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Article

Lightweight Co₃O₄/CC Composites with High Microwave Absorption at Low Frequency

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Abstract: With the rapid development of electronic and communication technology in military radar, the demand for microwave absorbing materials in low-frequency with thin layer is growing increasingly. In this study, flexible Co₃O₄/CC (carbon cloth) composites derives from Co-MOFs (metal-organic frameworks)/CC are prepared by hydrothermal and thermal treatment processes. The flexible precursors of Co-MOFs/CC are calcined with different calcination temperatures, which the material structure, dielectric properties and microwave absorption performance are changed. With the increase of calcination temperature, the minimum reflection loss of the corresponding Co₃O₄/CC composites gradually moves to the lower frequency with thinner thickness. In addition, the Co₃O₄/CC composites with 25 wt% filler loading ratio exhibit the minimum reflection loss (RL) of -46.59 dB at 6.24 GHz with 4.2 mm thickness. When the thickness is 3.70 mm, the effective absorption bandwidth is 3.04 GHz from 5.84 to 8.88 GHz. This study not only proves that the composite Co₃O₄/CC is a kind of outstanding microwave absorbing material with better flexibility, but also provides a useful inspiration for the low frequency and broadband microwave absorbing material.

Keywords: Low frequency; Co₃O₄/CC; Co-MOFs/CC; thinner thickness

1. Introduction

With the rapid development of science and technology, the influences of electromagnetic radiation on human body and environment have attracted extensive attention. In terms of this background, the development of microwave absorption materials can resist electromagnetic radiation and it has become a research hotspot in the world [1]. An ideal microwave absorbing material should have excellent properties such as light weight, thin thickness, strong reflection and broad effective absorption bandwidth [2]. In recent years, researchers have studied a variety of ideal microwave absorbing materials including the magnetic loss materials and dielectric loss materials. For instance, Wang et al. synthesized the Fe₃O₄@SnO₂/RGO ternary composites with an efficient and rapid three-step method improving the impedance matching and obtaining excellent electromagnetic wave absorption performance [3]. Moreover, ZnO [4], Fe₂O₃/N-Graphene/CNTs [5], composite nonlinear feedback (CNF)/SiO₂ [6] and MOFs derivatives [7] et al. have also been widely developed to improve the attenuation characteristics of electromagnetic waves. In view of the loss mechanism of electromagnetic waves, it is generally easier to achieve strong broadband microwave absorption at high frequencies. However, the absorbing materials at low-frequency normally have thick thickness and narrow frequency bandwidth, which is difficult to meet the needs of practical application. Therefore, it is essential to research lightweight thinner layer with wide bandwidth absorber at low frequency.

Among a large number of lightweight electromagnetic absorption materials, MOFs derivatives have been extensively manufactured because of their variety of metals, diversity of morphology and highly surface areas [8–11]. For example, Ma et al. synthesized TiO₂/C nano-porous carbon composites by pyrolysis of MIL-125 (Ti-based MOFs), which exhibited maximum reflection loss of -49.60 dB with

thinner thickness of 1.6 mm and obtained 4.6 GHz effective absorption bandwidth [12]. In addition, Shi et al. studied the nanoporous Co_3O_4 nanosheets derived from Co-MOFs, when the matching thickness is 2.5 mm, the maximum reflection loss can reach -32 dB, and the effective absorption band width can reach 4.2 GHz because of its better impedance matching of the composites [13]. Besides, Liu et al. also designed Co-MOFs derivatives to improve the absorption performance in the low frequency. The better impedance matching has been achieved with the Co/C composite, which minimum reflection loss value was about -20 dB and the effective absorption bandwidth reached 3.84 GHz in C-band frequency range [14]. Therefore, MOFs derivatives could obtain broadband absorption performances with thinner thickness in low frequency. In addition, MOFs can be combined with other functional materials to improve the microwave absorption performance. For example, Yang et al. fabricated SiC/Ni/NiO/C by annealing SiC/Ni-MOFs nanoparticles, which minimal RL value reached -50.52 dB [15]. Furthermore, Fe/Fe₃C/C derived from MIL-101-Fe and MIL-88-Fe was obtained by Miao et al. The minimum RL of the composites is -59.2 dB with a thickness of 4.32 mm and the effective bandwidth achieve 6.5 GHz with 2 mm [16]. Thus, the combination of MOFs derivatives and another material is better for the loss of microwave.

In recent decades, carbon cloth (CC) has been applied to various fields owing to its light weight, high strength, corrosion resistance and aging resistance [17-23]. Luo et al. prepared NiSe₂/CC as an electrode material without adhesive, which show good hydrogen evolution reaction performance and chemical stability in 0.5 mol/LH₂SO₄ solution [24]. Zhan et al. grow porous arrays of CoP nanoparticles derived from MOFs on carbon cloth using for effective alkaline hydrogen evolution [25]. Along with these, carbon cloth can also be used to prepare flexible microwave absorber. Che et al. made ZnO grow on CC flexible substrate, which constructed three-dimensional conductive network with multiple interfaces [26]. Moreover, Liu et al. grow of Co-MOF on conductive CC, which derived CC@NPC/CoS₂ composites of RL reaching -59.6 dB with 2.8 mm [27]. Based on the above, the carbon cloth can be combined with metal-organic framework materials for preparing excellent electromagnetic wave absorber with thin layer.

In this study, the Co_3O_4 /CC derived from Co-MOFs/CC is synthesized, the result indicates that the absorbing properties of Co_3O_4 /CC composite can be adjusted by controlling different calcination temperatures. When the calcination temperature is 500 °C, the Co_3O_4 /CC composites showed an RL_{min} (minimum reflection loss) of -46.59 dB at 6.24 GHz with a filler loading ratio of 25 wt%. Furthermore, the Co_3O_4 /CC material exhibits an effective microwave absorption bandwidth of 3.04 GHz from 5.84 GHz to 8.88 GHz with a thickness of 3.70 mm. Thus, the Co_3O_4 /CC composite derived from the metal-organic framework/CC can be as a promising wide-band absorbing material with thin layer in low frequency.

2. Materials and Methods

2.1. Preparation of Precursors of Co-MOFs/CC

A piece of 2*2 cm² CC was placed in acetone cleaning by deionized water and ethanol, which was ultrasonicated for 20 min respectively and then dried in a vacuum oven at 60 °C for 6 h. The preparation of Co-MOFs/CC is as Figure 1, dissolving 432 mg cobalt nitrate hexahydrate, 300 mg terephthalic acid and 3 g PVP K30 (Polyvinylpyrrolidone K30) in a 60 mL mixture solution (deionized water: DMF: ethanol =1:1:1 v/v/v) and stirring vigorously with a blender until completely dissolved. Then put the cleaning CC into the bottom of the 100 mL Teflon autoclave, and transfer the mixture solution into it. Finally, the Teflon autoclave was heated in an oven at 150 °C for 10 hours, and the Co-MOFs/CC was obtained. The precursor of Co-MOFs/CC was obtained after washing with deionized water and ethanol for three times and drying in an oven at 60 °C for 12 hours.

2.2. Synthesis of Co_3O_4 /CC Composites

The Co-MOFs/CC composite was heated to 300 °C, 400 °C, 500 °C respectively in a muffle furnace with a heating rate of 2 °C/min and heat two hours to obtain multilayer Co_3O_4 /CC composites, named as S1, S2 and S3, respectively.

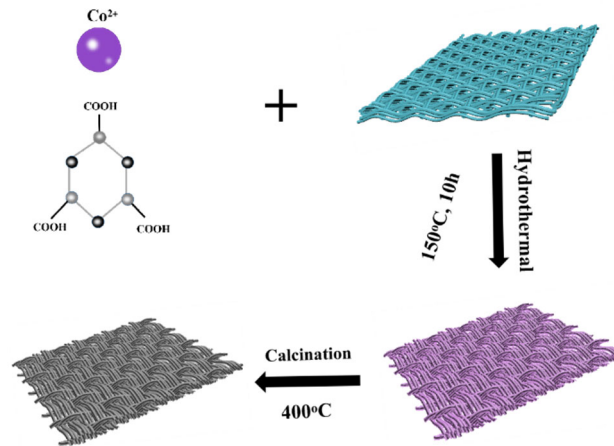


Figure 1. The structure diagram of experimental process.

2.3. Characterization and Measurement

The morphology of $\text{Co}_3\text{O}_4/\text{CC}$ and pure CC was characterized by field emission scanning electron microscopy (FE-SEM, S-4800, Hitachi), X-ray diffractometer (XRD) was used to collect powder diffraction data in the range of $10\text{--}70^\circ$ with $\text{Cu K}\alpha$ radiation ($\lambda=1.5418 \text{ \AA}$). XPS was obtained by model PHI 5000 Versa Probe. Then, the relative complex permittivity and permeability in the frequency range of $2\text{--}18 \text{ GHz}$ were measured by the Agilent PNA N5224A vector network analyzer with coaxial method and filler loading ratio of 25 wt%.

3. Results and Discussion

The sample phase composition and crystallinity of $\text{Co}_3\text{O}_4/\text{CC}$ composites can be observed by XRD diffraction analysis. In order to detect the successful synthesis of $\text{Co}_3\text{O}_4/\text{CC}$, the XRD patterns of the composites are analyzed in Figure 2. The diffraction peaks of Co_3O_4 were observed at 31.27° , 36.85° and 44.81° for S1-S3, which are corresponding to (220), (311) and (400) lattice plane. In addition, obvious diffraction peaks of C were observed at 22.76° (120), 26.57° (103) and 29.31° (113), which proved the existence of Co_3O_4 and carbon [28]. At the same time, no other impurity peaks are observed, indicating that the sample has a high purity and crystallinity [29]. It is obvious that $\text{Co}_3\text{O}_4/\text{CC}$ has been successfully synthesized.

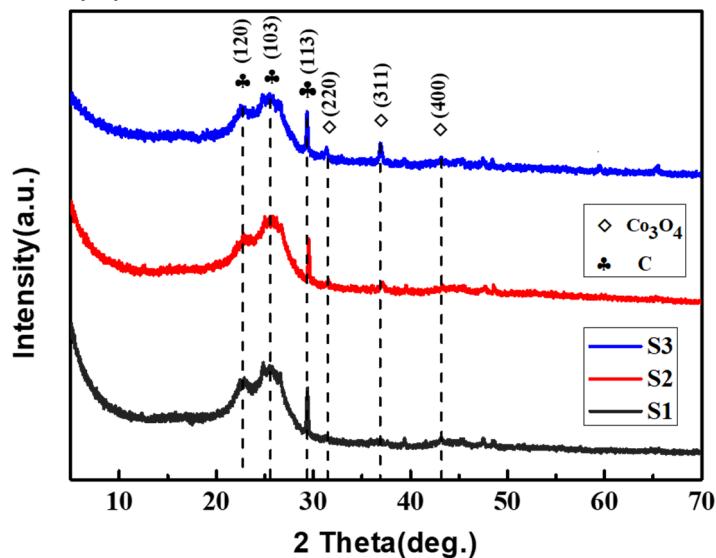


Figure 2. The XRD patterns of S1-S3.

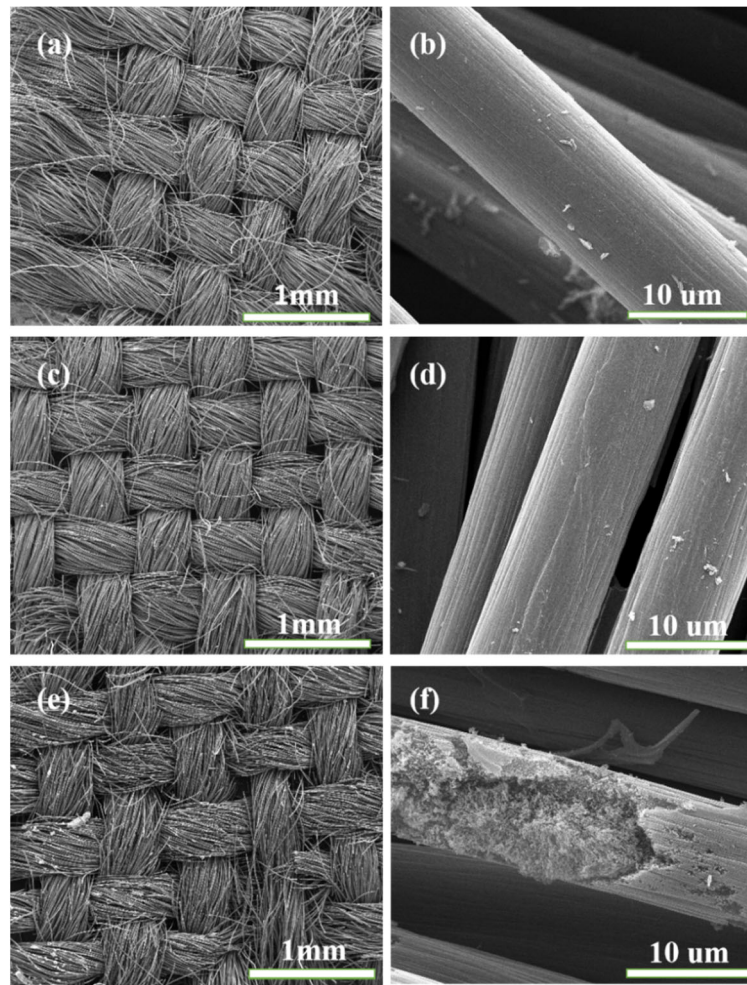


Figure 3. SEM images of (a-b) S1, (c-d) S2 and (e-f) S3.

The morphology of $\text{Co}_3\text{O}_4/\text{CC}$ composites with different temperatures were characterized by SEM pictures, which were shown in Figure 3. It can be seen from Figure 3 that the morphology of the samples with three different temperatures is very similar, the cross arrangement of the cylindrical morphology is well maintained and the structure size is quite uniform. However, with the temperature increasing, the morphology of $\text{Co}_3\text{O}_4/\text{CC}$ become roughly. In particularly, it can be seen from Figure 3f that most of the cylinder has been broken and presents a loose and porous structure, which may be caused by the collapse of the Co-MOFs. In addition, the surface of CC is relatively smooth and the floccule appears in the broken CC, which demonstrates the Co_3O_4 is grown inside of the CC. The floccule could provide loose and porous structure, which can offer more contact sites and increase the attenuation of electromagnetic waves.

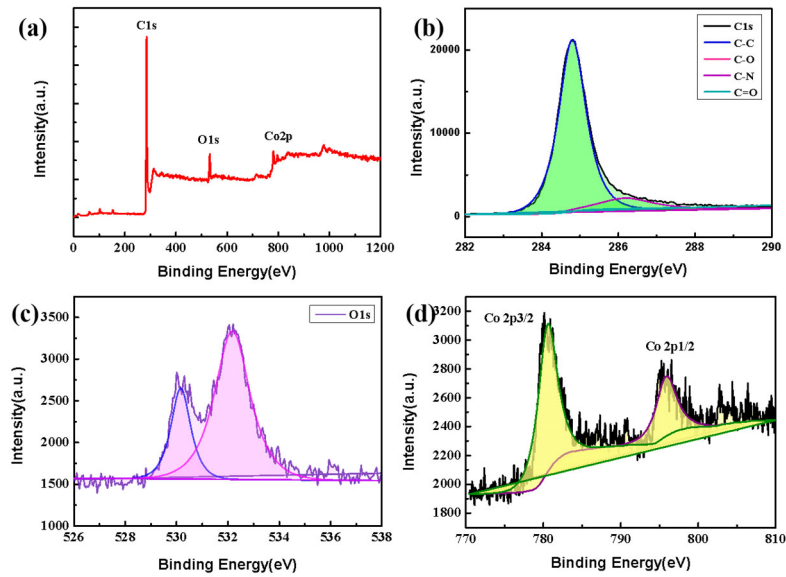


Figure 4. (a) XPS survey spectrum of S3; (b) C 1s, (c) O 1s and (d) Co 2p of core level spectrum in S3.

The valence states of elements in the composites can be clearly indicated by XPS [30]. The XPS results of Co₃O₄/CC are shown in Figure 4. Figure 4a shows the wide spectrum of Co₃O₄/CC composite material, and the Figure 4b-d shows the spectrum of C 1s, O 1s and Co 2p of S3 composite respectively. As can be seen from Figure 4b, two characteristic peaks appear in the C-1s spectrum of Co₃O₄/CC, which was peak-divided with the C-C/C=C peak appearing at 284.6 eV and the C-O peak emerging at 286.7 eV. It is obvious that the C-C/C=C bond signal is the strongest, which indicates that most of the precursor has become amorphous carbon after calcination and carbonization [31]. The results of XPS analysis in Figure 4c show that two characteristic peaks also appear in the O 1s spectrum of Co₃O₄/CC, in which the peaks are loading at 531.6 eV and 532.5 eV. In Figure 4d, the spectral peaks of Co 2P at 781.0 eV and 797.1 eV belong to Co 2P 3/2 and Co 2P 1/2, respectively [32]. The characteristic peaks can be attributed to cobalt nitrate hexahydrate and also indicates the presence of C and O elements.

Different calcination temperatures of precursor Co/CC could change the internal structure of Co₃O₄/CC composites, which affect the dielectric constant and magnetic permeability of the composite and adjust its microwave absorption property. In this study, we only pay attention to the dielectric characteristic analysis on account of the weak magnetism of the generated Co₃O₄/CC complex. The real parts (ϵ') and imaginary parts (ϵ'') of the complex dielectric constant are on behalf of the dielectric storage and dielectric loss capacity of the material, respectively [33]. It can be revealed from Figure 5 that the ϵ' of the Co₃O₄/CC complex generally decreases with the increase of frequency. It's worth noting that the ϵ' increases several times after the high frequency part of 9 GHz, which is mainly bring from exchange resonance [34]. According to Debye theory, the real and imaginary parts of the complex dielectric constant can be expressed in the following form [35]:

$$\epsilon' = \epsilon_{\infty} + (\epsilon_s - \epsilon_{\infty}) / (1 + \omega^2 \tau^2) \quad (1)$$

$$\epsilon'' = (\epsilon_s - \epsilon_{\infty}) \omega \tau / (1 + \omega^2 \tau^2) + \sigma_{ac} / \omega \epsilon_0 \quad (2)$$

where ϵ_s represent the static permittivity, ϵ_{∞} is the infinite static permittivity, ω belongs the angular frequency, τ is the polarization relaxation time, σ_{ac} is on behalf of the electrical conductivity and ϵ_0 represents the vacuum permittivity. According to equation (1), the decrease of the real part of the dielectric is independent of the increase of the angular frequency ω . The essence of this phenomenon is made from the existence of polarization relaxation at low frequency. Generally, it is important to note that the general relationship between the real part and imaginary part of the dielectric constant of the Co₃O₄/CC composite with three temperatures is S1 > S2 > S3. The reason why the complex

dielectric constant decreases gradually is that with the increase of calcination temperature, more and more Co_3O_4 is generated, which make the electrical conductivity gradually weakened of the $\text{Co}_3\text{O}_4/\text{CC}$.

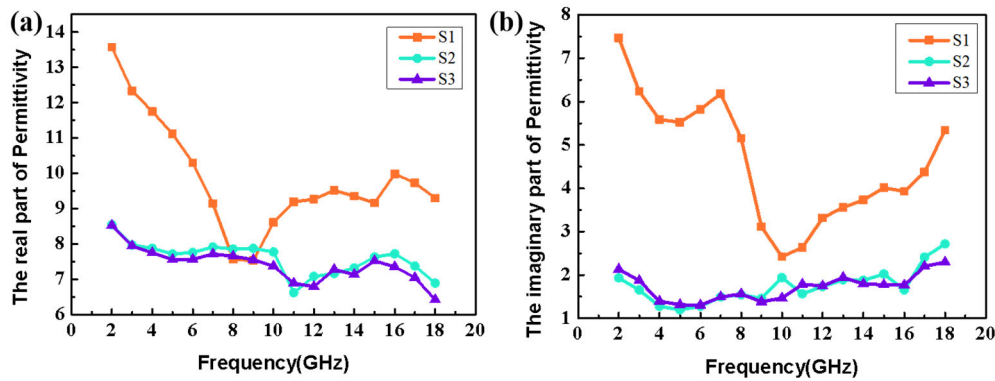


Figure 5. The real (a) and imaginary (b) parts permittivity of the $\text{Co}_3\text{O}_4/\text{CC}$ complex with three temperatures.

After analyzing the complex permittivity of $\text{Co}_3\text{O}_4/\text{CC}$ composite, the reflection loss of the generated composite is analyzed. Figure 6 shows the three-dimensional image of RL with different calcination temperatures and matching thickness, which can directly observe the RL value of $\text{Co}_3\text{O}_4/\text{CC}$ composite. The calculation formulas for RL of electromagnetic wave are as follows:

$$Z_{in} = Z_0(\mu_r/\epsilon_r)^{1/2} \tan h[j(2\pi f d/c) (\epsilon_r\mu_r)^{1/2}] \quad (3)$$

$$R_L = 20 \log (Z_{in} - Z_0)/(Z_{in} + Z_0) \quad (4)$$

where, Z_0 represents the impedance value of the free space. Figure 6a shows the three-dimensional (3D) RL of S1, and it can be seen that the minimum RL is -44.68 dB at 11.72 GHz with the thickness of 2.10 mm. Although the matching thickness of S1 is relatively thin, the frequency corresponding to the minimum reflection loss is still at the high frequency range (Ku band). The minimum reflection loss of S2 in Figure 6b is -33.95 dB, and the corresponding frequency and thickness are 5.4 GHz and 4.90 mm. The RL value of S3 is shown in Figure 6c, and the minimum RL can reach -46.59 dB at 6.24 GHz with a 4.2 mm thickness. It can be concluded that the frequency of the minimum RL gradually moves to low frequency with the increase of calcination temperature. According to Figure 6a, RL value is less than -10 dB in the frequency range of 10.76 to 14.16 GHz for S1, thus, the effective absorption band width of S1 is 3.4 GHz. Similarly, S2 (Figure 6b) has an effective absorption band width of 1.88 GHz in the frequency range of 4.52 ~ 6.4 GHz, while S3 (Figure 6d) has an effective absorption band width of 2.6 GHz with a frequency range of 5.12 ~ 7.72 GHz. It is well known that a good electromagnetic wave absorber should have large attenuation ability, superior impedance matching performance, minimum value of RL and a wide effective absorption band width and so on [36-37]. According to the above analysis, it's obvious that in low frequency area (< 6 GHz) the S3 have better reflection loss and thinner matching thickness. Therefore, the $\text{Co}_3\text{O}_4/\text{CC}$ complexes with calcination temperature of 500 °C is suitable for the microwave absorbing materials in low frequency.

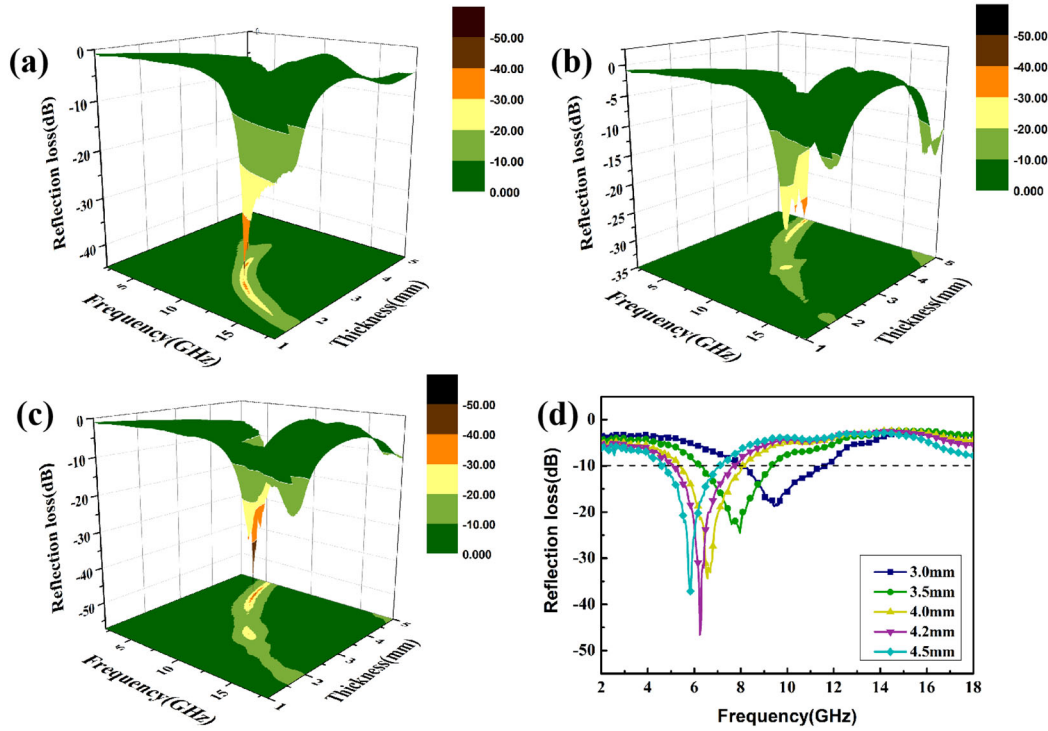


Figure 6. 3D plots of RL values for $\text{Co}_3\text{O}_4/\text{CC}$ with different calcination temperatures of (a) S1, (b) S2, (c) S3 in the frequency range of 2-18 GHz; (d) the RL values of S3 with different thicknesses.

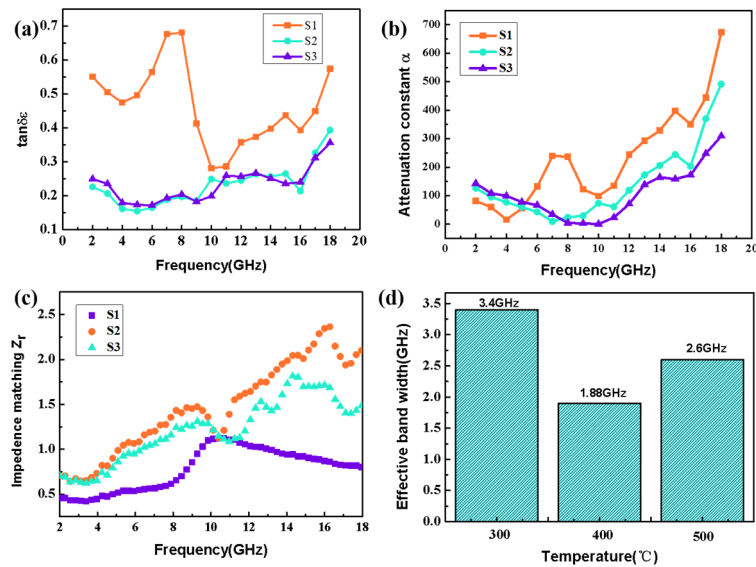


Figure 7. Frequency dependence of (a) $\tan \delta_\epsilon$, (b) α and (c) Z_r of S1-S3; (d) effective absorption bandwidth with three calcination temperatures.

In order to further explore the mechanism why the $\text{Co}_3\text{O}_4/\text{CC}$ composites have different absorbing properties with different calcination temperatures, the dielectric loss, attenuation ability of electromagnetic wave and impedance matching of the composites were further analyzed in this study. The dielectric loss is usually presented by the loss tangent of permittivity, the higher loss tangent indicating better dielectric loss. The loss tangent of permittivity can be expressed in the following formula [38]:

$$\tan \delta_\epsilon = \epsilon'' / \epsilon' \quad (5)$$

Figure 7a presents the variation of dielectric loss of Co₃O₄/CC composites with different calcination temperatures. In general, $\tan \delta_\epsilon$ with three temperatures demonstrate an upward trend in terms of the increase of frequency. Besides, a few relaxation peaks appear in low frequency, which proves that there are polarization relaxations in low frequency indicating better microwave absorption. For S1-S3 samples, the S1 composite has higher dielectric loss.

In addition to the dielectric loss, the attenuation factor α [39] and impedance matching Z_r [40] also play vital roles in the performance of electromagnetic wave absorption. Attenuation α refers to the amplitude or power attenuation in the electromagnetic wave transmission process, which is related to the dielectric loss. As can be seen from Figure 7b, the magnitude relationship of the attenuation ability of the three samples is $\alpha_{S1} > \alpha_{S2} > \alpha_{S3}$, indicating the S1 sample has better attenuation ability for electromagnetic waves. This is because with the enhance of calcination temperature, the amount of Co₃O₄ generated gradually added, leading to the decrease of permittivity. This phenomenon will bring about the decrease of dielectric loss and the attenuation loss, which illustrated the decrease of attenuation α is connected with the increase of calcination temperatures. For electromagnetic absorbent, an outstanding absorbing material should consider not only the strong electromagnetic propagation loss, but also the impedance matching. The attenuation constant α and impedance matching Z_r can be described in the following formulas:

$$\alpha = \frac{\sqrt{2}\pi f}{c} \sqrt{(\mu''\epsilon'' - \mu'\epsilon') + \sqrt{(\mu''\epsilon'' - \mu'\epsilon')^2 + (\mu'\epsilon'' + \mu''\epsilon')^2}} \quad (6)$$

$$Z_r = Z_{in}/Z_0 \quad (7)$$

where Z_r represents the impedance matching value, Z_0 is the free-space impedance value, Z_{in} is on behalf of the incident impedance matching value, c represents the speed of light. When $Z_r = 1$, the electromagnetic wave can realize zero reflection on the absorber surface, that is, the absorption effect reaches the best and the condition of $\epsilon_r = \mu_r$ should be satisfied. According to Figure 7c, the frequency corresponding to the minimum reflection loss of S1 is 11.72 GHz, and the corresponding impedance match at this frequency is 1.06. Similarly, the impedance matching corresponding values of S2 and S3 are 0.53 and 1.00 with the minimum reflection loss, respectively. In addition, the impedance matching values of S3 are closer to 1 at 6-8 GHz. Therefore, it illustrated that S3 has achieved almost perfect impedance matching at low frequencies. On the basis of this result, the S3 has better electromagnetic wave absorption performance and attenuation ability at low frequencies. Besides, as shown in Figure 7d, the S1 has more broadband effectively absorption bandwidth of the composites with three different calcination temperatures.

Therefore, the S3 has excellent microwave absorption performance in low frequency, the minimum reflection loss and effective band absorption width of S3 are further analyzed in this paper. In Figure 8a and 8b are minimum RL of S3 with different of matching thicknesses. When the thickness is 4.2 mm, the minimum reflection loss observed at 6.24 GHz is -46.59 dB, indicating elegant electromagnetic wave absorption characteristics. When the thickness of the S3 is reduced to 3.7 mm, the maximum effective absorption bandwidth is 3.04 GHz from 5.84 GHz to 8.88 GHz, at the moment, the minimum reflection loss is -32.13 dB. Compared with the previous work that has been reported in Table 1, the Co₃O₄/CC composites have thinner matching thickness and stronger reflection loss in low frequency.

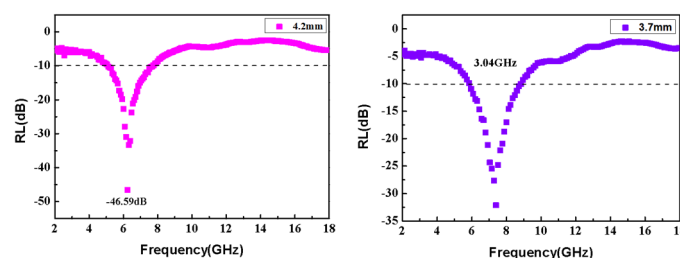
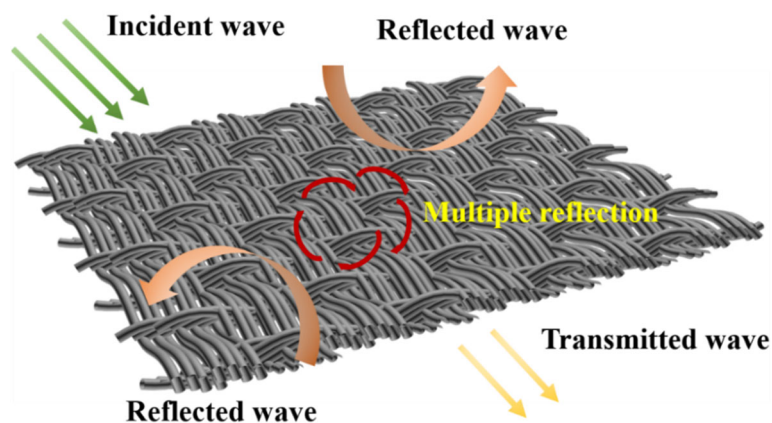


Figure 8. RL curve of S3 at thicknesses of (a) 4.2 mm and (b) 3.7 mm.

Table 1. Microwave absorption performance of the similar materials.

<i>Filler</i>	<i>RL (dB)</i>	<i>Thickness(mm)</i>	<i>Effective bandwidth (GHz)</i>	<i>Ref.</i>
(Zn _{0.8} Mn _{0.2}) ₂ Y and Co ₂ Z department hexagonal ferrite	-15	>5	3.2	40
NiZn Spinel type ferrite	-10	12	0.7	41
Magnesium doped barium ferrite	-10	6.0	3	13
Mg doped lithium zinc ferrite	-39	5.5	6.5	13
Co ₃ O ₄ /CC	-46.59	4.2	3.04	This work

Based on the above, the Co₃O₄/CC composites have better microwave absorption property with thinner thickness and widely effective absorption bandwidth in low frequency. The possible mechanisms of the microwave absorption are as in Figure 9: firstly, the carbon cloth and Co₃O₄ is a whole, which cause loss for incident electromagnetic wave; secondly, the carbon cloth could provide the network structure that allow more incident wave enter into the internal; finally, when the incident wave pass into the network structure the Co₃O₄ and CC could consume it, and generate dielectric loss. Therefore, the CC and Co₃O₄ synergistically improve the microwave absorption.

**Figure 9.** Diagram of microwave attenuation mechanism for Co₃O₄/CC composites.

4. Conclusions

In summary, the precursor Co-MOFs/CC was synthesized by the simple hydrothermal method, and the Co₃O₄/CC derivatives were generated through different calcination temperatures. It was concluded that with the increase of calcination temperature, the electromagnetic wave absorption performance was improved frequency gradually in the low frequency. When the calcination temperature was 500 °C, the minimum reflection loss RL can reach -46.59 dB at 6.24 GHz with 4.2 mm thickness. When the thickness is 3.7 mm, the effective absorption band width was 3.04 GHz in C band, and the minimum reflection loss was -32.13 dB. The synergistical effect of CC and Co₃O₄ made better microwave absorption performance with thinner thickness and widely effective bandwidth in the low frequency. This provides useful inspiration for the growth of absorbing materials on carbon cloth in low frequency, which is worthy of further investigation.

Author Contributions: Conceptualization, B.A. and X.L.; Data curation, B.A.; Formal analysis, B.A. and X.Y.; Investigation, B.A., M.W. and X.L.; Supervision, X.L. and C.F.; Validation, X.L. and C.F.; Visualization, B.A.; Writing-original draft, B.A.; Writing-review and editing, X.L., Z.M. and C.F. All authors approved the final version of the manuscript.

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Data Availability Statement: The data that support the findings of this study are available from the corresponding authors, upon reasonable request.

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Conflicts of Interest: The authors declare no conflict of interest.

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