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Recent progress of MOF-derived porous carbon materials for microwave absorption

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Microwave absorbing materials (MAM) have attracted considerable attention over the years in stealth and information technologies. Metal–organic framework (MOF) with a unique microstructure and electronic state has become an attractive focus as self-sacrificing precursors of microwave absorbers. The MOF-derived porous carbon (PC) materials exhibit a high absorbing performance due to the stable three-dimensional structure and homogeneous distribution of metal particles. MOF-derived PC materials are promising for ideal MAM *via* tuning of the structure and composition, resulting in appropriate impedance matching and the synergistic effect between magnetic and dielectric loss. In this review, the MOF-derived PC materials and their basic absorption mechanisms (dielectric loss, magnetic loss and impedance matching) are introduced, as well as the characters of various MOF-derived PC materials. In addition, this review provides a comprehensive introduction and tabulates the recent progress based on the classification of the MOF-derived metallic state, such as pure PC (without reduced metals), mono-metal/PC, multi-metal/PC, metal oxides/PC and other derived PC composites. Finally, the challenges faced by MOF-derived PC materials are overviewed, and their further development is mentioned.

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1. Introduction

In recent decades, the world has witnessed an eruptible development in stealth, information technology and electronics industry that bring public convenience and high-efficiency.¹ However, it is worth noting that the living environment has deteriorated dramatically with the explosive growth of modern electronic technology, which not only induces electrical interference with precision equipment and apparatus but also makes an impact on public health.² Thus, developing ideal microwave absorbing materials (MAM) with higher attenuating capacity, wider absorption bandwidth and lighter weight is an urgent proposition.³

Metal–organic framework (MOF) materials attract a lot of attention owing to their intriguing structure with a large pore volume, high surface area and uniformity of metals. MOFs have exhibited significant applications in gas storage and separation, sensing, catalysis, proton conduction, drug delivery and other fields.^{4–10} In addition, the synthesis process of MOF materials is simple and environment friendly, which meets the development requirements of green functional materials in modern society.¹¹

Up to now, more than twenty thousand monomers of MOF are prepared with different metal ions and organic ligands, exhibiting rich and varied micro-morphology such as sphere-like, flake-like, cube-like, octahedron-like, dodecahedron-like and so on,^{12–20} which have received attention from various fields. And the number of MOF materials reported is refreshed constantly with endless emergence of new monomers.

The metal ions cover almost all kinds of metals like main group metals, transition metals, lanthanide metals and rare-earth metals. As for the organic ligands, the common ligands are polyamines (especially amines produced from imidazole, oxalic acid and benzene), carboxylates, pyridyl, porphyrins, cyano groups, crown ethers and phosphonates.²¹ Therefore, there are various and complicated classification methods of MOF materials depending on what public focuses on. Different types of MOF can be converted into each other simply by changing the microstructure or one of the elements due to the highly adjustable character.

MOF possesses the advantages of high porosity, low density, large specific surface area, regular channel, adjustable pore structure and topological structure diversity.¹⁹ MOF-derived materials, especially the carbon materials, have become a new member of the microwave absorption field. The publications of MOF (almost all of publications searched are about MOF-derived porous carbon (PC) materials) as microwave absorbers are increasing progressively in recent ten years, as shown in Fig. 1. Compared with the MOF materials, MOF-derived PC materials are more promising as microwave absorbers.

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Fig. 2 Schematic diagram of microwave absorption mechanism of MOF-derived metal/C composites, this figure has been adapted from ref. 41, 70 and 71 with permission from Applied Surface Science, Copyright 2020, Composites Communications, Copyright 2020 and Applied Surface Science, Copyright 2020, respectively.

produced *via* pyrolysis in inert atmosphere. And the absorbers effectively combined with metallic components exhibit improving Debye dipole polarization and multiple interface reflection.⁶² Diversiform metals and metallic compounds can be combined *via* simple doping method. As a simple example, Fe-Co alloy is an ideal absorber for constructing a composite of magnetic material and carbon material with high saturation magnetization, Curie temperature and conductivity, which are better than those of PC doped with Co only.¹⁴ In addition, FeCo or FeNi alloys generally exhibit greater permeability, Snoek's limit and higher saturation magnetization than ferrites, which avail the modification and tuning of absorbing properties.⁶³

3.3 Synergistic effect of magnetic/dielectric loss

The strong microwave energy loss is common achieved through a multitude of loss mechanism.⁶⁴ Microwave absorbers composed of dielectric and magnetic loss materials are promising with optimized impedance matching and synergistic effect between dielectric loss and magnetic loss. And the impedance matching characteristic is typically represented by reflectivity R and term Z_{in} , as shown in eqn (3)–(5).⁶⁵ Nearly all of microwaves can enter into absorbing materials when Z_{in} is close to 1 (nearly zero reflection at interface). The single-component carbon materials are not conducive to impedance matching because of the large dielectric constant, low magnetic permeability and narrow EAB. And single metal materials with over-high magnetic loss are difficult to meet an ideal microwave absorber. Some of incident waves are reflected with only high eddy current loss, which is not conducive to the absorbing property.⁶⁶ MOF-derived PC materials are coupled with carbon

skeleton and metal particles, and carbon skeleton not only improves the corrosion resistance to some degree, but also effectively isolates metal particles, reducing eddy current losses to maintain excellent absorbing performance.⁶²

$$Z_{in} = \left(\sqrt{\frac{\mu_r}{\epsilon_r}} \right) \tanh \left[j \left(\frac{2\pi f d}{c} \right) \left(\sqrt{\mu_r \epsilon_r} \right) \right] \quad (3)$$

$$Z_0 = \sqrt{\frac{\mu_0}{\epsilon_0}} \quad (4)$$

$$R = \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \quad (5)$$

The impedance matching and the absorbing characteristic should be considered comprehensively for an ideal absorber, and the absorbing property is generally indicated by the RL, which is expressed as eqn (6).

$$RL = 20 \lg \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (6)$$

For MOF-derived PC materials, the unique embedded structure of the metals and carbon exhibits abundant interfaces with a large amount of free charge accumulating on them, resulting in the interface polarization. Besides, multiple scattering reflection and interfacial polarization are increased in the porous structure of MOF-derived PC materials, enhancing the absorption performance.³⁸ For instance, Li *et al.* obtained magnetic Co-C@C composites from ZIF materials. The composites showed the smaller RL of -58.0 dB and wider



bandwidth of 5.7 GHz compared with previously reported composites, due to the favorable impedance matching and strong attenuation.⁶⁷ The size of metal nanoparticles and pores is adjusted with the different pyrolysis temperature, which can tune the impedance matching.⁶⁸ The introduction of pores could decrease the density of the absorbers and have an effect on the permittivity of carbon materials. Ding *et al.* studied MIL-53(Fe) derived MCC/rGO composites by gradient temperature calcination and discussed the effect of temperature for absorption performance.⁶⁹ The framework enhanced the internal scattering, and the graphene provided a larger absorption surface, efficiently enhancing the absorbing performance.

In a word, compared with common materials, the MOF-derived PC materials with nanoscale quantum dots exhibit better absorbing performance. The synergistic effect of absorbers can achieve excellent impedance matching and high absorbing capacity,⁵³ as shown in Fig. 2.

4. MOF-derived PC materials for microwave absorption

The MOF-derived PC composites exhibit huge potential application as high-performance microwave absorption materials. And the derived PC mentioned herein from recent literatures are divided into a series of subsets for categorization. Based on the metal state, MOF-derived PC composites can be classified as pure PC, mono-metal/PC, multi-metal/PC, metal oxides/PC and

other MOF-derived PC composites. The derived pure PC also be employed not only as absorbers but also as reactive materials.

4.1 Pure PC

As mentioned above, PC exhibits potential application for microwave absorption. MOF-derived pure PC is a high porosity carbon material without metals, which is generally obtained *via* high temperature pyrolyzation process to remove low-boiling point metals. In addition, it can also be prepared through acid etching to remove reduced active metals. MOF-derived pure PC absorbers are often composited with other loss materials because of their restriction as wide frequency bandwidth microwave absorbers.

Various composite forms of derived pure PC are reported. Some are applied as central core of core-shell or yolk-shell structure with other cooperate materials assembling upon their surface. Besides, some are grown on the matrix to construct the porous and stable carbon shell. Derived pure PC can also be combined with two-dimensional materials to play a disperse and dopant role. Zhao and co-workers synthesized nanopolyhedron PC decorating on the surface of reduced graphene oxide (rGO) by *in situ* pyrolysis.⁷² ZIF (zeolitic imidazolate framework) is a typical precursor to obtain pure PC materials, such as ZIF-8 (Zn-MOF). It is demonstrated that the grown of ZIF-8 particles is restrained by GO and the derived PC reduces the re-stacking of rGO nanosheets, which yield optimized performance with minimum RL value of -66.2 dB at the thickness of 2.89 mm at 6.2 GHz (as shown in Fig. 3). Because of the low boiling point of metal Zn, ZIF-8 has played a significant

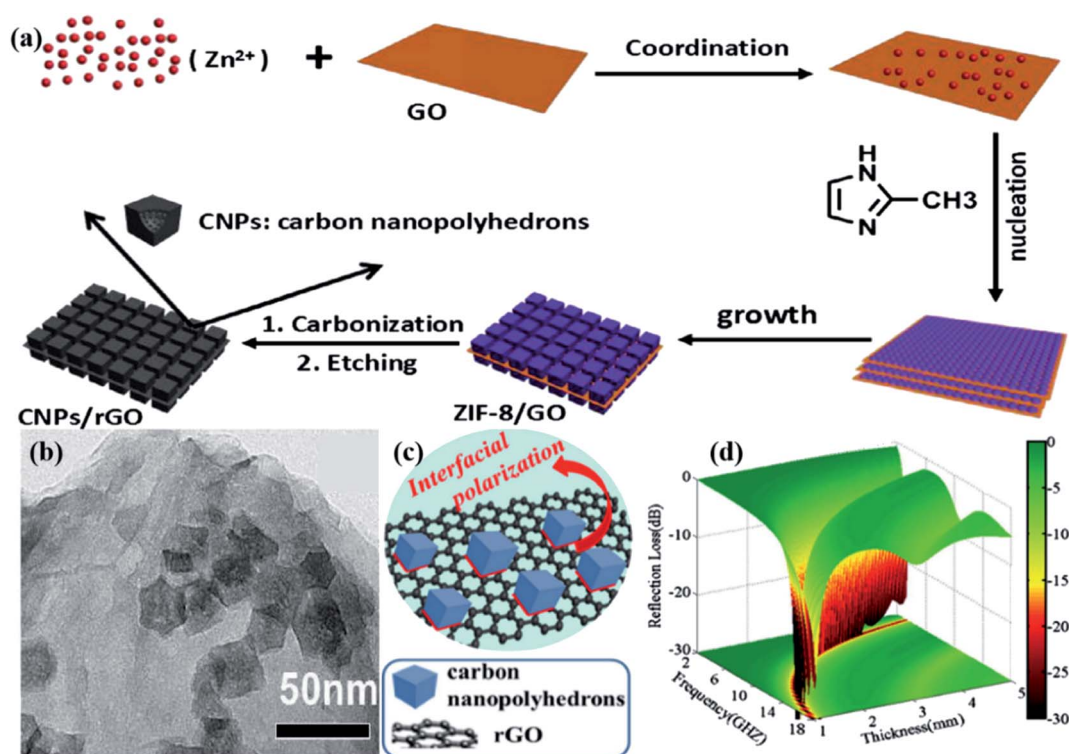


Fig. 3 The fabrication of CNPs/GO derived from ZIF-8/GO (a), TEM images of CNPs/GO (b), interaction (c) and the RL of CNPs/GO (d). This figure has been reproduced from ref. 72 with permission from Journal of Colloid and Interface Science, Copyright 2018.



Table 1 The microwave absorbing performance of MOF-derived pure PC materials

Absorbers	Loading (wt%)	RL _{min}		EAB		Ref.
		Thickness (mm)	Value (dB)	Thickness (mm)	Value (GHz)	
CNPs/GO	40	2.89	-66.2	1.5	3.6	72
Co@NPC	25	1.45	-48	1.65	5.2	73
Hollow GO@PC	10	3.7	-32.43	3.5	4.2	74
N-doped PC	50	4.0	-39.7	4.0	4.3	75
MCCMs	30	2.0	-28.5	1.8	5.7	76
NPC@CPI-2	50	1.9	-46.86	1.65	3.74	77
C@Co/NC	25	2.2	-52.5	2.2	4.4	78
Fe/C	60	3.0	-40.0	1.5	6.0	79
Fe-N/C	33.3	1.7	-30.98	1.7	5.04	80

role in acting as admirable templates to obtain pure PC materials.^{34,73,74} Wu's group also used ZIF-8 as the derived loose PC precursor to prepare high-performance absorbers.⁷⁵ By the way, the MOF-derived composites are reported that they can be effectively dispersed in one-dimensional and two-dimensional materials such as carbon fiber and graphene, which play a role in avoiding agglomeration.

It is feasible to obtain pure PC *via* using other metal-based MOF-derived composites as precursors and adding acid solution to get rid of metals. For example, Du *et al.* used Ni-MOF as templates to synthesize the multi-chamber carbon microspheres (MCCMs) *via* the pyrolysis progress and etching treatment.⁷⁶ The unique multi-chamber structure possessed better impedance matching characteristic than hollow carbon microspheres. Peng *et al.* obtained PC through removing Cu from the prepared HKUST-1 (Cu-MOF) and studied the microwave attenuation of the derived PC composites.⁷⁷

Similar to above absorbers of MOF-derived PC, an increasing number of PC composites have been developed with other magnetic loss and dielectric loss materials to induce microwave absorption, through the increased research of MOF precursors. The minimum RL and EAB with their corresponding loading and thickness of these similar materials are tabulated in Table 1.

4.2 Mono-metal/PC

The derived-PC composites filled with metallic particles possess the increased magnetic interaction with incident microwave and the optimized impedance matching. As a result, the efforts of developing novel microwave absorbers have been devoted to study compounds of carbon materials and magnetic metal nanoparticles in recent years. When MOF-derived PC composites are combined with single component metal and carbon, the composites can be classified as MOF-derived mono-metal/PC materials. Typical magnetic loss absorbing metallic materials

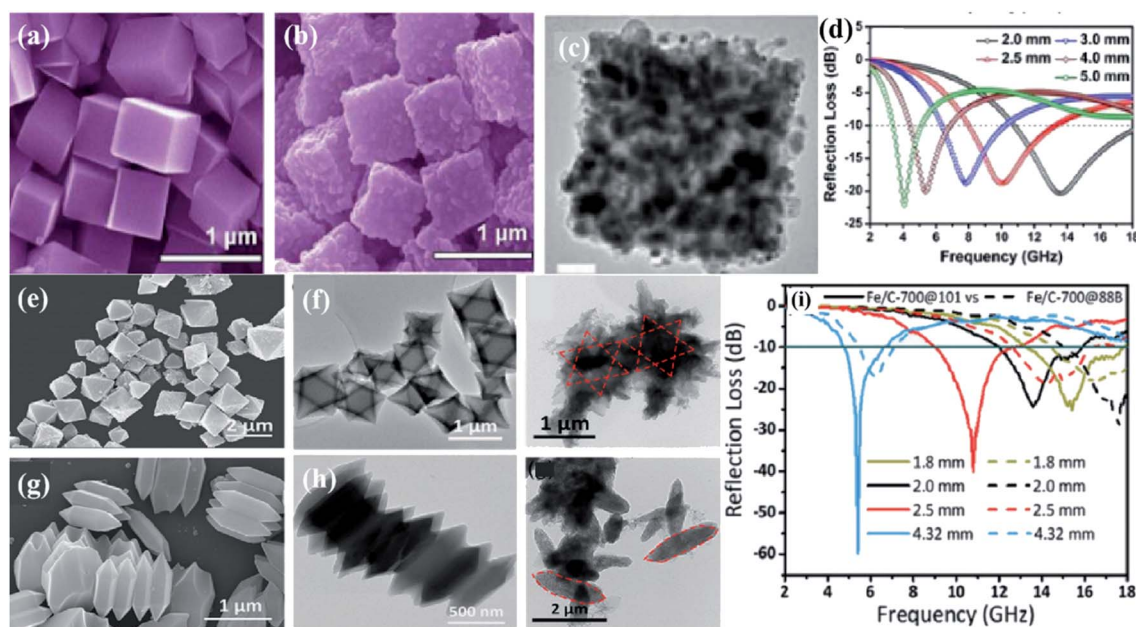


Fig. 4 SEM and TEM images of PB (a), Fe/C-PB (b and c), MIL-101-Fe (e), Fe/Fe₃C/C-800@101 (f), MIL-88B-Fe (g), and Fe/Fe₃C/C-800@88B (h), respectively. RL of Fe/C-PB (d), Fe/C-700@101 and Fe/C-700@88B (i), respectively. This figure has been reproduced from ref. 18 and 83 with permission from Advanced Materials Interfaces, Copyright 2020 and Journal of Materials Chemistry A, Copyright 2015.



includes Fe, Co and Ni, which are also mainly applied in the MOF-derived PC materials.^{49,81,82}

Prussian blue (PB) is one of common Fe-based MOF which consists of $\text{Fe}_4[\text{Fe}(\text{CN})_6]_3$ (a mixed-valence iron(III) hexacyanoferrate(II) compound) with a face-centered-cubic (fcc) crystal structure. And it is an eligible precursor candidate for derived Fe/C composites because of abundant contents of carbon and iron species.^{83–85} Xu *et al.* used PB as a MOF precursor to fabricate core-shell Fe@graphitic carbon nanoparticles under different pyrolysis temperature (600–700 °C), and the results displayed strong dielectric loss and magnetic loss properties (Fig. 4).⁸³ The PB derived Fe/C and its composites have been noticed as new microwave absorbers.⁸⁵ In addition, the more efforts are devoted on MIL (named based on the discovery place: Materials of Institut Lavoisier) series for Fe-based PC derivatives. Miao *et al.* reported the morphology effect on microwave absorbing by annealing the two MOF precursors (MIL-101-Fe and MIL-88B-Fe).¹⁸ Both of them had almost identical chemical composition and microstructure, including element content, valence state, pore size and volume, but with different topology. The Fe/C-700@101 and Fe/Fe₃C/C-800@101 (the pyrolysis temperature of MIL-101 precursor was 700 °C and 800 °C) were reduced at high temperature from ferric oxide by CO and reductive carbon generated, showing the excellent performance, as shown in Fig. 4. However, in comparison with pure iron, ferric oxides exhibit chemical stability and have wider application for microwave absorption, which are tabulated in the fourth part of this chapter.

The Ni/C composites with the shape of porous spheres are formed by H₃BTC (BTC = benzene-1,3,5-tricarboxylate) and Ni²⁺. The amorphous spheres formed with Ni²⁺ and H₃BTC ligands dissolved and diffused to the surface in the process of

crystallization, resulting in rough surface of Ni-based MOF precursors.^{15,22,86–88} Liu *et al.* successfully prepared waxberry-like Ni@C core-shell microspheres with hierarchical architecture.¹⁵ The composites obtained after pyrolysis at 700 °C showed optimum absorbing performance, including the RL value of -73.2 dB and the EAB of 4.8 GHz with only 1.8 mm applied thickness (Fig. 5). Similarly, the hollow Ni-MOF spheres were synthesized with different surface morphology by tuning the time of hydrothermal reaction, as shown in Fig. 5.⁸⁸ And it was found that the samples with 8 h reaction time could reach a broad EAB of about 6.8 GHz with only 1.8 mm thickness. Moreover, rod-like,⁸⁶ flower-like,⁸⁹ accordion-like⁹⁰ and other sphere-like Ni/C composites⁸⁷ derived from Ni-based MOF also exhibited strong microwave absorption and attenuation performance, and these results are listed in Table 2.

ZIF materials are designed through the expansion of the zeolite topology with larger pores and higher porosity.⁹¹ ZIF-67 and ZIF-8, with the same microporous structure and organic ligands, are formed by bridging the 2-methylimidazolate anions and metal cations (Co²⁺ and Zn²⁺), respectively. Due to high porosity, easy preparation and large apertures, ZIF-67 is widely applied as precursor of Co-based highly PC matrix absorbers in the recent years. Lu *et al.* fabricated porous Co/C composites by the directly decomposing ZIF-67 at different temperatures (500 °C, 600 °C and 800 °C).¹⁷ Co/C composites obtained at 500 °C exhibited best absorbing performance with the minimum RL of -35.3 dB, EAB of 5.80 GHz and the absorbing thickness of 2.5 mm. Chen *et al.* designed and synthesized rod-like Co/C composites derived from cubic Co-based MOF *via* hydrothermal and calcination processes.⁹² The prepared composites exhibited optimized absorbing ability with the minimum RL of -47.6 dB with 2.0 mm. In addition, Co-based

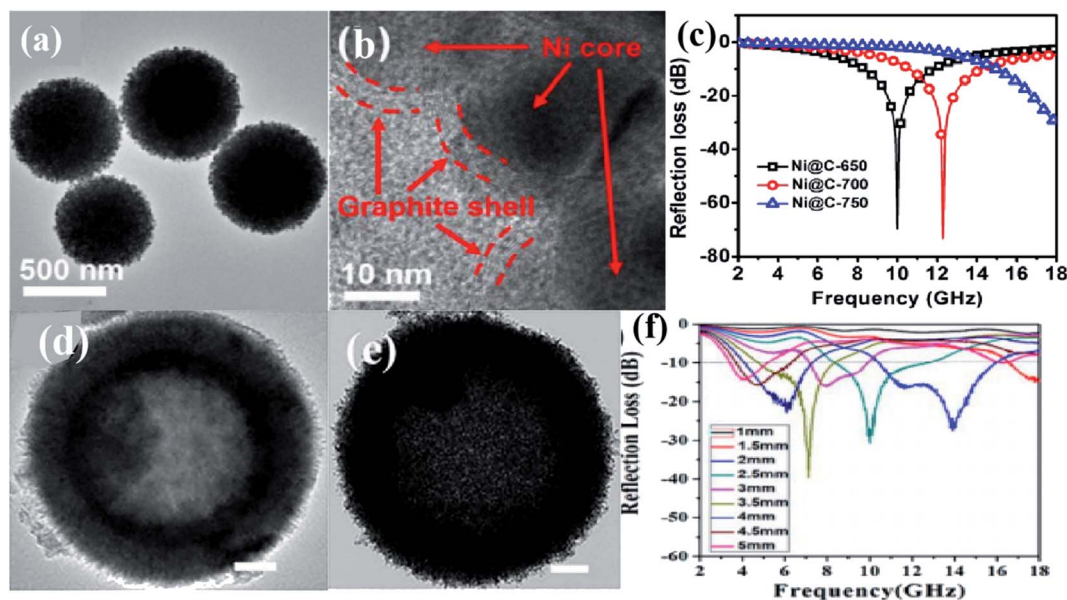


Fig. 5 SEM (a), TEM (b) images and RL (c) of waxberry-like hierarchical Ni/C (annealing at 650 °C), TEM images of Ni-MOF precursors with 8 h (e), Ni/C annealed at 600 °C (d) and RL of Ni/C-8 h (f). This figure has been reproduced from ref. 15 and 88 with permission from Journal of Materials Chemistry C, Copyright 2019 and ACS Applied Nano Materials, Copyright 2019.



Table 2 The microwave absorbing performance of mono-metal/PC materials

Absorbers	Loading (wt%)	RL _{min}		EAB		Ref.
		Thickness (mm)	Value (dB)	Thickness (mm)	Value (GHz)	
Fe/carbon	15	2.5	−29.5	2.5	4.3	48
Fe ₃ O ₄ @PB	60	1.55	−48.04	2.6	4.5	58
Fe/carbon	40	2.0	−20.3	2.0	7.2	83
Fe/C@Co ₃ O ₄	50	1.5	−38.1	2.5	7.65	98
Fe ₃ O ₄ -C/RGO	15	3.6	−60.5	1.5	5.5	98
Fe/NPC	20	1.8	~−28	1.8	5.6	99
Ni@C	33	3.5	−45.8	3.5	5.1	22
Ni@C	40	2.7	−86.8	2.7	7.4	22
Ni/C rod	40	2.6	−51.8	2.6	3.48	86
Ni@C@ZnO	25	2.5	−55.8	2.0	4.1	87
Ni/C-8 h	50	—	—	1.8	6.8	88
Ni@C	25	1.8	−86.9	1.8	6.6	90
Ni@C	30	1.85	−57	1.85	6.0	100
Ni/C nanosheets	10	2.2	−71.6	2.2	4.7	101
Co/C	40	4.0	−35.3	2.5	5.8	17
Co/C	40	2.8	−41	2.0	5.6	46
Rod-like Co/C	22	2.0	−47.6	2.0	5.11	92
Co/C	60	2.0	−26.4	2.0	6.6	93
Co@NCNT	25	1.8	−53	2.0	6.2	95
Co@NPC@TiO ₂	50	1.5	−31.7	1.0–5.0	13	96
Co/C	25	3.0	−30.31	3.0	4.93	102
Co/N-doped C	20	2.5	−65.1	3.2	9.4	103
Co/NC@MnO ₂	15	3.7	−58.9	3.7	5.56	11
Co/C@ZnO@GO	30	2.0	−45.4	2.0	5.4	97
Co@C@RGO	20	2.6	−67.5	2.0	5.4	104
Co@C@NRGO	20	2.0	−73.4	2.0	5.3	105
Co-C-MWCNTs	17.5	0.9	−25.27	1.6	2.38	106
Co/C@V ₂ O ₃	50	1.5	−40.1	1.5	4.64	107
CNT/Co/C	10	2.9	−53.3	1.6	8.02	108
Mo ₂ C/Co@C	35	1.7	−47.98	1.6	6.0	109

MOF can also be synthesized with different organic ligands (glucose, 2,5-dihydroxyterephthalic acid and so on) in a more convenient and environment-friendly way.^{93,94} Co@NC composites with multi-dimensional shapes (sheet-, flower-, cube-, dodecahedron- and octahedron-like) from ZIF-67 precursors were fabricated *via* regulating the anion/linker ratio and the solvent.⁹⁵ The observed results provide the way to fabricate high-absorbing MAM with controlled morphology. Other similar Co-based MOF derived PC composite materials are listed at Table 2.

Core-shell structure is a significant research direction for MOF-derived microwave absorbers. For example, Ji *et al.* developed novel multi-interfaced yolk-shell Co@NPC@TiO₂.⁹⁶ As a typical semiconductor, TiO₂ is widely applied as a supplemental material to optimize impedance matching owing to its strong dielectric loss. In addition, the porous core-shell Co/C@ZnO-decorated rGO was synthesized *via* a facile method, displaying the exceptional microwave absorbency (Fig. 6).⁹⁷

The MOF-derived PC materials are not only applied as the central core, but also the lightweight carbon shell of the composites. Recently, Liu *et al.* prepared two different types of ZIF to synthesize nano core-shell carbon cages, made up of N-doped carbon inner shell from ZIF-8 and Co/N-doped carbon outer shell from ZIF-67.⁷⁸ Derived core-shell composites

exhibited hierarchical porous, high specific surface area and thin thickness, which induced excellent microwave attenuating performance. The strong RL reached −52.5 dB with 25 wt% filler loading of composites, and the EAB of 4.4 GHz with the thickness of 2.2 mm. The results provided a new strategy of carbon nanocages fabrication with tunable chemical composition and the impedance matching. Besides the examples discussed above, other mono-metal/PC composites with excellent microwave absorbing performance are tabulated in following Table 2.

4.3 Multi-metal/PC

The MOF-derived multi-metal/PC composites (with two or more metals embedded in the derived carbon matrix) often show the better absorption performance than mono-metal/PC, due to the multi-metal/PC composites artfully integrating the advantages of multi metals and carbon. Effective doping of other metallic components to compose functional multi-metal MOF is served as a prospective approach. The compositions and the structures can be optimized to achieve efficient microwave attenuation like improving Debye dipole polarization and multiple interface loss.^{110,111} The optimizing absorption properties are due to the synergetic effects of the high porous structure and the multiple





Fig. 6 The scheme for the synthesis of Co/NPC@ZnO/rGO (a), SEM images of ZIF-67 (b) and Co/NPC@ZnO/rGO (d), and the RL of Co/NPC@ZnO/rGO (c). This figure has been adapted from ref. 97 with permission from Journal of alloys and compound, Copyright 2020.

components. This expansion method not only provides interesting references for further studying MOF derived carbon-based lightweight MAM, but also broadens the application of MOF materials.

Nowadays, FeCo particles and their composites derived from MOF have been widely reported as microwave absorbers. For example, Ji *et al.* developed carbon composite with FeCo bimetal nanoparticles embedded *via* pyrolysis process of ZIF-

67.¹⁴ The composites showed a strong RL of -21.7 dB with the thin thickness of only 1.2 mm and a broad EAB of 5.8 GHz (covering from 12.2 GHz to 18 GHz). The saturation magnetization gradually increased with the augment of Fe element content. Compared with pure Co/NPC derived from ZIF-67, Fe-Co/NPC exhibited effectively microwave absorption properties, as showed in Fig. 7.



Fig. 7 SEM image of Fe-Co/NPC (a), field-dependent magnetization curve of Fe-Co/NPC (b), the RL (c) and attenuation constant (d) of the Co/NPC, Fe-Co/NPC. This figure has been adapted from ref. 14 with permission from Nanoscale, Copyright 2015.



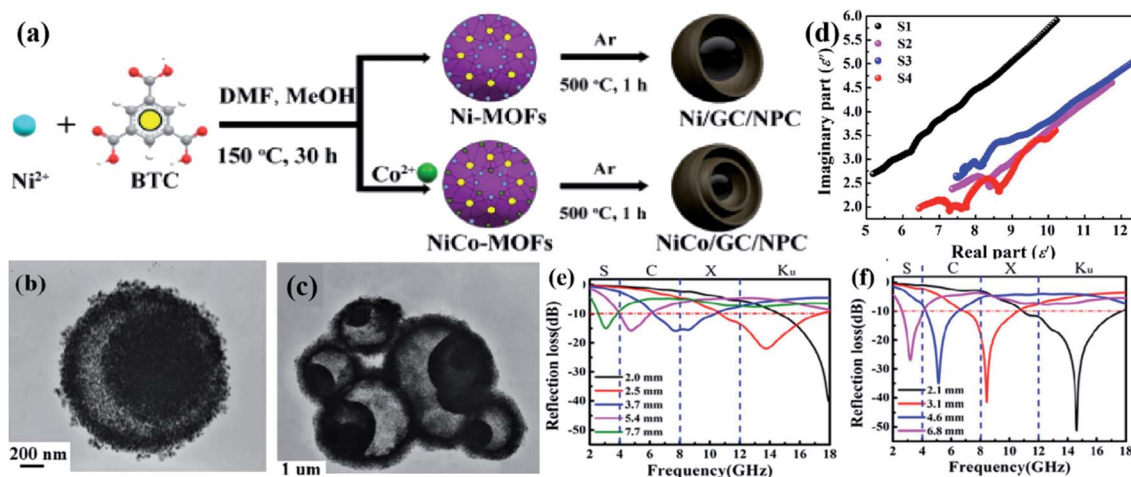


Fig. 8 The schematic formation process of the NiCo/GC/NPC composites (a), TEM images of S1 (b), S4 (c), the Cole–Cole semicircles of S1–S4 (d), the RL with various thicknesses for S1 (e), S4 (f) (S1 represents Ni/GC/NPC, and the content of Co increases from S1 to S4). This figure has been reproduced from ref. 71 with permission from Applied Surface Science, Copyright 2020.

Besides, hollow sphere-like FeCoNi@C absorbers were obtained *via* high-temperature carbonization, which used trimetallic FeCoNi-MOF as the precursor, and more excellent absorption properties are obtained compared with the carbonized mono-metal MOF or bi-metal MOF.¹¹² Doping with metallic elements can also be achieved through opposite direction, which illustrates the replaceability of similar valence transition metals. Dielectric loss material manganese dioxide (MnO₂) with flower nanostructures was assembled on the derived CoFe@C nanocube *via* a hydrothermal reaction, giving rise to the improvement of the impedance matching.¹¹³ The Co element was added into the PB nanocube and reduced to Co magnetic metallic nanoparticles at the high annealing temperature. The

CoFe@C@MnO₂ achieved a minimum RL of –64 dB and a maximum EAB of 9.2 GHz from 8.8 GHz to 18 GHz with a thickness of 1.3 mm and 1.6 mm, respectively. Yang *et al.* designed NiFe@C nanocubes derived from NiFe PB on the GO nanosheets, having a minimum RL of –51 dB at a thin thickness of 2.8 mm.¹¹⁴

Sometimes the doping elements can not only enrich the composition, but also result in a novel structure. Lu and his group designed the MOF derived nanocomposites NiCo/GC/NPC with hierarchical double yolk–shell structure, which were synthesized by regulating Co elements doped into Ni-based MOF.⁷¹ The emergence of yolk–shell could be ascribed to the heterogeneous decomposition induced by Co and Ni

Table 3 The microwave absorbing performance of multi-metal/PC materials

Absorbers	Loading (wt%)	RL _{min}		EAB		Ref.
		Thickness (mm)	Value (dB)	Thickness (mm)	Value (GHz)	
Fe–Co/NPC	50	1.2	–21.7	1.2	5.8	14
CoFe@C@MnO ₂	50	1.3	–64	1.6	9.2	113
CoFe@C	10	2.8	–61.8	2.8	9.2	117
Solid Fe/Co/C	33	2.0	–54.6	2.5	8.8	118
FeCoNi@C	38	2.1	–64.75	2.47	8.08	112
NiFe/C@GO	40	3.0	–65.5	3.0	4.5	114
CoZn/C/graphene	6	1.5	–47.31	2.2	4.01	2
CoZn/C	40	2.5	–45.2	2.5	5.7	41
CoZn/NPC	30	2.0	–49.0	2.0	5.3	119
CoNi/C-650	30	1.8	–74.7	1.8	15.1	24
Air@NC/Ni–Co	25	2.2	–36.5	2.3	6.55	70
NiCo/GC/NPC	30	2.1	–52.2	2.1	7.2	71
CoNi/C	10	2.0	–61.02	2.0	5.2	115
Ni _{0.8} Co _{0.2} @C	25	3.5	–39.3	2.0	4.8	116
G/CoNi@NCNTA	10	2.0	–44.23	1.6	4.63	120
CoNi@NG-NCPs	35	3.0	–45.73	2.5	4.32	121
MoW-NC	30	2.8	–55.6	2.8	8.8	122
CoMo@NC	30	2.5	–44.8	2.5	6.56	123



component, which not only decreases the density of composites, but also facilitates the multiple interfacial polarization (Fig. 8). Similarly, the additional metal Ni was introduced into the derived porous Co/C composites to obtain hollow structure, which yield the RL of -61.02 dB and the EAB of 5.2 GHz.¹¹⁵ Similarly, Che *et al.* successfully synthesized porous $\text{Ni}_{1-x}\text{Co}_x\text{@C}$ composites derived from MOF with different Ni, Co millimole content.¹¹⁶ The generated carbon/alloy particles acted as catalysis to encourage the carbon sp^2 arrangement through the thermal decomposition, forming special micro architecture. Other MOF-derived multi-metal/PC composites mentioned above are tabulated in Table 3.

4.4 Metal oxides/PC

If the obtained derived PC is formed by metal oxides and reduced carbon, it can be categorized as MOF-derived metal oxides/PC composites. The saturation magnetization and the compatible dielectric loss of metal oxides are lower than those of magnetic metal nanoparticles (*e.g.* Fe, Co, Ni and related alloys).⁸³ However, they have the great chemical stability, characteristic magnetic property and facile manipulation. Some metal oxides like Fe_3O_4 , have been often served as magnetic promoters for the enhancement of microwave absorbing, and

other non-magnetic metallic oxide (ZnO , Co_3O_4) applied to optimize impedance matching. In addition, it is reported that MOF-derived metal or metal oxide particles carbon composites are mainly based on the reduction potential of metal ions at the pyrolysis process. The metal oxides nanoparticles tend to form when the reduction potential of metal ions is lower than -0.27 V. On the contrary, the metal nanoparticles tend to form at a reduction potential higher than -0.27 V.¹²⁴

Fe_3O_4 is one of the widely accepted magnetic metal oxide microwave absorbers because of its low cost, chemical stability and high absorption performances. And it exhibits optimized impedance matching after being composed with other materials. Xiang *et al.* obtained magnetic $\text{Fe}_3\text{O}_4\text{@NPC}$ composites through two thermolysis processes with increasing saturation magnetization.⁵¹ The sample exhibited excellent absorbing properties, including a strong RL of -65.5 dB as well as a broad EAB of 4.5 GHz with a matching thickness of 3.0 mm. Similarly, Shu *et al.* compounded MOF-derived $\text{Fe}_3\text{O}_4\text{@C}$ and rGO by solvothermal and pyrolysis.⁹⁸ The obtained $\text{Fe}_3\text{O}_4\text{-C/RGO}$ displayed the minimum RL of -60.5 dB with a thickness of 3.6 mm and the EAB of 5.5 GHz with an ultrathin thickness of merely 1.5 mm. Moreover, derived metal oxides can translate into stronger dielectric loss absorbers as the raw materials. Currently, metal

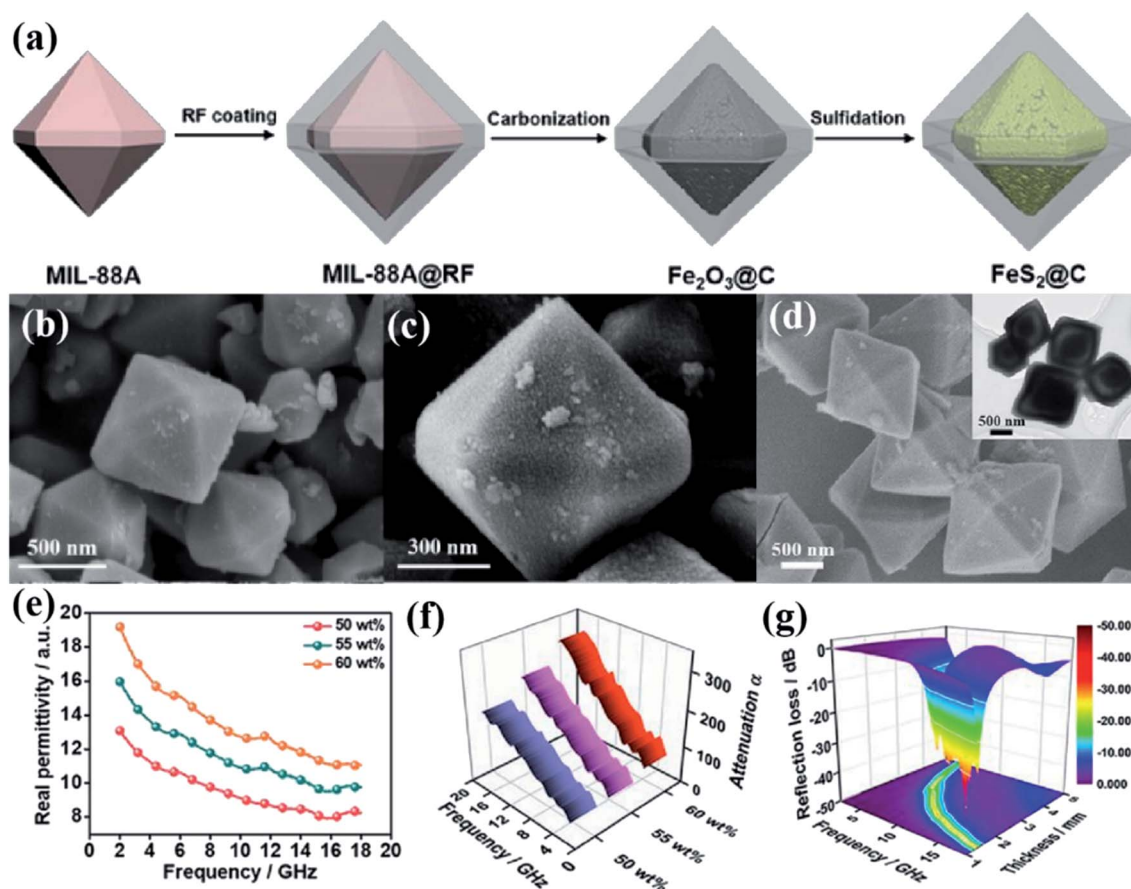


Fig. 9 The schematic formation process of $\text{FeS}_2\text{/C}$ composites (a), SEM images of Fe-MOF (b), $\text{Fe}_3\text{O}_4\text{/C}$ (c), SEM and TEM images of $\text{FeS}_2\text{/C}$ (d), the real permittivity (e), attenuation constant (f) and the RL of $\text{FeS}_2\text{/C}$ composites (g). This figure has been reproduced from ref. 16 with permission from Nano Letters, Copyright 2020.



Table 4 The microwave absorbing performance of metal oxides/PC materials

Absorbers	Loading (wt%)	RL _{min}		EAB		Ref.
		Thickness (mm)	Value (dB)	Thickness (mm)	Value (GHz)	
FeS ₂ @C	55	1.45	-45	1.2-5.0	15.4	16
Fe ₃ O ₄ @NPC	40	3.0	-65.5	3.0	9.8	51
Fe ₃ O ₄ -C/RGO	15	3.6	-60.5	1.5	5.5	98
Fe ₃ O ₄ /RGO	5	2.2	-67.1	1.8	5.24	131
γ-Fe ₂ O ₃ /CrGO	50	3.25	-43.13	3.6	11.68	132
Fe ₃ O ₄ @NC@rGO	25	2.0	-72.6	2.0	5.5	133
ZnO/NPC	40	1.55	-25	—	—	125
ZnO/NPC/RGO	40	2.4	-50.5	2.6	7.4	126
NC-Co ₃ O ₄ /CP	40	1.6	-41.27	—	—	52
Co ₃ O ₄ /N-C	40	1.5	-42.63	1.5	4.14	127
ZrO ₂ /C	50	1.5	-58.7	—	—	128
TiO ₂ /C	60	1.6	-49.6	1.6	4.6	129
CoO/Co/C	25	3.3	-66.7	5.1	1.8	130
Co ₃ O ₄ @C@α-Fe ₂ O ₃	20	3.7	-52.2	2.5	6.6	134
CoO/Co@C	70	1.5	-38.46	2.0	4.8	135
SnO ₂ /Co ₃ Sn ₂ @C	30	2.0	-46.8	2.5	4.8	136

sulfides such as MoS₂, CdS and CuS develop fast owing to their remarkable RL value. Based on this, Man *et al.* reported a yolk-shell FeS₂@C nanocomposite for microwave absorption.¹⁶ The MIL nano-spindles were firstly synthesized and then converted to Fe₂O₃@C after annealing in N₂ atmosphere. Finally, the FeS₂@C was obtained *via* a sulfidation process. The as-fabricated composites showed excellent absorbing properties having the RL of -45 dB with the matching thickness of 1.45 mm, and the broad bandwidth of 15.4 GHz, as shown in Fig. 9.

It is accepted that nonmagnetic metal oxides can be used to tune impedance matching. For example, Liang *et al.* designed a hetero-structured ZnO/NPC by using Zn-based MOF ZIF-8 as precursor materials, and the minimum RL achieved -25 dB with merely 1.55 mm of absorber thickness.¹²⁵ Similarly, Ji *et al.* compounded the rGO and ZnO/NPC composites through the simple hydrothermal method.¹²⁶ Thereinto, the ZnO/NPC were obtained *via* high-temperature carbonization at 700 °C under N₂ flow. The minimum RL of ZnO/NPC/RGO could reach -50.5 dB with a thin thickness of 2.4 mm. Ji and his group synthesized Co₃O₄ nanoparticles embedded in the carbon matrix, and the Co₃O₄ nanoparticles grow on the carbon paper, which exhibited the excellent absorbing properties. The samples showed RL value of -41.38 dB at 7.32 GHz with the absorbing thickness of 2.3 mm.⁵² Besides, Bai *et al.* fabricated stratiform Co₃O₄/N-doped carbon using a Co-based MOF as the precursor.¹²⁷ The optimal RL of the samples reached -42.63 dB with the thickness of only 1.5 mm. Besides the above metal oxides/PC composites, some similar composites and other certain elements derived materials like ZrO₂/C,¹²⁸ TiO₂/C¹²⁹ and CoO/Co/C¹³⁰ are listed in Table 4.

4.5 Other MOF-derived PC

In addition to those mentioned above, there are some PC composites with other composition, such as rare-earth metal, bimetal oxides and so on. Up to now, the rare-earth metal

organic frameworks (RE-MOFs) for microwave absorption are barely reported. However, RE-MOFs are one of the attractive members of MOFs family in the recent years due to the unique 4f electron layer of RE metal ions. And the highly connected structures of RE-MOFs would induce ultra-high surface areas, resulting in excellent absorbing performance.¹³⁷ Zhu *et al.* synthesized four RE-MOFs by hydrothermal reactions for microwave absorption with synergetic and complementary of permittivity and permeability.¹³⁸

Metal and metal oxides encapsulated in MOF-derived PC are generally received more attention. And multiple component composites are significant candidates with high property as microwave absorbers. Xu *et al.* synthesized flower-like heterogeneous Co/MnO@C, deriving from Co/Mn bimetal oxides MOF derivatives.¹³⁹ When the molar ratio of Co and MnO reached 2 : 1, composites exhibited an optimized RL of -55.3 dB with an absorber thickness of 2.4 mm and the EAB of 4.6 GHz from 7.4 GHz to 12.0 GHz. Moreover, necklace-like CNFs@Co/CoO composites derived from ZIF-67 penetrated throughout the carbon nanofibers were synthesized by simple carbonization process (Fig. 10).¹⁴⁰ Attributed to the unique conductive network and the synergistic effect of dielectric loss, magnetic loss and impedance matching, the incident microwaves would be sufficiently absorbed after the multiple reflections, scattering and attenuation. The more similar composites are tabulated in Table 5.

Bimetal oxides are explored to act as microwave absorbers. Bi *et al.* found the unique polaron excitations-enhanced absorbing properties *via* synthesizing PC with CoNiO₂ through ZIF-67. This properties effectively improved electron polarization, multiple internal reflections and high RL.¹⁴¹ In addition, metal sulfides and selenides can be homogeneously dispersed in the derived PC under the appropriate reaction conditions. For example, the CoS₂ was obtained after the carbonization and sulphidation process of Co-MOF precursors.¹⁴² Mao *et al.*



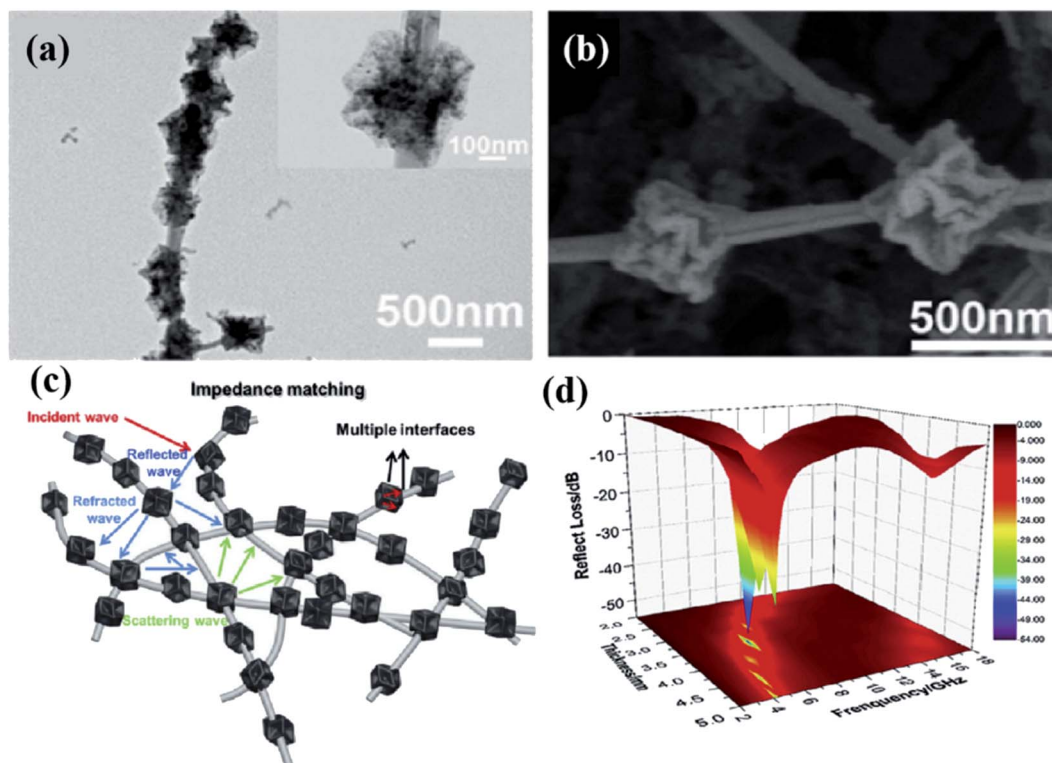


Fig. 10 TEM (a) and SEM (b) images, schematic diagram of microwave absorption mechanisms (c) and the RL (d) of CNFs@Co/CoO composites. This figure has been reproduced from ref. 140 with permission from Journal of Alloys and Compound, Copyright 2018.

Table 5 The microwave absorbing performance of other MOF-derived PC materials

Absorbers	Loading (wt%)	RL _{min}		EAB		Ref.
		Thickness (mm)	Value (dB)	Thickness (mm)	Value (GHz)	
[Er ₂ (MH) ₆] _n	20	5	-22.78	5	2.24	138
[Yb ₂ (MH) ₆] _n	20	4.5	-19.99	4.5	2.12	138
Co/MnO@C	25	2.4	-55.3	2.4	4.6	139
CNFs@Co/CoO	20	3.54	-53.1	2.0–5.0	13.52	140
C/CoNiO ₂	40	2.5	-53	—	—	141
C@NC/CoS ₂	30	2.8	-59.6	2.8	7.2	142
NiSe/C	50	3.7	-59.70	2.1	4.67	143
Co/ZnO/C	30	3.0	-52.6	2.5	5.8	144
Fe/Fe ₂ O ₃	30	2.2	-70.2	2.2	5.2	145
Fe ₂ N@NC	50	1.55	-59.3	1.9	4.32	146
NiO/Ni/C@Air@NiO/Ni/C	50	1.7	-34.5	1.7	~5.5	147
ZnO/C@Co/C	50	1.9	-28.8	1.9	4.2	148

successfully synthesized rod-like NiSe/C composites *via* the *in situ* selenization of Ni-MOFs with excellent microwave absorption properties.¹⁴³ The results and the performance of other MOF-derived PC materials are displayed in the Table 5.

5. Summary and conclusions

In the past few decades, MOF-derived PC composites have been increasingly reported for microwave absorption. And the diverse and unique chemical structures (light-weight, specific surface area, chemical stability and high porosity), tunable properties

(electronic conductivity, impedance matching) and facile preparation methods make derived PC composites be sought-after. The varied microstructures and compositions of derived PC are ascribed to the special nature of MOF, and the porosity can be adjusted *via* controlling the carbonization temperatures. These indicate that MOF-derived PC composites are admirable candidates of microwave absorbers.

The synergistic effect between the dielectric loss and the magnetic loss contributes to the efficient and strong attenuation capability, which can be tuned by well designing the constitution of derived PC materials and other microwave



absorbers and adjusting the proportion of effective constituent. Up to now, the MOF-derived PC composited magnetic metals or metal oxides and nano-PC are growing fast, which can overcome the problem of nano-scale particles agglomeration and protect the metal particles from external corrosion. The present of derived carbon skeleton with rich pores establishes the conductive path for electron migrating and hopping, and enhances the interfacial polarization and multiple reflections.

However, there are still some challenges for MOF-derived PC materials to face. For example, the toxic solvents and harsh fabrication condition are generally required in the fabrication process of MOF materials. It is highly desirable to design an environment friendly and facile fabrication strategy to construct MOF materials. The compositions of current absorbers of MOF-derived carbon materials are mainly large density metal like Fe, Co and Ni, which have been studied comprehensively in the previous research. However, the chemical composition is much dependent on the corresponding MOF precursors to be tuned, besides, the heavy metal composites often lead to the high filler loading. Thus, it is significant to find novel composite, diverse and lightweight MOF-based PC composites to achieve preferable microwave absorption. In addition, the research progress is almost theoretical stage. The practical applications as microwave absorbers are rarely reported due to the challenge of mass production. Based on this, MOF-derived PC materials are expected to be one of admirable candidates with efficient microwave absorption performance and facile preparation process. With the widespread attentions, MOF-derived PC materials would increasingly promote the advancement of microwave absorption field.

Author contributions

Ma Mingliang: conceptualization, resources, writing-reviewing and editing Bi Yuxin: conceptualization, investigation, writing-original draft preparation, writing-review and editing Tong Zhouyu: writing-reviewing and investigation Liu Yanyan: editing Lyu Ping: supervision Wang Rongzhen: investigation Ma Yong: writing-review and editing Wu Guanglei: editing Liao Zijian: investigation Chen Yan: investigation.

Conflicts of interest

There are no conflicts to declare.

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