

Research Progress and Current Bottlenecks of MOF-Based Microwave Absorbing Materials

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Abstract: Due to the explosive growth of wireless communications, radar detection, and electronics, the development of high-performance microwave-absorbing materials to attenuate harmful electromagnetic waves and achieve electromagnetic compatibility and stealth protection holds significant scientific importance and urgent application demands. MOF-based materials serve as an ideal platform for integrating dielectric and magnetic losses. MOF-based materials are a perfect medium where dielectric and magnetic losses may be integrated. By carefully designing the composition and structure, several loss mechanisms can be combined in situ to create a synergistic effect that effectively balances the contradictory requirements of impedance matching and effective dissipation, which makes it highly promising in the area of microwave absorption. This paper has been systematically reviewed to provide a summary of the research progress on the combination of MOFs with zero-dimensional, one-dimensional, and two-dimensional nanomaterials to absorb microwaves. Beginning with the loss mechanisms underlying different dimensional composite strategies, the synergistic effects are explained in turn: the incorporation of the magnetic loss and heterogeneous interfaces due to 0D magnetic nanoparticles, the increase in the conduction loss and multiple scattering due to 1D conductive networks, and the improvement in the interfacial polarization and impedance matching due to 2D lamellar structures. The effectiveness and shortcomings of each strategy with regards to the goals of thin thickness, light weight, broadband, and high absorption are outlined through typical case studies. Finally, there is a summary of the results obtained in MOF-based microwave absorbers and recommendations on the future development directions.

Keywords: MOF; Microwave Absorption; Multiscale; Dielectric Loss

1. Introduction

Because of the fast advancement of wireless telecommunication, radar, and other electronic gadgets, electromagnetic wave pollution is now the fourth major type of environmental pollution after water pollution, air pollution, and noise pollution. Electromagnetic radiation does not just disrupt the normal functioning of high-precision electronic devices, but also inflicts some damage on the human body. Hence, the creation of high-performance microwave-absorbing materials that can reduce adverse effects of electromagnetic waves and provide electromagnetic compatibility and stealth protection has great scientific importance and is practically needed. Ideally, microwave-absorbing materials would be thin, lightweight, have wide bandwidth, and strong absorption, namely, they would be low-matching thickness, low density, wide effective absorption bandwidth, and have high reflection loss. Nevertheless, conventional absorbing materials like ferrites, metallic powder, and conductive polymers have high density, limited absorption bands, poor thermal stability, or less extensive mechanisms of loss, which makes it challenging to meet the ever-increasing demands of service. Consequently, the quest for new high-efficiency microwave-absorbing material systems has become a research hot topic in the said area.

Metal-organic frameworks (MOFs) are crystalline porous materials formed by metal ions or clusters with organic ligands self-assembling through coordination. Because of their ultra-high specific surface area, large number of pore structures, tunable metal/ligand compositions, and easy functionalization, MOFs have recently demonstrated significant application possibilities in the area of microwave absorption. By pyrolyzing MOFs in an inert atmosphere, they can, on the one hand, be in situ transformed into porous carbon/metal/metal oxide nanocomposites; the resulting carbon structures serve as conductive loss and dipole polarization, metal or oxide nanoparticles cause magnetic loss, and

porous structures are used to optimize the impedance match and increase the propagation lengths of electromagnetic waves, which results in the synergistic combination of several loss mechanisms. Alternatively, the structural designability of MOFs enables scientists to freely control the electromagnetic properties of composite materials either by changing the metal species, the type of ligand, or the extent of the pyrolysis process, or by adding different dimensional nanounits, offering a perfect platform to design high-performing absorbers on demand.

In recent times, the studies of microwave-absorbing materials based on MOFs have been diversified beyond simple carbonization of a single-component system to the multi-dimensional heterostructure composite phases, leading to various research directions, including the synthesis of MOFs with zero-dimensional (0D), one-dimensional (1D), and two-dimensional (2D) nanomaterials. In addition to the enhancement of the loss mechanisms due to the introduction of nanounits of different dimensions, the materials are endowed with special physical and functional properties: 0D nanoparticles can induce magnetic loss and form nanoscale heterogeneous interfaces; 1D carbon skeletons can combine to form conductive networks to improve conduction loss and multiple scattering; and 2D lamellae can form multilevel heterogeneous interfaces by having large van der Waals contact areas to increase interfacial polarization. However, these significant advances notwithstanding, there are still a number of challenges that remain, like challenging synthesis, mutual limiting properties of various functional parameters, which are challenging to obtain at the same time, low frequency absorption ability, poor environmental performance, and inadequate knowledge of loss mechanisms that should be summarized and discussed further.

The present review focuses on summarizing the state-of-the-art research on the integration of MOFs with various dimensional nanomaterials (0D, 1D, and 2D) in microwave absorption and gives an in-depth comment on the attenuation mechanisms as well as the structure-absorption performance relationships of every composite system. Chapter 2 initially covers the classifications of the MOF-0D material combinations, MOF-1D material combinations, and MOF-2D material combinations, and complements them with representative literature samples with analysis. Chapter 3 provides an overview of the current situation in this area and makes forecasts regarding the further development tendencies. It is anticipated that this review will be a valuable source of information when designing, synthesizing, and using MOF-based high-efficiency microwave absorber materials.

2. Research Progress of MOF-Based Microwave Absorbing Materials

2.1 Compositing MOFs with 0D Materials

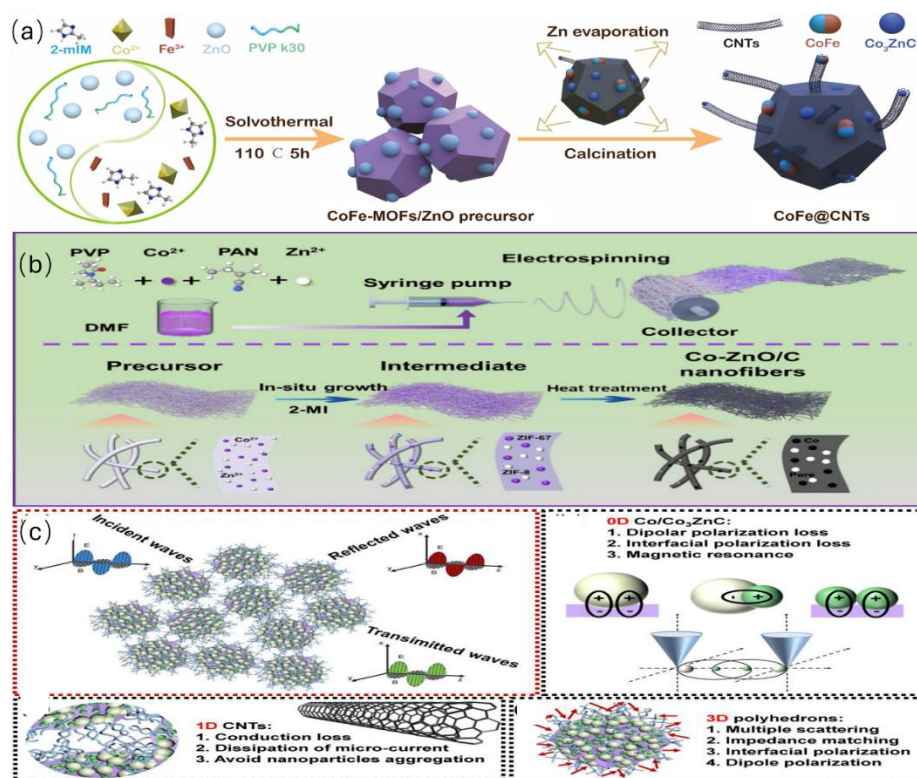
Pure MOFs are mostly insulators or wide-bandgap semiconductors with extremely low electrical conductivity and a lack of magnetic loss. Its loss depends solely on the weak dipole polarization by ligand defects or polar groups, and hence it has a single loss type and a limited frequency band. Besides, due to their relatively small permittivity, their impedance is significantly higher than that of free space, which results in strong reflection of electromagnetic radiation and ineffective absorption. On the other hand, 0D magnetic metal/alloy nanoparticles and oxides have large saturation magnetization and permeability, and may offer significant magnetic losses (natural resonance and exchange resonance) to overcome the lack of magnetic loss. At the same time, their small-size factor and large specific surface area also serve as alternative dipole polarization sources, allowing efficient broadband and strong attenuation.

Zhang et al. ^[1] suggested a 0D/1D composite optimization strategy that involves component design and microstructure engineering to improve the electromagnetic attenuation of MOF-derived absorbers. This design uses cubic dual-phase CoFe alloy as the magnetic loss backbone. At the same time, in situ carbon nanotubes add significant conduction loss to the nanocomposites and, at the same time, increase interfacial polarization and dipole polarization. The obtained CoFe@CNTs nanocomposite (Fig. 1(a)) has a lowest RL of -49.63 dB at 16.88 GHz (1.5 mm thickness) and an EAB of 6.32 GHz (1.71 mm). When compared to single-component Co nanoparticles, the CoFe@CNTs nanocomposite has a lower RL, a higher bandwidth, and a thinner thickness with good microwave absorption properties. It is thus a potential candidate to be used in effective electromagnetic absorption.

The paper by Wang et al. ^[2] presented an innovative synthesis approach, combining electrospinning, a seed-driven growth of zinc-cobalt bimetallic metal-organic framework (MOF), and carbonization, to fabricate one-dimensional (1D) porous ZnO/C fibers containing zero-dimensional (0D) cobalt nanoparticles (Co-ZnO/C) (Fig. 1(b)). Changing the proportion of cobalt ions in the fibers can be used

to regulate the level of graphitization and electromagnetic properties of the fibers in order to make them better suitable to absorb microwaves. Moreover, the small Co particles greatly enhance interfacial polarization and magnetic loss, and the hierarchical porous architecture is used to match impedance. Consequently, increasing the cobalt content improves RL_{min} from -50.83 dB to -63.42 dB, whereas EAB_{max} slightly decreases from 7.28 GHz (10.72–18.00 GHz) at 2.50 mm to 7.04 GHz (9.12–16.16 GHz) at 2.90 mm. Excessive addition of cobalt ions also leads to degradation of the microwave absorption performance. The unique one-dimensional porous structure extends the electron transport path of the Co-ZnO/C composite and improves its thermal conductivity. The corresponding thermal conductivity reaches 0.318 W m⁻¹ K⁻¹, which is 40% higher than that of pure epoxy resin. Co-ZnO/C composites have good electromagnetic wave absorption performance and high thermal conductivity, with potential applications in the development of future wireless communication equipment and high-power devices.

The research of Zhang et al. [3] developed a multi-dimensionally integrated Co₃ZnC/Co@C heterostructure made up of 0D Co and Co₃ZnC nanoparticles, 1D carbon nanotubes, and 3D porous carbon polyhedra via pyrolyzing a bimetallic MOF. Porous carbon polyhedra, which are grafted with carbon nanotubes, have been uniformly filled with Co and Co₃ZnC nanoparticles, which form a 0D-1D-3D integrated heterostructure. Component-modulated electromagnetic parameters and microwave absorption performance are shown to be possessed by the Co₃ZnC/Co@C composite that may be easily tuned by varying the Co/Zn ratio of the MOF precursor (Fig. 1(c)). With a filler loading of 30 wt% the best Co₃ZnC/Co@C heterostructure has a minimal reflection loss (RL) of -66.78 dB at 7.43 GHz with a thickness of 4.5 mm, which is within the C band. The maximum effective absorption bandwidth (RL ≤ -10 dB) is highest at 2mm (4.8GHz (12.9-17.7GHz)). High-efficiency microwave absorption performance is due to the capability of the multi-dimensionally integrated heterostructure to make full use of the synergistic effects of the individual components. Evenly distributed 0D Co and Co₃ZnC nanoparticles can enhance magnetic loss and interfacial polarization. These results offer some new perspectives on how to achieve high-performance microwave absorption based on component modulation and multi-dimensional structural integration.



(a) schematic illustration of the synthesis process of CoFe@CNTs nanocomposites;
 (b) schematic illustration of the synthesis route of Co-ZnO/C fibers;
 (c) schematic representation of the composition and multi-dimensional improved microwave absorption processes in the Co₃ZnC/Co@C heterostructure.

Figure 1: Preparation procedure of MOF composite with 0D materials and mechanism of increased microwave absorption.

To summarize, combining 0D magnetic nanoparticles and carbon structures based on MOF produces a magnetic-dielectric composite loss system that incorporates several loss factors. The multi-dimensional heterogeneous porous structure helps improve the interfacial polarization and optimize the impedance matching, which in turn enables the passage and dissipation of electromagnetic waves. At small thicknesses, adjustability of compositions results in strong absorption and broadband absorption, and certain ones of them have thermal conduction capability as well, which is a prospective design approach of structural-functional integration. Nevertheless, there are bottlenecks: magnetic parts are prone to high sensitivity, and over-concentration causes impedance mismatch; high absorption, high bandwidth, and thin thickness are interrelated; multi-dimensional structure is hard to achieve, and filler filling is not beneficial to the light-weight design. Future work must carefully arrange the components and microstructures, create low-filler-loading multi-dimensional designs, green synthesis, and multifunctional characteristics, including thermal conductivity and high-temperature resistance, and thus advance application to practice.

2.2 Compositing MOFs with 1D Materials

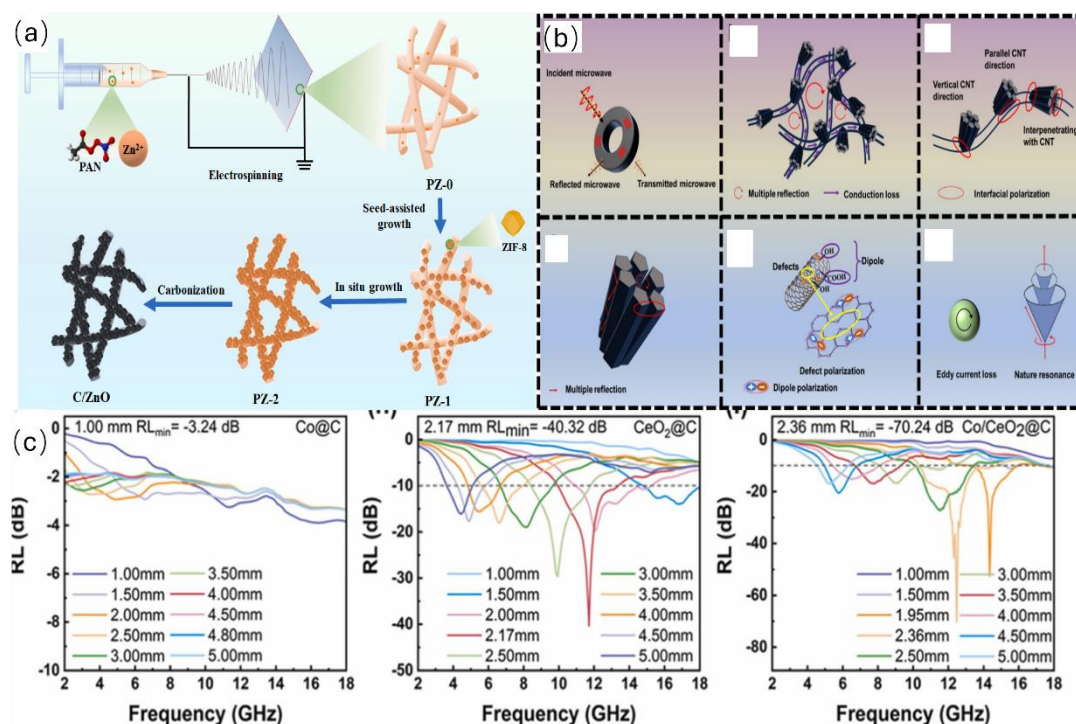
In contrast to 0D composites, 1D materials have a number of benefits. Due to the high aspect ratio, it is possible to build three-dimensional conductive pathways in the matrix at extremely low filler loading, improving conduction losses and not compromising the impedance match. The stretched geometry causes any incident electromagnetic radiation to be reflected and scattered internally many times, which augments the effective path length as the radiation propagates. The large surface area has plenty of dipole polarization sites and dangling bonds, and there are wide areas of heterogeneous interfaces in the axial direction, which generate strong interfacial polarization relaxation. Moreover, their excellent flexibility allows for creating free-standing films or flexible fabrics that have lightweight properties yet perform mechanically.

C/ZnO nanofiber composites have been synthesized by Wang et al.^[4] through a seed-assisted in situ electrospinning technique using a zinc-based imidazolate framework (ZIF-8) as the precursor (Fig. 2(a)). The special porous structure and different interfaces between C and ZnO allow both scattering and multiple reflections of microwaves. Due to the joint action of interfacial polarization loss, dipole polarization loss, and conduction loss, the C/ZnO nanofiber composite has very high microwave absorption performance with RL_{min} -58 dB at 7.5 GHz and EAB of 6.5 GHz.

The team of Guo et al.^[5] synthesized Ni-MOF with various forms of microstructures in situ onto carbon nanotube monofilaments with a hydrothermal process at varying ratios of molar concentrations of nickel ions to organic ligands. The ultra-flexible standing Ni-MOF/CNT buckypaper (BP) was produced by the directional pressure filtration method based on these reasons. The skeletal porosity of BP and the distinctive porosity of Ni-MOF give efficient impedance matching. The existence of carbon nanotubes adds to the conduction loss, the cross-scale heterogeneous interfaces that are created during the formation of Ni-MOF/CNT BP result in a high amount of interfacial polarization loss, and the porous structure makes the propagation of microwaves more complicated (Fig. 2 (b)). These all factors combine to give the Ni-MOF/CNT BP great microwave absorbency properties. The lowest reflection losses of the Ni-MOF/CNT BPs loaded with granular, hollow porous prism, and porous prism geometries are -50.8, -57.8, and -43.3 dB, respectively. The associated effective absorption bandwidths are 4.5 GHz, 6.3 GHz, and 4.8 GHz, respectively. Also, the BPs are very flexible because they can be rolled over a glass rod (4 mm in diameter) up to hundreds of times without causing any structural damage. The paper suggests a novel idea of creating ultra-flexible self-supported microwave-absorbing materials with hierarchically interpenetrating porous structures based on MOFs, which may find applications in flexible electronics.

Yu et al.^[6] successfully synthesized Co/CeO₂@C porous nanorods through the high-temperature calcination method in the Ar atmosphere using Zn-Co-Ce-MOF-74 nanorods as the templates. With a filler level of 20 wt%, Co/CeO₂@C porous nanorod demonstrates the least reflection loss of -70.24 dB at 12.48 GHz and 2.36 mm, and the effective absorption bandwidth is 4.34 GHz (Fig. 2(c)). Moreover, CST simulations indicate that the composite Co/CeO₂@C has a good radar cross-section (RCS) reduction ability. Co/CeO₂@C composite shows a reduction rate of 31.86 dB m² and effective absorption of electromagnetic waves at any angle of incidence. The good microwave absorption characteristics of porous Co/CeO₂@C nanorods are due to their peculiar structure that enhances impedance matching and offers a variety of loss mechanisms, such as conduction loss, dipole polarization loss, magnetic loss, and interfacial polarization loss. The present work gives a practical pathway towards the preparation of porous Co/CeO₂@C nanorods, which can be applied to other one-dimensional dielectric/magnetic nanocomposites with required microwave absorption properties.

Consequently, once MOFs have interacted with 1D materials, the interconnected 1D units create a three-dimensional conductive network, which leads to higher conduction losses and better matching of impedances. Lengthening the propagation distance and increasing the attenuation is achieved due to the porous structure that causes multiple internal reflections and scattering of electromagnetic waves. Various interfaces of different sizes cause intense interfacial polarization relaxation phenomena and link to heteroatom dipole polarization and magnetic loss; thus, enabling high reflection loss and wide absorption bandwidths. Certain systems may be bent repeatedly without the structural failure. But multi-step procedures are unwieldy, and it is hard to effectively manipulate one-dimensional unit morphologies and interfacial assembly forms. Also, there are natural contradictions between intensive absorption, large bandwidth, ultra-thinness, and small filler load. Hence, future studies ought to investigate more straightforward and economical synthesis processes, enhance the coordination of design between compositions and microstructures even more, and use in situ characterization and computational simulation to explain the interactions between several loss mechanisms and thus develop pre-research design and multi-system integration.



(a) graphical description of the sample preparation strategy;
 (b) schematic illustration of the absorption mechanism of Ni-MOF/CNT BP;
 (c) one-dimensional RL curves of Co@C, CeO₂@C, and Co/CeO₂@C composites.

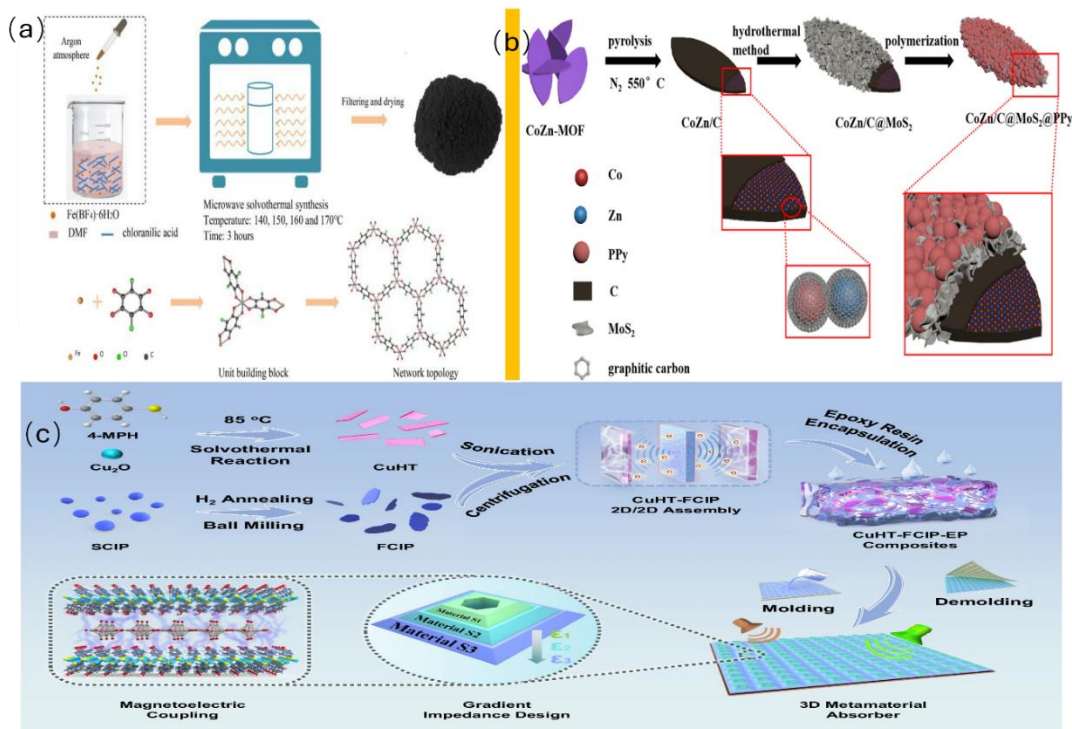
Figure 2: Sample preparation strategy, microwave absorption mechanism of Ni-MOF/CNT BP, and RL performance of the composites.

2.3 Compositing MOFs with 2D Materials

2D materials have planar anisotropy and specific surface areas of ultra-large scale. The wide contact area between sheets may develop high-density heterogeneous interfaces, triggering severe interfacial polarization relaxation; surface functional groups, defects, and vacancies can be used as dipole polarization centers. Intercalation spaces will force electromagnetic waves to repeatedly reflect internally and scatter, increasing the propagation length, and the effective permittivity may be adjusted by changing the intercalation distance and stacking type, and thus the impedance match is enhanced. The ability to conduct electricity and have polarity makes some 2D materials capable of creating a magnetic-dielectric-dual-loss system, and they are flexible enough to enable the production of lightweight free-standing absorbers.

According to Wei et al.^[7], they offered a new idea which was grounded on the use of the intrinsic properties of MOFs. Sheet-like redox-active iron-quinone MOFs (Fig. 3(a)) were produced by a simple and fast microwave solvothermal process, which varied depending on the reaction temperature, both in terms of morphology and crystallinity. The fabricated MOF has an electrical conductivity of up to $2 \times$

10^{-3} S/m, comparable to that of semiconductors. Moreover, magnetization hysteresis loops were measured at 300 K. Reaction temperature is the main factor that can realize maximum microwave absorption with a reflection loss of -73.5 dB at 13.8 GHz and a matching thickness of 3.3 mm. At a broadband frequency range of 9.8-15.9 GHz, an effective absorption bandwidth with reflection loss less than -10 dB is also achieved. The best performance of microwave absorbers is mostly due to the fact that magnetic loss and dielectric losses are simultaneously realized within a single iron-quinone MOF, thus attaining adequate impedance matching using variable magnetic and electrical properties. The created iron-quinone-based MOF materials provide new possibilities for implementing efficient, lightweight, and adjustable microwave absorbers.



(a) schematic illustration of the synthesis process and self-assembly of iron-quinone MOF;
 (b) schematic illustration of the formation process of CoZn/C@MoS₂@PPy composites;
 (c) schematic illustration of the preparation of the CuHT-FCIP-EP metamaterial absorber.

Figure 3: Preparation of MOF composites.

The MOF self-templating strategy was used by Bi et al.^[8] to synthesize CoZn/C@MoS₂@polypyrrole (PPy) composites (see Fig. 3(b)). By combining MoS₂ sheets with a PPy shell, the impedance match between the two-dimensional (2D) CoZn/C composite can be enhanced, and MoS₂ sheets combined with a PPy shell enhance the microwave absorbing properties of the composite. The produced CoZn/C@MoS₂@PPy composite has the lowest RL of -49.18 dB at a thickness of just 1.5 mm; besides, the effective absorption bandwidth (EAB, RL ≤ -10 dB) is also 4.56 GHz, which means that it is more effective in absorbing microwaves than the CoZn/C composite at lower thicknesses (<2 mm). Besides providing a simple way of making MOF-derived carbon-based composites as microwave absorbers, this paper widens the use of MOF materials.

It is also extremely important to combine macroscopic design of structure with microscopic design of materials in order to create more effective electromagnetic wave (EMW) absorbing materials. Qu et al.^[9] suggested a metamaterial design (Fig. 3(c)) to overcome the long-standing issues of this area, such as limited bandwidth, low-frequency bottlenecks, and, in particular, the acute problem of strength (i.e., oblique incidence, and polarization incidence). The absorber of Qu et al.^[9] has a semiconducting metal-organic framework/iron 2D/2D assembly (CuHT-FCIP) that has plenty of crystalline/crystalline heterojunctions and a robust magnetoelectric coupling network. Based on the above, a design with high-performance EMW absorption across a wide frequency range (2-40 GHz) was achieved with a thickness of just 9.3 mm, good stability at an oblique incidence range (up to 75°), and across different EMW polarizations (TE and TM). At the same time, this SAM has high specific compressive strength (201.01 MPa·cm³ g⁻¹) and low density (0.89 g·cm⁻³), allowing preparation of high-performance strong EMW absorbers.

The combination of MOFs with 2D materials has been developed through induced carbonization to intrinsic electromagnetic behaviour of two-dimensional MOFs, which may incorporate electrical conductivity and magnetism. Interfacial polarization is increased by high-density heterogeneous interfaces and core-shell structures. Metamaterial design, ultrabroad bandwidth, high resistance to oblique incidence, and lightweight high strength can be combined to create a cross-scale synergy. Currently, intrinsically active systems are limited and poorly stabilized; heterogeneous construction is intricate and costly; the conflict between ultrathin thickness, broadband, and low-frequency absorption is significant; and there is no study of synergistic loss mechanisms. The further development of stable systems with a focus on green large-scale synthesis, precise coordination of components and microstructure, and resolving the performance indicator conflicts should be considered in future work, as well as the combination of in situ dynamic characterization and multiscale simulations to discover the synergistic character and accomplish rational design.

3. Conclusion

Due to their highly tunable constituents, high pore count, and ease of multi-dimensional compositing, MOFs and their derivatives have become ideal construction systems to develop high-performance microwave absorbing materials. It has been systematically reviewed how the compositing of MOFs with zero-, one- and two-dimensional nanomaterials has been studied, and it was found out that various loss mechanisms and structure-activity relationships were observed at various dimensional strategies. Overall, present work has shifted towards a multi-dimensional heterogeneous coupling rather than a single component-derived carbonization. Through the realization of dielectric and magnetic dual losses and polarization effects enhancement by multi-scale heterogeneous interfaces, and impedance transition by porous conductive networks, significant progress has been achieved in the thin, light, wide, and strong performance. Besides, first-principle application opportunities of functional integration like flexibility, heat transfer, and load-bearing have been identified, which implies that MOF-based absorbent materials are evolving in terms of performance optimization into performance and functionality integration.

Nevertheless, there are numerous deep-rooted issues that persist between laboratory samples and practical applications. Refined three-dimensional heterogeneous structures are obtained through a complex multi-step preparation process; it is challenging to replicate the micro/nanostructures on a large scale; certain precursors and nanomaterials are costly; green and environmentally friendly methodologies of synthesis are absent. Moreover, ultrathin thickness, broad spectral response, and high absorption efficiency have intrinsic conflicts. The best performance usually only occurs at particular matching thicknesses and fairly high filler loading, and low-frequency absorption capability is poor. One-component MOFs with intrinsic electromagnetic dual-response properties are quite uncommon, and their survivability when exposed to extreme operating conditions, as well as their multifunctional composite use, are at an early experimental level. Further down, there is a lack of thorough combination of in situ dynamic analysis and multi-scale theoretical modeling in the synergism of various loss mechanisms at multi-dimensional interfaces. There are no standardized testing and evaluation systems, which also hinders rational comparison between different systems, and thus material design still has not essentially departed the paradigm of empirical trial and error.

The future development of MOF-based microwave absorbing materials must be directed towards the following areas: (1) formulation of green, straightforward, and scalable synthetic strategies, and implementation of the transformation of MOF materials into the laboratory and pilot scale, and industrial production; (2) enhancement of the precise coordinated control of components and hierarchical microstructures, and incorporation of big data methods like machine learning and high-throughput screening to expedite the rational design of optimal composition-structure-performance combinations; (3) increasing the multi-scale interaction of macro/microstructures, including metamaterials and materials, to eliminate all the contradictions between ultrathin thickness, broadband, high absorption, and angular polarization insensitivity; (4) enhancing low-frequency absorption efficiency and multifunctional integration, and creating multifunctional materials with wave absorption, thermal conductivity, high-temperature resistance, and corrosion resistance to satisfy the requirements of harsh working conditions; (5) formation of standardized performance testing and evaluation specifications, tight connection between in situ characterization methods and multi-scale simulations in order to understand the fundamental processes of electromagnetic wave propagation through complex hierarchical structures, and thus direct the design and synthesis of next-generation high-efficiency MOF-based microwave absorbing materials and bring them to practical application.

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