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State of the art and prospects in metal-organic framework-derived microwave absorption materials

Xiao-Xuan Fan[1], Xin-Ci Zhang[1,*], Lin Li[1,*], Mao-Sheng Cao[2,*]

[1]Key Laboratory for Photonic and Electronic Bandgap Materials Ministry of Education, School of Physics & Electron Engineering, Harbin Normal University, Harbin 150025, Heilongjiang, China.
[2]School of Materials Science and Engineering Beijing Institute of Technology, Beijing 100081, China.

[*]**Correspondence to:** Dr. Xin-Ci Zhang and Prof. Lin Li, Key Laboratory for Photonic and Electronic Bandgap Materials Ministry of Education, School of Physics & Electron Engineering, Harbin Normal University, 1 Shida Road, Hulan District, Harbin 150025, Heilongjiang, China. E-mail: zhangxinci@hrbnu.edu.cn; lil@hrbnu.edu.cn; Prof. Mao-Sheng Cao, School of Materials Science and Engineering Beijing Institute of Technology, No.5 Zhongguancun South Street, Haidian District, Beijing 100081, China. E-mail: caomaosheng@bit.edu.cn

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Abstract

Microwaves are currently used in many fields, including the military, medical, and communication. However, the ensuing electromagnetic radiation has seriously threatened human life. Therefore, the design of high-performance microwave absorbing materials (MAMs) has become an important development direction. Metal-organic frameworks (MOFs) are regarded as a bright new star among MAMs with broad application prospects due to their advantages of tunable structure, large specific surface area, high porosity, *etc*. This paper reviews the research progress of MAMs derived from MOFs in recent years, including preparation methods, properties and microwave absorption mechanisms. Finally, the problems and research prospects of MOF-derived MAMs are discussed.

Keywords: Microwave absorption materials, metal-organic frameworks, mechanisms of microwave, preparation methods

INTRODUCTION

The rapid advancement of technology has introduced microwave pollution, which can harm the human nervous system[1-10]. Consequently, the development of advanced microwave absorbing materials (MAMs)

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has become an urgent requirement. Ideal MAMs should possess excellent microwave absorption (MA) properties, a wide effective absorption bandwidth (EAB), and be lightweight and thin[11-15]. Developed MAMs can be broadly categorized into three groups: conductive polymers, conductive carbon matrices, and magnetic materials[16-23]. While these materials are easy to prepare and relatively inexpensive, their single-component nature often causes impedance mismatch, hindering development[24,25]. Therefore, integrating carbon-based materials and metals in MAMs presents an effective solution to the challenges[26-28].

Metal-organic frameworks (MOFs) consist of organic ligands and metal ions through coordination interactions, and possess unique advantages of tunable structure, high porosity, good thermal stability, and many metal coordination sites[29-31]. Therefore, they are often used as templates or precursors for MAMs, but their low conductivity poses a challenge. The commonly used workaround is to carbonize MOFs at high temperatures to synthesize porous carbon and metal or metal oxide composites with specific pore structures, designed to create numerous defects, rich interfaces, and additional magnetic losses[32,33]. Maintaining the morphology and porous structure of the original MOF can enhance multiple scattering and reflection, which further promotes electromagnetic wave (EMW) dissipation[34]. Additionally, MA performance can be optimized by changing the types of organic ligands and metal ions to achieve a combination of being "light, thin, wide and strong".

Previous research has mainly focused on component regulation to design and develop MAMs by altering metal ions or combining MOFs with other materials[35,36]. The dimensions and morphology of MOF-derived materials also significantly influence their properties, making classification by dimension an effective approach [Figure 1][37-44]. Each dimension offers unique advantages: zero-dimensional (0D) materials have abundant magnetic particles, one-dimensional (1D) materials form conductive networks, two-dimensional (2D) materials possess excellent dielectric properties, and three-dimensional (3D) materials create numerous heterogeneous interfaces. Thus, optimizing MA performance by adjusting precursor dimensions and morphology is a promising research direction[37].

This paper systematically summarizes recent research progress on MOF-derived MAMs, including morphological design, advantages, and MA characteristics. The electromagnetic response mechanism of MAMs is elucidated. Finally, it discusses current challenges and future development prospects.

MAMS DERIVED FROM MOF

Core-shell and hollow structures are the prevalent morphological types in MOF-derived 0D structures. The core-shell structure can form a rich heterogeneous interface to improve interface polarization, while the hollow structure promotes multiple reflections and scattering of EMW. In the carbonization process, graphitized carbon typically forms a carbon shell, and magnetic metal particles form a magnetic core, facilitating the preparation of core-shell MOF derivatives. Wang *et al.* successfully prepared yolk-shelled Ni@C@ZnO through the pyrolysis of a bimetallic Ni-Zn MOF precursor [Figure 2A][40]. Ni@C@ZnO has a layered porous yolk-shell structure [Figure 2B and C]. During pyrolysis, ZnO nanosheets are formed through the reaction of oxygen atoms with Zn^{2+}, which are embedded into carbon matrices [Figure 2D]. The interfacial polarization and magnetic-dielectric synergistic effects contribute to the remarkable MA performance of Ni@C@ZnO, with a minimum reflection loss (RL_{min}) of -55.8 dB at a thickness of 2.5 mm.

MOF derivatives were prepared by directly carbonizing MOFs, which was also effective for the preparation of hollow structures. Ma *et al.* synthesized MAMs with hollow bowl-like structures by straightforward solvent heating and carbonation [Figure 2E and F][45]. The synergy of the dielectric and magnetic components significantly optimizes impedance matching; the structure also has a large number of

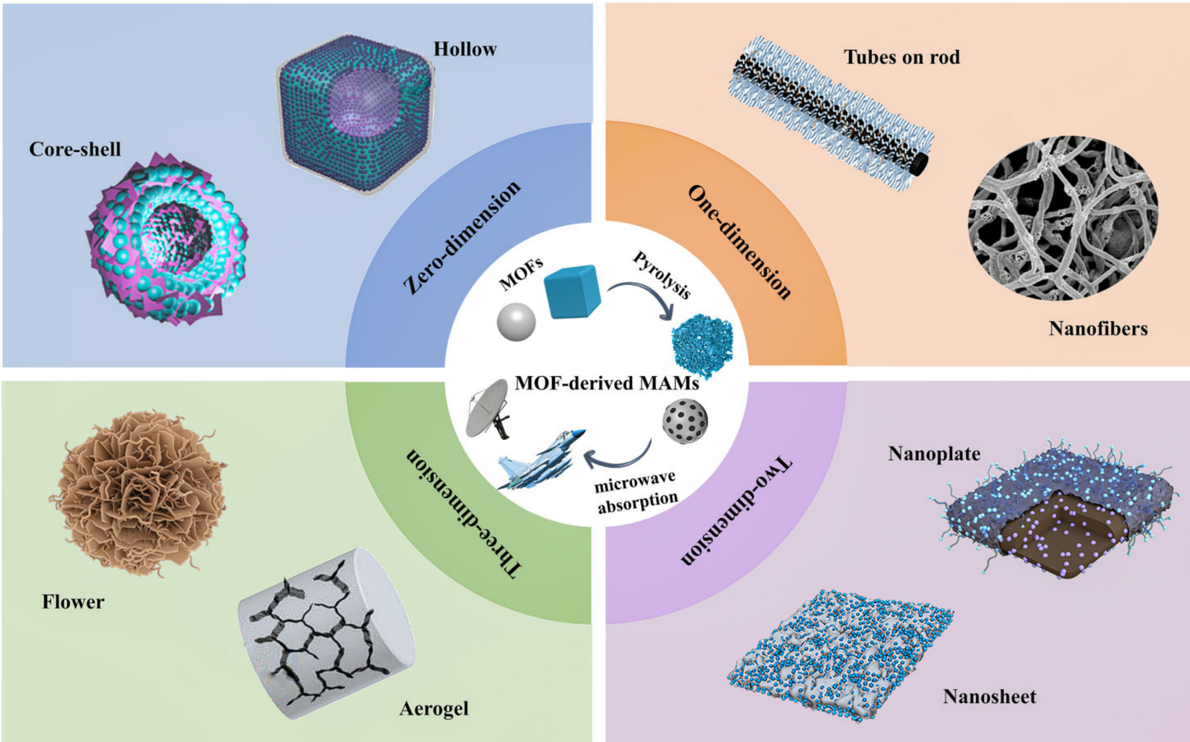

Figure 1. Schematic of MAMs derived from MOF in different dimensions. Reprinted with permission[37-44]. MAMs: Microwave absorbing materials; MOF: Metal-organic framework.

interfaces. Consequently, a RL_{min} of -52.66 dB is achieved, and the EAB achieves full coverage of the Ku-band [Figure 2G].

One-dimensional MOF-derived materials are crucial in MAMs for their unique structure that facilitates conductive network formation. Template strategies are often employed for their synthesis. Xue *et al.* successfully prepared Co/MnO/carbon nanotube (CNT) composites by template method[46]. ZIF-8@ZIF-67 was fabricated on MnO_2 nanorods using the precipitation method. Then, after metal catalysis and carbonation, a Co/MnO/CNT layered structure was formed [Figure 2H]. ZIF-8 is coated on the MnO_2 surface, forming a rough core-shell structure [Figure 2I]. Carbonization causes structural collapse, eventually forming a layered structure resembling a caterpillar [Figure 2J]. The structure has good impedance matching [Figure 2K]. Under the synergy of magnetic and dielectric loss, the composite material achieves a strong MA of -58.0 dB and 4.5 GHz at an ultrathin matching thickness.

Two-dimensional MOF-derived MAMs are gaining attention for their high specific surface area and low density, with the stripping method being a common preparation technique for 2D MOF derivatives. Yan *et al.* first mixed $Zn(NO_3)_2$ with methylimidazole to produce Zn-ZIF-L. Subsequently, metal chloride was added as a stripping agent and an etchant. The ZIF-L layer was exfoliated into N-doped porous graphene carbon nanonets (N-GN) [Figure 2L][47]. Because of the evaporation of Zn, the nanosheets are porous [Figure 2M and N]. The good structure, N heteroatoms, large pores, and abundant heterogeneous interfaces all lay the foundation for excellent MA properties. The optimum *RL* of -54 dB is achieved with a filler loading of 3 wt.%.

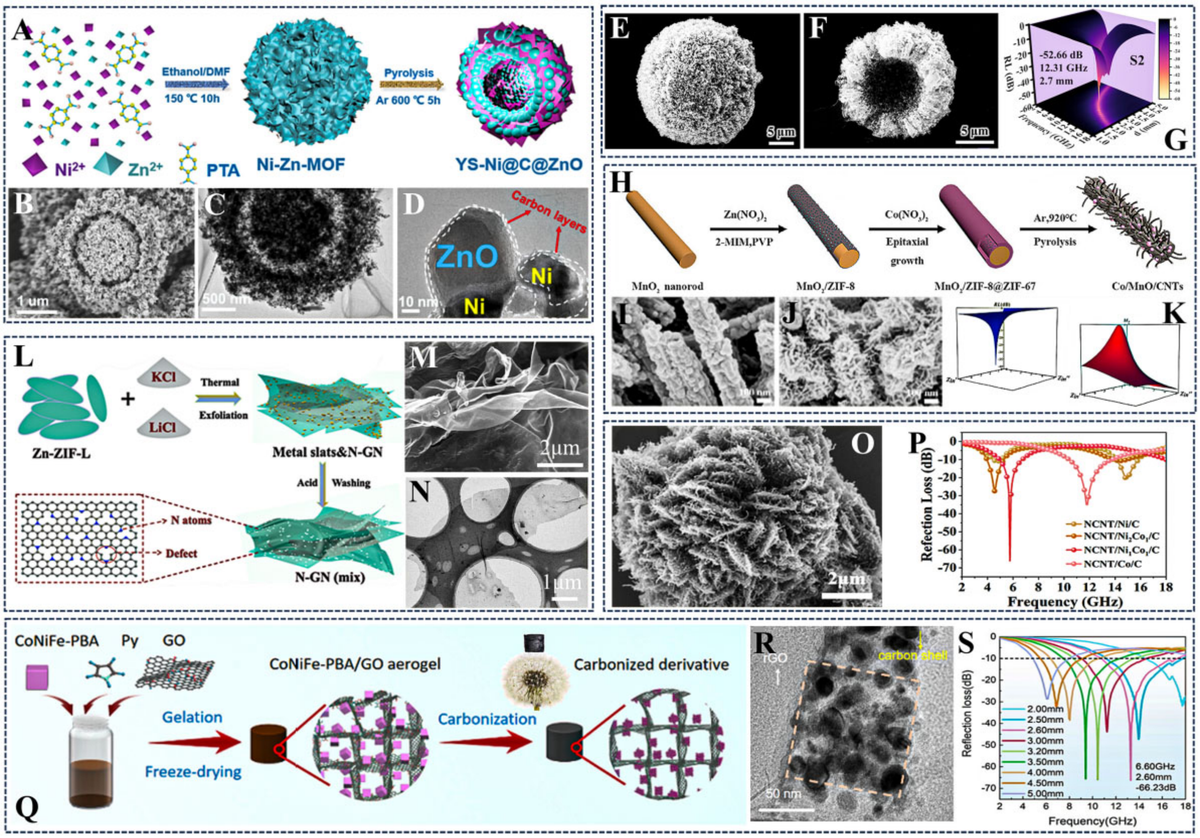

Figure 2. (A) Schematic diagram of the growth process of Ni@C@ZnO; (B) SEM image; (C, D) TEM plots. Reprinted with permission[40]; (E, F) SEM images; (G) 3D *RL* diagram of S2. Reprinted with permission[45]; (H) The illustration of the synthesis strategy of Co/MnO/CNTs, SEM plots for (I) MnO_2/ZIF-8 and (J) Co/MnO/CNTs; (K) The relationship between the *RL* and M_z with input impedance. Reprinted with permission[46]; (L) The illustration of synthesis strategy of N-GN; (M) SEM plots; (N) TEM images. Reprinted with permission[47]; (O) The SEM plots of NCNT/Ni_1Co_1/C, (P) RL_{min} value of all samples. Reprinted with permission[37]; (Q) The illustration of synthesis strategy of CoNiFe-PBA/GO aerogel derivatives; (R) TEM images; (S) RL_{min} value of S550. Reprinted with permission[33]. *RL*: Reflection loss; CNT: Carbon nanotube; N-GN: N-doped porous graphene carbon nanonets; NCNT: N-doped CNT; PBA: Prussian blue analog; GO: Graphene oxide; SEM: Scanning electron microscope; TEM: Transmission electron microscope.

The flower-like 3D structure enhances microwave energy loss by increasing microwave entry and extending the transmission path. Hou *et al.* synthesized floral N-doped CNT (NCNT)/NiCo/C nanocomposites by hydrothermal and vapor deposition processes using Bimetal-MOF as a precursor[37]. The composites show a porous flower-like structure, with CNTs uniformly distributed on the surface [Figure 2O]. The nanosheets are interconnected by nanotubes to form a unique 3D conductive network. Moreover, CoNi nanoparticles enhance the magnetic loss while forming a non-homogeneous interface. An optimal RL_{min} of -66.1 dB and EAB of 4.64 GHz can be achieved at a Ni/Co ratio of 1:1 [Figure 2P].

Carbon-based aerogels are ideal for MAMs because of their porous interconnect structure, low density, and good electrical conductivity. Three-dimensional aerogels can be constructed through the interfacial coordination of MOF precursors and graphene sheets. Wei *et al.* first assembled graphene oxide (GO) assisted by CoNiFe-Prussian Blue Analog (PBA) nano-cubes, then heat-treated to successfully fabricate CoNiFe-PBA/GO aerogel derivatives [Figure 2Q][33]. The square PBA derivatives were uniformly distributed in graphene oxide flakes [Figure 2R]. N-doped graphene oxide flakes produced abundant polarization loss. Furthermore, NiFe/CoFe nanoparticles encapsulated in defective carbon would form non-homogeneous interfaces and continuous conductive networks. Therefore, they exhibit strong *RL* and wide EAB in both the

X-band and Ku-band at very low filling contents [Figure 2S].

Table 1 summarizes and compares the EAB, *RL* and filling rates of MOF derivatives under different dimensions and morphologies.

MECHANISMS OF MA

When a microwave meets an absorber's surface, it can be absorbed, reflected, or transmitted [Figure 3A][48-50]. Deep penetration into the absorber causes microwaves to have multiple reflections on the inner surfaces, which helps dissipate microwave energy [Figure 3B][38,51,52].

Dielectric and magnetic losses are the main forms of microwave energy dissipation. Dielectric loss includes conductive loss and polarization relaxation loss [Figure 3C][40,43,44,53-57]. Conductive loss arises from the directional movement of free electrons within the medium under an alternating current electric field[58]. In MOF-derived MAMs, conductivity is enhanced by a strong conductive network due to the directional motion of charges, especially in 1D and 2D composites.

Polarization relaxation loss involves dipole and interfacial polarization, occurring during the directional rearrangement of electrons or molecules in response to an electric field[59]. High-temperature carbonization introduces new functional groups and defects in MOFs, disrupting electron equilibrium and creating dipoles. When the electric field changes or disappears, these dipoles are forced to rotate, leading to energy conversion and attenuation[60]. The heterogeneous interfaces between MOFs and other materials also enhance polarization relaxation losses.

In the 2 to 18 GHz band, magnetic losses include hysteresis loss, natural resonance, exchange resonance, and eddy current effects [Figure 3D][35,39,55,57,61-63]. Hysteresis loss arises from domain wall movement during magnetization, while eddy current loss heats the material and causes energy loss[64-66]. Natural resonances stem from the material's anisotropic field, and exchange resonance occurs in sub-micron or nano-sized particles.

There are two methods for adjusting magnetic loss in MOF-derived MAMs: introducing magnetic metals or transforming metal ions via carbonization. The introduction of magnetic metals can cause magnetic losses. Optimizing experimental conditions allows metal ions to be transformed into metals or metal oxides, regulating magnetic response to enhance magnetic loss.

CONCLUSION AND PERSPECTIVES

Researchers have developed various efficient MOF derivatives by carefully designing their morphology and dimensions. However, some issues still need to be addressed.

MOF synthesis strategy

The hydrothermal method is widely employed for preparing MOF precursors; however, its low yield and high costs pose a barrier to mass production. Therefore, it is urgent to explore a new, cost-effective preparation method capable of achieving mass production (e.g., the microwave-assisted method).

Development of conductive MOF

Low-conductivity ZIFs are often used as MOF precursors, requiring high-temperature carbonization to improve MA performance. However, this can lead to structural collapse, hindering further research. Therefore, it is essential to develop conductive MOFs that can be used directly for absorption.

Table 1. MA performance of MOF-based MAMs

Dimension	Morphology	MAMs	EAB (GHz)	*RL* (dB)	Filling ratio (wt.%)	Refs.
0D	Core-shell	Ni@C@ZnO	4.1	-55.8 (2.5 mm)	25	40
	hollow	Co/C	6.8	-52.66 (2.7 mm)	30	45
1D	Nanotubes	Co/MnO/CNTs	4.5 (1.32 mm)	-58.0	35	46
2D	Nanosheets	N-GN	-	-54 (2.1 mm)	3	47
3D	Flower	NCNT/NiCo/C	4.64	-66.1 (1.5 mm)	15	37
	aerogel	CoNiFe-PBA/GO	6.6	-66.23 (2.6 mm)	1.1	33

MA: Microwave absorption; MOF: Metal-organic framework; MAMs: Microwave absorbing materials; EAB: Effective absorption bandwidth; *RL*: Reflection loss; CNT: Carbon nanotube; N-GN: N-doped porous graphene carbon nanonets; NCNT: N-doped CNT; PBA: Prussian blue analog; GO: Graphene oxide; 0D: Zero-dimensional; 1D: One-dimensional; 2D: Two-dimensional; 3D: Three-dimensional.

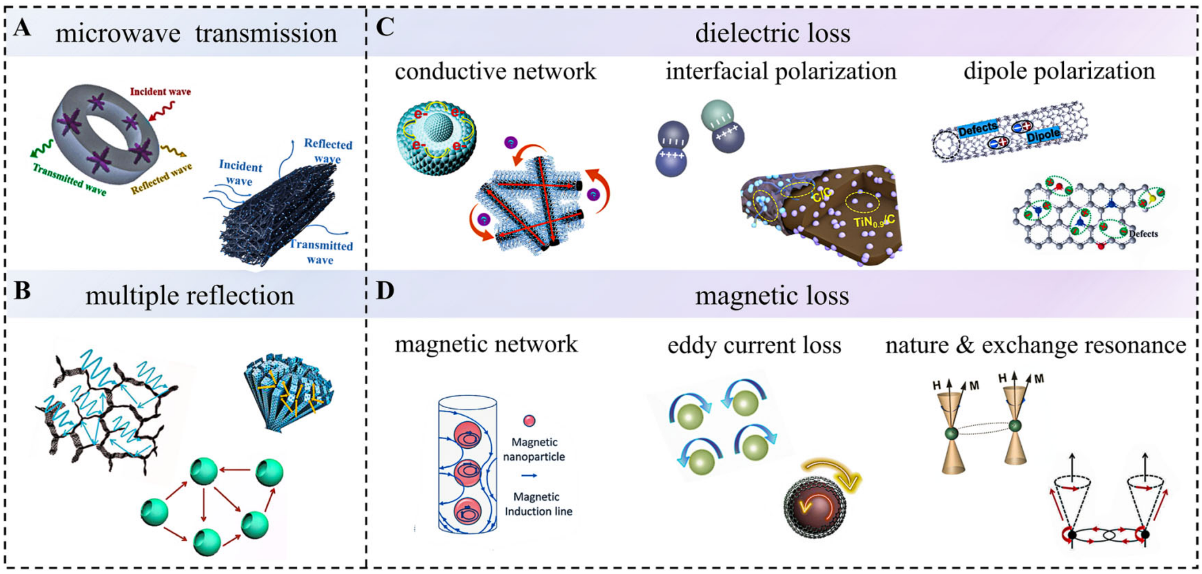

Figure 3. Schematic diagram of the MA mechanism, including (A) microwave transmission. Reproduced with permission[48,49]; (B) multiple reflection. Reproduced with permission[38,51,52]; (C) dielectric loss. Reproduced with permission[40,43,44,53,55,57]; (D) magnetic loss. Reproduced with permission[35,39,55,57,62]. MA: Microwave absorption.

Introduction of other group metal elements in MAMs

The metals introduced into MOF-derived MAMs mainly focus on transition metals, such as Mn, Ni, Fe, *etc*. However, some elements from other families also have the same chemical properties, such as rare earth elements (RE), which can also bond with organic ligands to form RE-MOFs. Developing MAMs containing metals from different families is a promising direction for advancement.

Research on the mechanism of microwave absorption

Most electromagnetic functional materials are complex systems, and their MA capability depends on the synergistic effects of multiple mechanisms. However, insufficient theories to quantify each mechanism's contribution to MA capacity complicate material design. Additionally, the relationship between microstructure and MA mechanisms requires further exploration.

Simulation and customization of new structural MOFs

Because of the limitations of physical mechanisms, the relationship between MAMs and their properties can only be validated through lengthy and inefficient duplicate experiments. To improve efficiency, researchers can customize the composition and structure of MOFs using advanced computer simulations for precise manipulation of electromagnetic properties.

In summary, research on MOF-derived MAMs has considerable potential for advancement. Researchers can explore efficient experimental methods, new MAMs and microwave mechanisms to facilitate the development of multifunctional materials and electromagnetic devices by scaling up experiments and applying computer technology.

DECLARATIONS

Authors' contributions

Substantial contributions to conception and design of the study and data analysis and interpretation: Fan XX

Data acquisition and administrative, technical, and material support: Zhang XC, Li L, Cao MS

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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