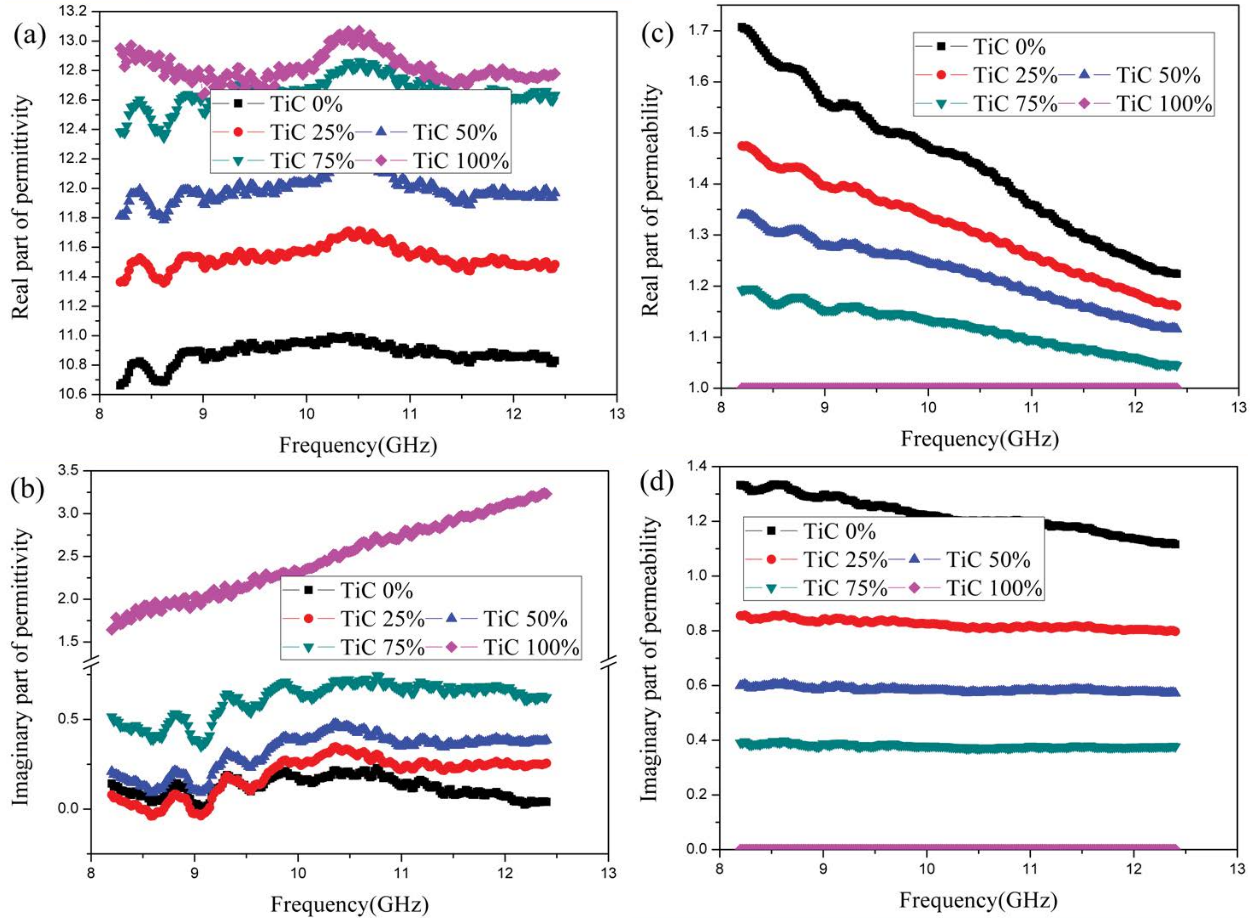

Figure 3: The complex permittivity (a & b) and permeability (c & d) of the composites with different weight fraction of TiC.

conductivity of CI is about 1×10^7 s/m [14]. According to the mixture rule, the electrical conductivity of the composites increases with increasing the TiC content.

The ε´´ is known as an expression of the loss ability of materials. According to Debye equation, the increase in the imaginary component of the permittivity as the content of filler increase may be the consequence of the increase of the relaxation polarization loss and the electric conductance loss [15].

$$\varepsilon'' = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau / (1+(\omega\tau)^2) + \sigma}{\varepsilon_0 \omega}$$

Where ε_s is the static permittivity, ε_∞ is the permittivity at the high frequency limit, ω is the angular frequency, τ is the relaxation time, σ is the electrical conductivity of the composites. It indicated that the imaginary part of permittivity increase as an increase of the dielectric conductivity and polarization loss. In this case, the increase of ε´´ could be mainly attributed to the relaxation polarization loss and the electric conductance loss.

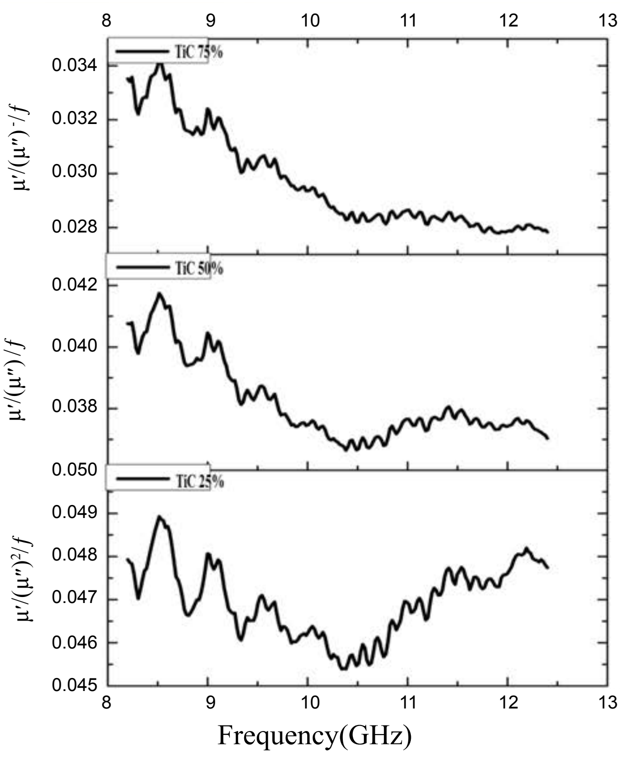

Figure 4: The values of μ´´/(μ´)^2/f as function of frequency.

Figure 3 also shows the complex permeability of the composites with different TiC weight content as a function of frequency. There is no doubt both the real part (μ´) and imaginary part (μ´´) of permeability should decrease with the increase of TiC content since the complex permeability of the hybrid powders mainly comes from CI. It is can also be found that the μ´ gradually decrease with the increase of the frequency owing to the domain-wall motion and relaxation [16]. The magnetic loss mainly consists of natural resonance, domain wall displacement, magnetic hysteresis and eddy current loss, and according to previous study, the hysteresis loss is negligible in weak applied electromagnetic field, and the domain wall resonance occurs for the most part only in the 1-100 MHz. If the eddy current loss contributed to the imaginary part of permeability, the values of μ´´/(μ´)^2/f should be a constant when the frequency is varied [17]. However, as depicted in figure 4, the values of μ´´/(μ´)^2/f

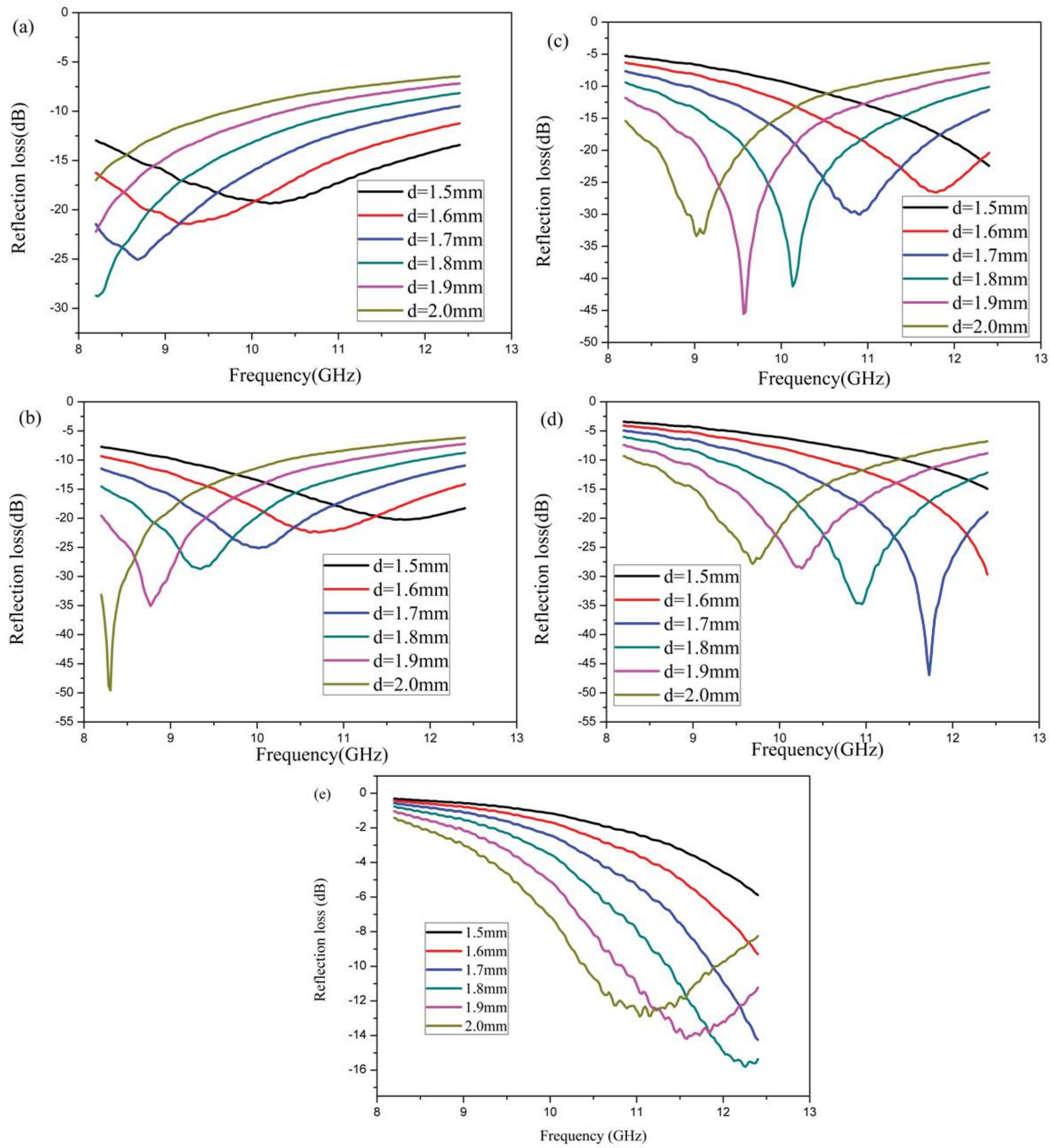

Figure 5: The reflection loss (RL) of the composites including different content of TiC were calculated **(a)** TiC 0%, **(b)** TiC 25%, **(c)** TiC 50%, **(d)** TiC 75% and **(e)** TiC 100%.

Table 1: Microwave absorbing properties of the composites.

Sample	Optimal RL (dB)	Dm (mm)	f_m (GHz) (Optimal RL)	(RL < -5dB)	(RL<-10dB)	(RL<-20dB)
TiC 0%	-19.4 dB	1.5 mm	10.2 GHz	8.2-12.4	8.2-12.4	-
TiC 25%	-25.1 dB	1.7 mm	10 GHz	8.2-12.4	8.2-12.4	9.4-10.5 GHz
TiC 50%	-41.3 dB	1.8 mm	10.1 GHz	8.2-12.4	8.3-12.4	9.6-10.7 GHz
TiC 75%	-34.6 dB	1.8 mm	10.8 GHz	8.2-12.4	9.3-12.4	10.4-11.5 GHz
TiC 100%	-12.8 dB	2.0 mm	11.0 GHz	9.6-12.4	10.4-11.9-	-

decrease with increasing the frequency. Therefore, it means that the magnetic loss is mainly caused by the natural resonance [17,18].

Microwave absorption properties

To further reveal the influence of CI/TiC ratio on the EM-wave absorption properties, based on the above relative complex permittivity and complex permeability at the given frequency, the reflection loss (RL) of the composites including different TiC content were calculated, as shown in Figure 5. It can be seen, all the absorption peaks shift to low frequency direction with increasing the thickness in the 8.2-12.4 GHz range. For TiC 0%, the minimum reflection loss value reaches -28.6 dB at 8.2 GHz with the thickness of 1.8 mm, while these values are -49.6 dB, 8.3 GHz, 2 mm for TiC 25%, -45.5 dB, 9.5 GHz, 1.9 mm for TiC 50%, -46.9 dB, 11.7GHz, 1.7 mm for TiC 75% and -15.6 dB, 12.3 GHz, 1.8 mm for TiC 100%, respectively. The optimal reflection loss and the bandwidth below -5 dB, -10 dB and -20 dB (99% power absorption) are listed in Table 1. When only using magnetic flaky CI as absorbent, the bandwidth (< -10dB) can be obtain in the whole frequency range with the thickness of 1.5mm, and there was no RL values below -20 dB. When only using dielectric absorbent TiC, it cannot get good absorbing properties with thin thickness, when the thickness was 2.0 mm, the optimum RL reached and the -10 dB bandwidth was 1.5 GHz. By integrating the two absorbent, the bandwidth and optimal reflection loss both increase, the bandwidth below -20 dB can obtain in 9.4-10.5 GHz for TiC 25%, 9.6-10.7 GHz for TiC 50%. Further increase the TiC content, the absorption properties became worse due to the main absorbent was dielectric absorbent. As we know, using magnetic absorbent, the absorbers have thin thickness while for the dielectric absorbent, thick thickness was needed. Therefore, if the hybrid powders want to have good absorption properties, the simulated RL curves should choose thick thickness. The variation in reflection loss results of the samples is coherent with the variation in the complex permittivity and permeability. With the increasing TiC content in the hybrid filler, the dielectric loss increases while the magnetic loss decreases. As we know, the design of EM waves absorbing materials with low reflection requires two important conditions [19]: impedance matching characteristic and attenuation characteristic. The impedance matching degree is determined by the combination of the six parameters, ε´´, ε´, μ´´, μ´, d and w, only the optimal combination of the six parameters, the impedance matching between the surface of the RAM and the air could be achieved good absorption properties. The attenuation constant **γ** can be calculated according the following equation:

$$\gamma = \omega(\mu'\mu_0\varepsilon'\varepsilon_0)^{1/2}\left\{2\left[\frac{\mu''\varepsilon''}{\mu'\varepsilon'}-1+\left(1+\frac{\mu''^2}{\mu'^2}+\frac{\varepsilon''^2}{\varepsilon'^2}+\frac{\mu''^2\varepsilon''^2}{\mu'^2\varepsilon'^2}\right)^{1/2}\right]\right\}$$

While ω is the angular frequency, the optimum RL can be explained by the considerable dielectric and magnetic loss in the absorbing materials. The results indicated that the CI/TiC composite with appropriate thickness and amount of TiC content is favorable for application in the X band as microwave absorption material.

Conclusions

In this study, carbonyl iron/TiC composite materials were successfully fabricated by the simple mechanical method and the electromagnetic and microwave absorption property of the hybrid powders are investigated. Complex permittivity increases while complex permeability decreases with increasing the TiC content due to the high electrical conductivity of TiC compared with CI and TiC is non magnetic material. When only using magnetic flaky CI as absorbent, the bandwidth (< -10 dB) can be obtain in the whole frequency range with the thickness of 1.5 mm, and there was no RL values below -20 dB. When only using dielectric absorbent TiC, it cannot get good absorbing properties with thin thickness, when the thickness was 2.0 mm, the optimum RL reached and the -10 dB bandwidth was 1.5 GHz. The hybrid powders have good absorption properties suggesting that both the bandwidth and optimal reflection loss increase by integrating the dielectric absorbent and magnetic absorbent. The microwave absorption property of the composites is optimized by mixing the ratio of the CI/TiC hybrid absorbents

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