

Electrically Insulating Electromagnetic Interference Shielding Materials: A Perspective

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Electromagnetic interference (EMI) shielding is critical in electronic applications. Over the past decades, numerous high-performance EMI shields are developed using electrically conducting materials. However, from an application aspect, EMI shielding materials, integral to the increasingly complex electronic systems, often need to be electrically insulating to prevent short-circuiting and enhance overall application reliability and flexibility. This requirement has frequently been overlooked in earlier research on EMI shielding materials. This paper summarizes the representative advancements in the design of innovative EMI shielding materials that minimize and eliminate reliance on electrical conductivity. Insights into the future prospects and directions for the development of electrically insulating EMI shielding materials are also provided, with the aim of pioneering materials that are both intrinsically insulating and effective at EMI shielding.

compositional or structural designs.^[4–11] On the other hand, it is important to note that the traditional design of EMI shielding materials primarily relies on the principle of the Faraday cage, which requires high electrical conductivity and sufficient free electric charges in EMI shielding materials. Metallic materials, characterized by their dense free electrons and ultrahigh electrical conductivity, thus offer superior EMI shielding efficiency (SE) and are the most commonly used materials in creating EMI shielding solutions.^[12] In exploring alternative synthesized EMI shielding materials, research efforts are also primarily concentrated on enhancing the electrical conductivity of materials, as this is universally acknowledged as the most effective strategy for boosting the EMI

shielding performance.^[13] However, given that EMI shielding materials are typically employed to safeguard electronic components, many applications also require these materials to be electrically insulating to prevent short-circuiting. This engineering requirement has often been overlooked in earlier research on EMI shielding materials.

It is commonly believed, often subconsciously, that EMI shielding materials must be electrically conductive, given that their shielding performance generally improves with increased electrical conductivity. Consequently, developing intrinsically insulating EMI shielding materials has long been considered highly challenging and largely unexplored. Traditional methods for achieving electrical insulation in EMI shielding materials generally involve applying external insulating coatings to conductive EMI shielding materials and creating multilayer structures, which still rely on the construction of highly conducting shielding layers within the materials. However, it is also often overlooked that while increased electrical conductivity does enhance EMI shielding capability, it is not the only scientific criterion for EMI shielding.^[12] In other words, effective EMI shielding does not necessarily require the connectivity of the conducting network within the materials. With this insight, some pioneering efforts have been directed toward the development of less conductive and intrinsically electrically insulating materials for EMI shielding (Figure 1). Initially explored for their high processability and tunability, composites containing electrically-isolated fillers were designed to achieve effective EMI shielding through the interaction of electromagnetic waves with the dispersed individual fillers, coupled with the scattering effect.^[14–16] Subsequently, inspired by the design concepts

1. Introduction

The rapid evolution of electronic and communication technologies has significantly heightened the complexity of electromagnetic environments, consequently escalating the risk of device malfunctions.^[1] As a result, there is a widespread need for effective electromagnetic interference (EMI) shielding materials to address the adverse effects of EMI, guaranteeing the stable functioning of devices.^[2,3] Over recent decades, a variety of high-performance EMI shielding materials have been developed, evolving from initial metallic materials to carbon-based substances, and subsequently to innovative nanomaterials and functional composite materials, achieving high EMI shielding performance and additional beneficial functions through thoughtful

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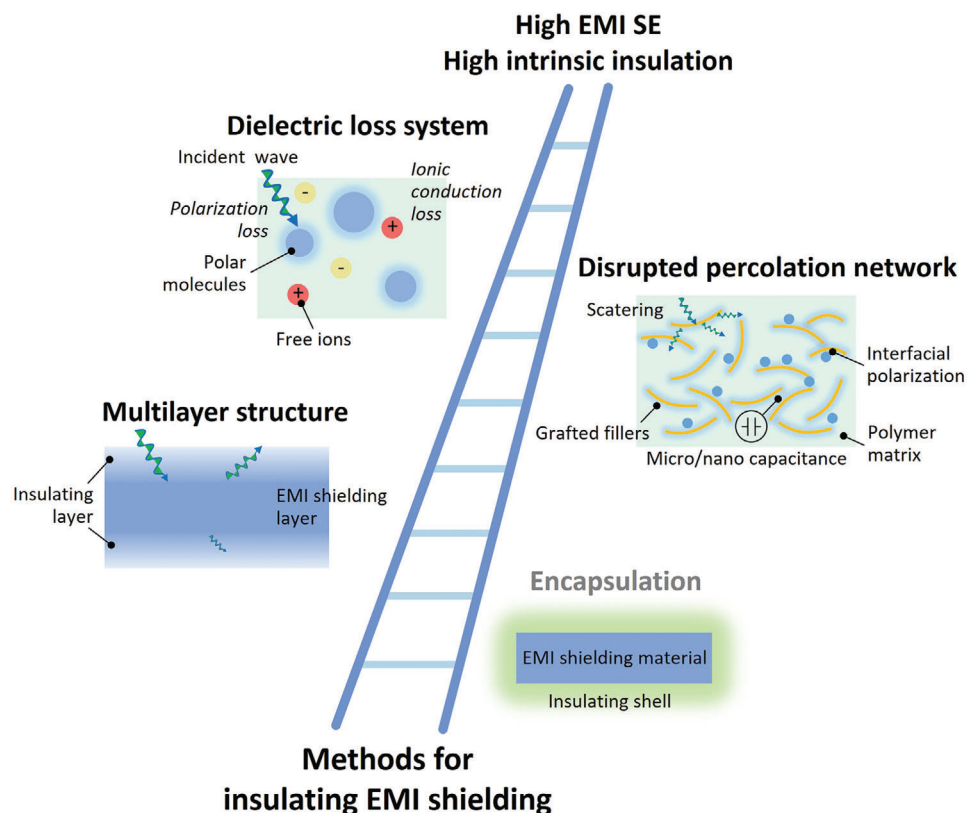


Figure 1. Schematic illustrating the reported strategies for insulating EMI shielding.

of microwave absorbers, dielectric systems (e.g., EMI shielding materials containing polar molecules) were developed. These systems demonstrate strong electromagnetic wave attenuation capability through non-conduction loss mechanisms, such as interfacial, dipolar, and defect-induced polarization losses, effectively minimizing transmission.^[17,18] These preliminary advancements have offered both inspiration and new possibilities for further research into intrinsically insulating EMI shielding materials.

In this perspective, we begin by providing a critical overview of the most significant advancements in the design of EMI shielding materials that aim to minimize and eliminate reliance on electrical conductivity. Then, we shift our focus to highlighting the potential strategies and viable solutions for the development of intrinsically insulating EMI shielding materials. Additionally, the significant problems and challenges currently confronting the field will be discussed.

2. Methods for Creating Electrically Insulating Electromagnetic Interference Shielding Materials

2.1. Encapsulation using Insulating Materials

Encapsulating electrically conductive EMI shielding materials with electrically insulating materials, such as plastics, rubber and silicone, is the most commonly used method for insulating EMI shielding materials. This method is simple, flexible and scalable, allowing for adjustments in the types of insulating encapsulation

and shielding materials to achieve the desired insulation and EMI shielding levels for various working environments. However, this method still has some inherent limitations. For example, the outside insulating polymer materials may age, harden, or become brittle over time, which can lead to a decline in insulation performance, potentially increasing the risk of short-circuiting or electrical faults. In addition, since the EMI shielding materials and the external insulating layers cannot form an integrated structure, cracks or damages can easily occur between them when subjected to excessive mechanical stress or impact, leading to a failure in insulation. Therefore, it is still imperative to develop EMI shielding materials that possess intrinsic insulation properties.

The design of multilayer structures shares principles with the creation of external insulation layers.^[19] In a multilayer structure, the EMI shielding layers are sandwiched between insulating layers, ensuring effective electrical insulation along the *z* direction.^[20] This method demonstrates greater compatibility with polymer nanocomposites. Notably, within multi-layer polymer nanocomposites, the EMI shielding layers and the insulating layers can share the same polymer matrix, and gradient structure can also be formed.^[21,22] This feature facilitates seamless interfaces between the layers, resulting in superior structural integrity and stability. Meanwhile, the number of layers, the composition and thickness of each layer can all be rationally designed and tailored for efficient performance regulation and optimization. For instance, Liu et al. developed a multilayer EMI shielding film via a repeated spin coating process, in which the conductive MXene/polydimethylsiloxane (PDMS) EMI shielding layers and the

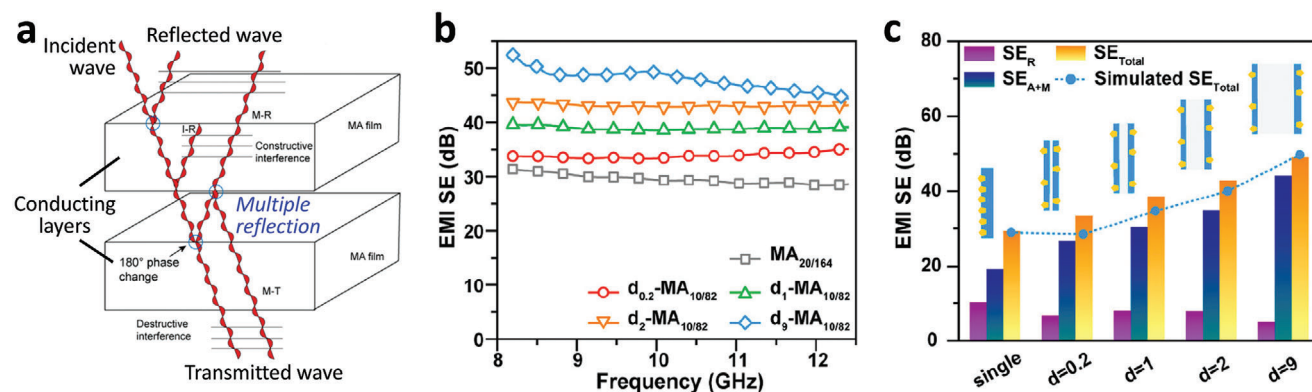


Figure 2. (a) Schematic illustration of microwave interference mechanisms of the multilayer EMI shielding films. (b) EMI shielding performances of multilayer films with various gap distances. (c) Experimental and simulated SE values for homogeneous film and multilayer films with various gap distances. Reproduced with permission.^[24] Copyright 2020, American Chemical Society.

insulating yet thermally conductive boron nitride (BN)/PDMS layers are sequentially deposited. After curing, these different PDMS-based layers can be tightly bonded together to form an integrated EMI shielding structure. A 11-layers film with a thickness of 1.2 mm can possess an EMI SE of 35.2 dB, an ultrahigh volume resistivity of $2.9 \times 10^{12} \Omega \text{ cm}$, and a breakdown strength of 3.29 kV mm^{-1} .^[23] It is also worth noting that a layered structure design can optimize the EMI shielding performance without additional loading of conducting components (Figure 2).^[24] This effect is due to the transmitted microwaves being repeatedly reflected between the conductive layers, leading to more effective attenuation. When the spacing of the conductive layers is an integer multiple of quarter of the wavelength, optimal EMI shielding performance can be achieved. This work provides a valuable guideline for designing high-performance insulating EMI shielding materials based on multilayer structures. Despite the effectiveness, adding external insulating phase will inevitably result in a thicker EMI shielding material, which may lead to issues related to the additional physical space occupied and may also affect the mechanical flexibility and weight. These factors are critical considerations for applications such as aerospace and wearable electronics. In addition, the shielding capability of multilayer structures still relies on the presence of continuous conducting layers, which limits their in-plane insulation properties.

2.2. Design of Dielectric Systems

EMI shielding aims to minimize the transmission of incident electromagnetic waves, which can be enabled through either reflection or absorption.^[1] The design of EMI shielding materials with high conductivity primarily contributes to reflection. However, if a material can effectively absorb the incident radiation through multiple attenuation mechanisms to minimize transmission, high EMI SE can also be achieved. Dielectric loss represents one kind of the most effective attenuation mechanisms, which depends on not only the conductive properties but also the polarity of the EMI shielding materials.^[25] For example, in a polar system, numerous permanent dipoles exist due to the substantial difference in electronegativity between two atoms in the polar molecules. This difference causes one atom to attract elec-

trons more strongly, becoming more negatively charged, while the other becomes more positively charged. Under an alternating electromagnetic field, the dipoles will absorb energy to reorient themselves toward the direction of the electric fields, a process known as dipole polarization. When the rotation of these dipoles cannot keep pace with the changes in the electric fields, polarization hysteresis occurs, leading to attenuation of the electromagnetic field. Therefore, the absorption of radiation can be facilitated through various polarization-based mechanisms including dipole polarization, interfacial polarization, defect-induced polarization, and ionic polarization (Figure 3a).^[26]

Highly polar molecules, such as water, have been demonstrated to be capable of effectively attenuating electromagnetic waves due to the high static dielectric constant and strong polarization loss capability.^[17,27] For example, by incorporating abundant high-polarity water molecules into polymer networks, gel-type materials have been developed for EMI shielding applications.^[28–35] The polar water molecules and hydrogen-bonded polymer networks within the composite systems can be polarized under the incident electromagnetic field and attenuate its energy through polarization losses.^[1] Therefore, even with poorly connected conducting networks and low electrical conductivities, these gel-type materials can still exhibit good EMI shielding performance through an absorption mechanism. These efforts have highlighted a viable direction for further developing electrically insulating EMI shielding materials through the design of dielectric systems. More recently, Wu et al. conducted a comprehensive study on the polarization loss-dominated EMI shielding materials (Figure 3b).^[17] A series of gel-based systems with single electromagnetic mechanism dependence was cleverly designed, ensuring high polarity correlation while avoiding the occurrence of conduction-related properties. It was demonstrated that polar liquids (such as water and water/DMSO mixture) immobilized in inert polymer networks and the existing non-covalent bonding can provide strong polarization loss to attenuate incident electromagnetic waves. These interactions dramatically affect the dielectric properties and relaxation time of the gels by changing the short-range alignment and orientation barriers of the molecules, leading to an EMI SE of $\approx 20 \text{ dB}$ for the resulting hydrogels (Figure 3c,d). The diversity of polar molecules also makes it possible to build a

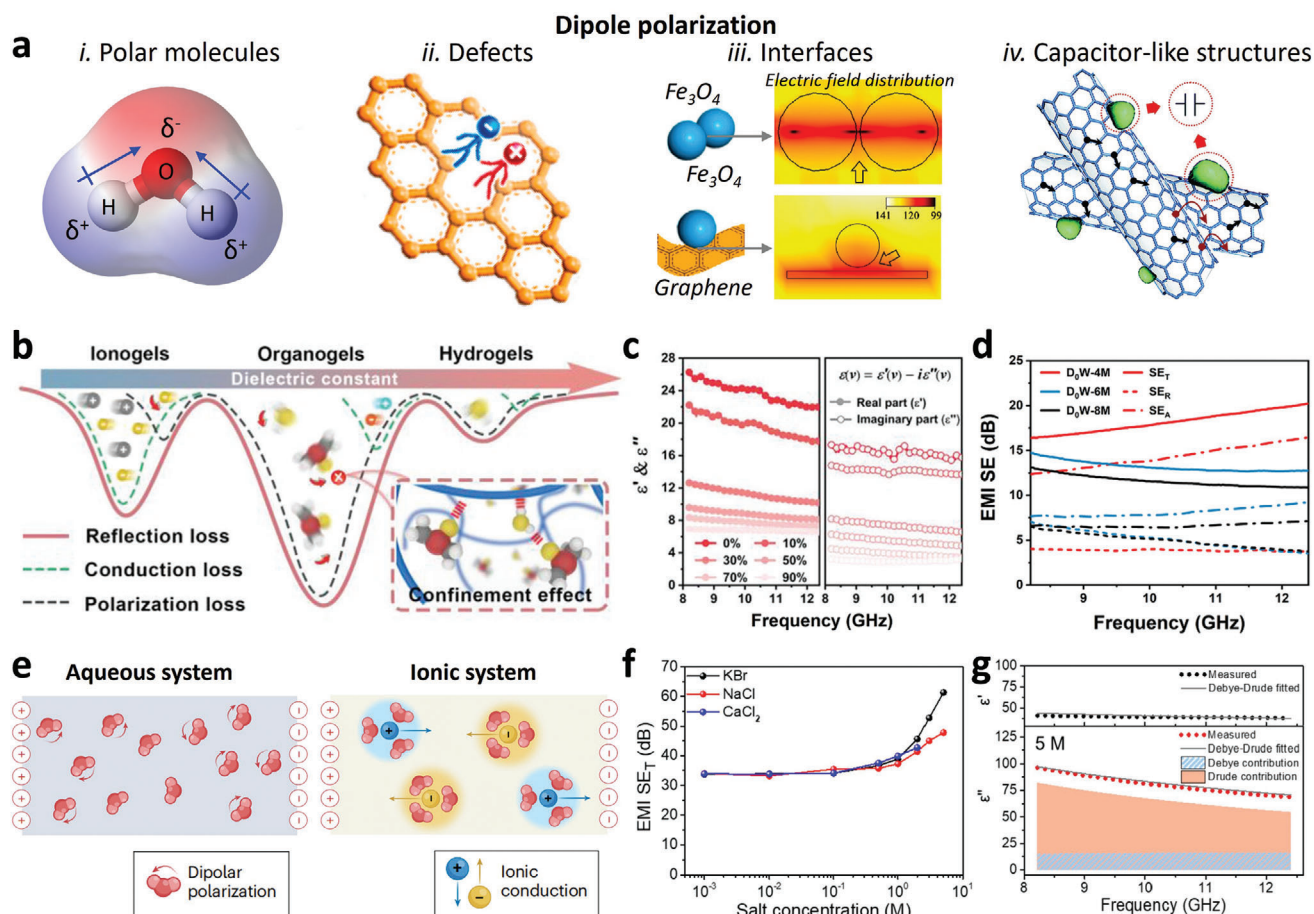


Figure 3. (a) Schematic illustrating showing models for dipole polarization: i. Polar molecules. ii. Defects and iii. Interfaces. Reproduced with permission.^[36] Copyright 2021, Elsevier. iv. Capacitor-like structures. Reproduced with permission.^[37] Copyright 2014, Royal Society of Chemistry. (b) Schematic diagram of the attenuation characteristics of gel-based systems. (c) Complex permittivity of hydro/organogels. (d) EMI shielding performance of water-containing polyacrylamide hydrogels. Reproduced with permission.^[17] Copyright 2022, Wiley-VCH. (e) EMI shielding mechanisms for aqueous (left) and ionic (right) solutions via dielectric polarization of water molecules and ionic conduction of free ions, respectively. (f) EMI SE of 5-mm-thick KBr, NaCl, and CaCl_2 ionic solutions with different concentrations. (g) Experimentally obtained and theoretically calculated real and imaginary components of permittivity of 5 M KBr solution. Reproduced with permission.^[38] Copyright 2022, Elsevier.

dielectric database for further designing more efficient gel-based EMI shields. This study initially provides insightful guidelines for precisely engineering electromagnetic functional materials with specifically designed electromagnetic mechanisms and opens up new possibilities for developing insulating EMI shielding materials based on polar liquid-containing systems.

In another experimental work, Koo et al. proposed the use of ionically conductive aqueous ionic solutions of alkali-metal halide salts (e.g., KBr, NaCl, and CaCl_2) as effective EMI shielding materials (Figure 3e).^[38] It was demonstrated that adding salts into water will reduce the dielectric constant (real part of the permittivity) but can increase the dielectric loss (imaginary part of the permittivity, which relates to the attenuation ability). These ionic solutions can attenuate the energy of incident radiation through orientational polarization of water molecules and ionic conduction of mobile ions, showing a distinct EMI shielding mechanism compared to conventional solid materials. As a result, the EMI shielding performance can be easily modulated and enhanced by adjusting the salt concentration and the thick-

ness of the solution shield, reaching an unprecedented value of 61.3 dB for 5 M KBr ionic solution at a thickness of 5 mm (Figure 3f,g). It was demonstrated that a low salt concentration results in poor ionic conductivities, which limits its contribution to enhancing EMI shielding performance through ionic conduction loss. Thus, to achieve an enhanced EMI SE, the ion concentration and ionic conductivity should be kept at a sufficient level. However, it must be recognized that the ion concentration is also directly relevant to the electrical conductivity of the salt solution. Higher ion concentration generally leads to higher electrical conductivity because more ions are available to carry electric charge, facilitating a greater flow of current through the solution. Thus, a more systematic study is still needed to elucidate the relationship between the ion concentration, EMI shielding performance, and electrical conductivity of the solution-based shields, in order to effectively manage the trade-offs.

Despite their effectiveness, current EMI shielding materials based on polar molecules and liquids still encounter significant challenges, such as the evaporation of liquids, poor

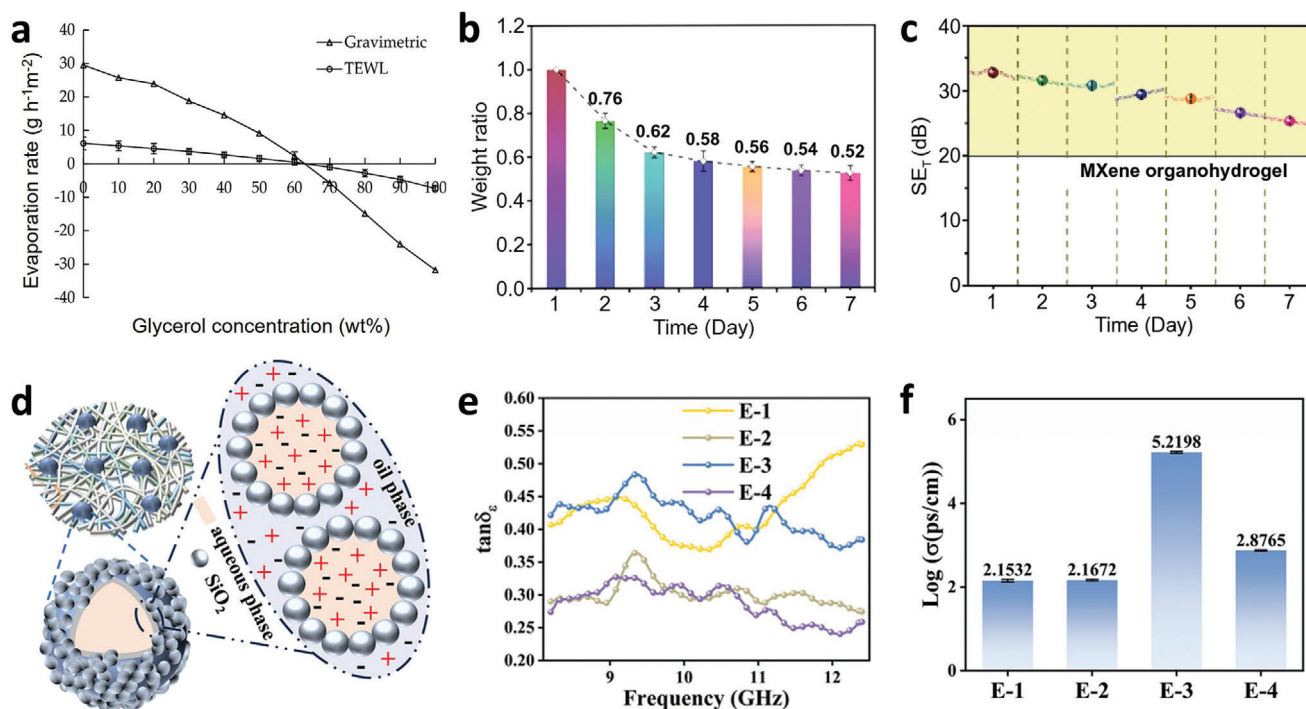


Figure 4. (a) Evaporation rate of glycerol solutions of various concentrations. Reproduced with permission.^[40] Copyright 2022, Springer Nature. Changes in (b) weight ratio and (c) EMI shielding performance of MXene organohydrogel over a 7-day storage period under environmental conditions of 25 °C and 45% humidity. Reproduced with permission.^[31] Copyright 2022, Springer Nature. (d) Schematic of the liquid marble microarchitectures. (e) The dielectric loss tangent and (f) electrical conductivity of the composites. Reproduced with permission.^[18] Copyright 2024, Wiley-VCH.

low-temperature tolerance, and compatibility issues with electronic systems. To address these challenges, strategies such as developing binary liquid systems have been proposed to enhance the anti-drying capability and stability of gel-type shields. For instance, partially replacing the water with glycerol in the MXene composite hydrogel can significantly reduce water evaporation and enhance performance stability (Figure 4a–c).^[31] It should also be noted that while ideal polar liquids, such as pure water, are excellent insulators, the presence of dissolved impurities can exponentially increase their electrical conductivity.^[30,39] In a recent study, Wu et al. proposed an innovative microstructural engineering strategy to endow designed composite materials containing polar liquids with remarkable stability, effectively addressing the aforementioned challenges (Figure 4d).^[18] They found that polar liquid droplets, specifically a NaCl solution, could be stabilized by encapsulating them in a protective nano-SiO₂ layer through a self-assembly process, resulting in the formation of liquid marble microarchitectures. These robust marble microarchitectures containing high-concentration salt solutions can be seamlessly integrated into different polymer matrices, significantly enhancing polarization loss to effectively attenuate electromagnetic waves (Figure 4e). Additionally, the nano-SiO₂ layer restricts the mobility of Na⁺ and Cl⁻ ions, preventing the formation of an excessive ionic network within the composite, leading to ultrahigh electrical resistivity ranging from 10⁷ to 10¹⁰ Ω cm (Figure 4f). Although this study concentrated on investigating electromagnetic wave absorption properties of the designed composites, it provides valuable guidelines for designing electrically insulating EMI shielding materials based on dielectric

loss mechanisms. It is anticipated that by further refining the structural design, adjusting the composition, controlling the distribution of polar components, and including synergistic wave attenuation effects, it will be possible to achieve ultrahigh electrical resistivity while enhancing electromagnetic wave dissipation, thereby achieving more efficient electrically insulating EMI shielding. Nevertheless, further advancements are still necessary to develop more stable dielectric composite systems containing polar molecules, thereby broadening their potential for practical applications.

Conduction loss is another typical dielectric loss mechanism, which generally exists in highly conductive materials. When electromagnetic waves penetrate conductive materials, the electric field induces the movement of free electrons or other charge carriers, generating microscopic currents. These currents encounter the inherent resistance of the material as they flow, converting electromagnetic energy into thermal energy. In composite materials, electrons can either migrate within a single conducting filler or jump across defects or interfaces between fillers, identified as migrating and hopping models, respectively.^[26,41] Thus, the existence of non-continuous conducting network within an insulating EMI shielding material would limit the capacity for conductive loss dissipation due to the high energy barrier for the electron hopping process.^[18,26] While high electrical conductivity is crucial for significant conduction losses, it is important to recognize that the formation of a localized microscopic conductive network within the material may still facilitate conductive losses, even if the material demonstrates ultralow conductivity at a macroscopic level.

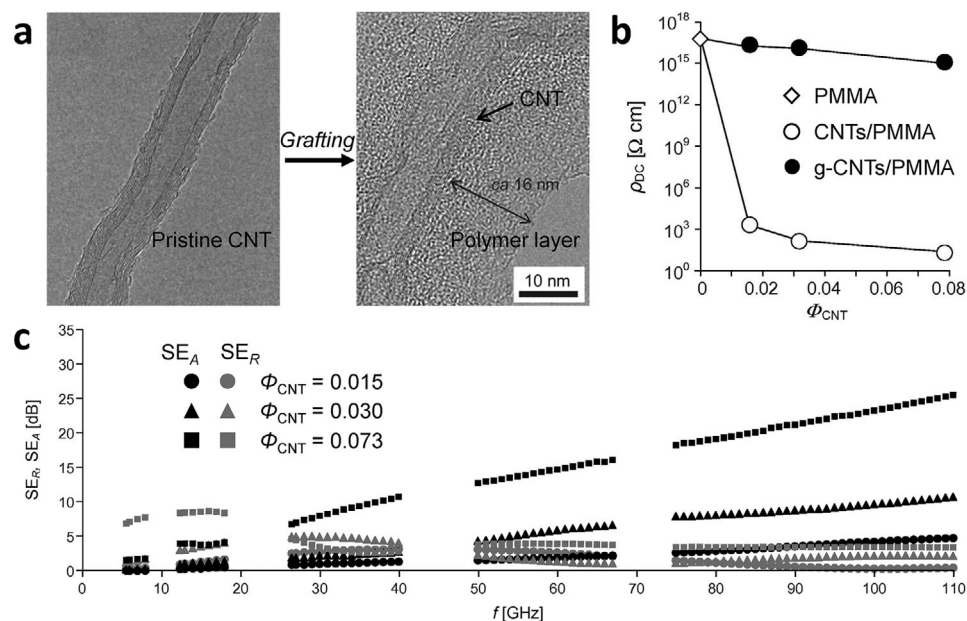


Figure 5. (a) TEM images of the pristine CNT and polymer-grafted CNT. Reproduced with permission.^[48] Copyright 2012, Wiley-VCH. (b) Electrical resistivity of PMMA, CNTs/PMMA composite, and g-CNTs/PMMA composite with disrupted CNTs network. (c) EMI shielding performance of g-CNTs/PMMA composite. Reproduced with permission.^[14] Copyright 2015, Elsevier.

2.3. Design of Composites with Disrupted Percolation Networks

Composite materials have shown great potential in EMI shielding applications due to their light weight, corrosion resistance, and highly tunable composition, structure and properties.^[42,43] Generally, EMI shielding relies primarily on a reflection mechanism, which requires the EMI shielding materials to possess sufficient mobile charge carriers for effective interaction with the electromagnetic waves.^[12] According to Simon's formula ($EMI\ SE = 50 + 10\log(\sigma/f) + 1.7d(\sigma/f)^{0.5}$,^[44] where f is the frequency, σ and d is the electrical conductivity and the thickness of the shield, respectively), the EMI shielding performance of a composite material monotonically increases with the increase in its electrical conductivity. Therefore, previous research on EMI shielding composite materials has primarily focused on improving the continuity of conducting fillers and lowering the percolation thresholds, aiming to achieve improved EMI SE with reduced filler content and decreased thickness.^[45,46] However, it has often been overlooked that while electrical conductivity does enhance EMI SE, it is not the sole scientific criterion for shielding, and effective EMI shielding in composites does not necessarily require the connectivity of the fillers.^[12] When a continuous conductive network is absent in the EMI shielding composite material, the mobile electrons and dipoles within the conducting filler and the existing interfaces can also interact with the electromagnetic waves to a certain extent.^[12,25,47]

In a pioneering work, Hayashida et al. designed an isolated filler network in polymer/carbon nanotubes (CNTs) composites.^[14] In this design, the CNTs are individually grafted with poly(methyl methacrylate) (PMMA) chains through a surface-initiated atom transfer radical polymerization process (Figure 5a). The grafted PMMA effectively separate the individual CNTs by a sufficient distance (>10 nm) within the polymer matrix

to prevent tunneling conduction, thereby maintaining the high electrical resistance of the composites (Figure 5b).^[48] This occurs even when the CNTs loading surpasses the percolation thresholds typical for conventional CNTs/PMMA composites, indicating the formation of a disrupted percolation network. Meanwhile, the separated CNTs in the composite can still reflect and absorb the electromagnetic waves in the frequency range of 1–110 GHz through electron-electric field interaction and interfacial polarization, showing a maximum EMI SE of 29 dB at 110 GHz (Figure 5c). It is worth noting that the composite demonstrates a significant absorption behavior within higher frequency ranges, which indicates the minimal contribution of reflection. It was also proposed that reducing the electron mobility in the filler to levels significantly lower than the electric field of the electromagnetic wave could further enhance the dielectric loss capability, thereby contributing to the overall EMI shielding performance. However, in this study, the observed EMI SE still falls within a relatively low range, which should be attributed to two main factors: 1) the polarization loss provided by the defect-free carbon nanotubes is insufficient, and 2) as 1D fillers, the small size of CNTs limits their capacity to effectively scatter electromagnetic waves, hindering the attenuation capability. Although outstanding EMI SE was not obtained, this work innovatively provides a new concept for EMI shielding composites: the compatibility of EMI shielding performance and electrical insulation can be achieved by disrupting the percolation network of fillers. It is worth noting that the conducting fillers can also be isolated using a variety of functional yet insulating components, not limited to polymers. For instance, Gu et al. suggested that depositing magnetic nanoparticles (such as Fe_2O_3 , Fe_3O_4 , and $NiFe_2O_4$) with the effect of attenuating electromagnetic waves on electrically conductive fillers can enhance the EMI shielding performance and also facilitate insulation modification.^[19]

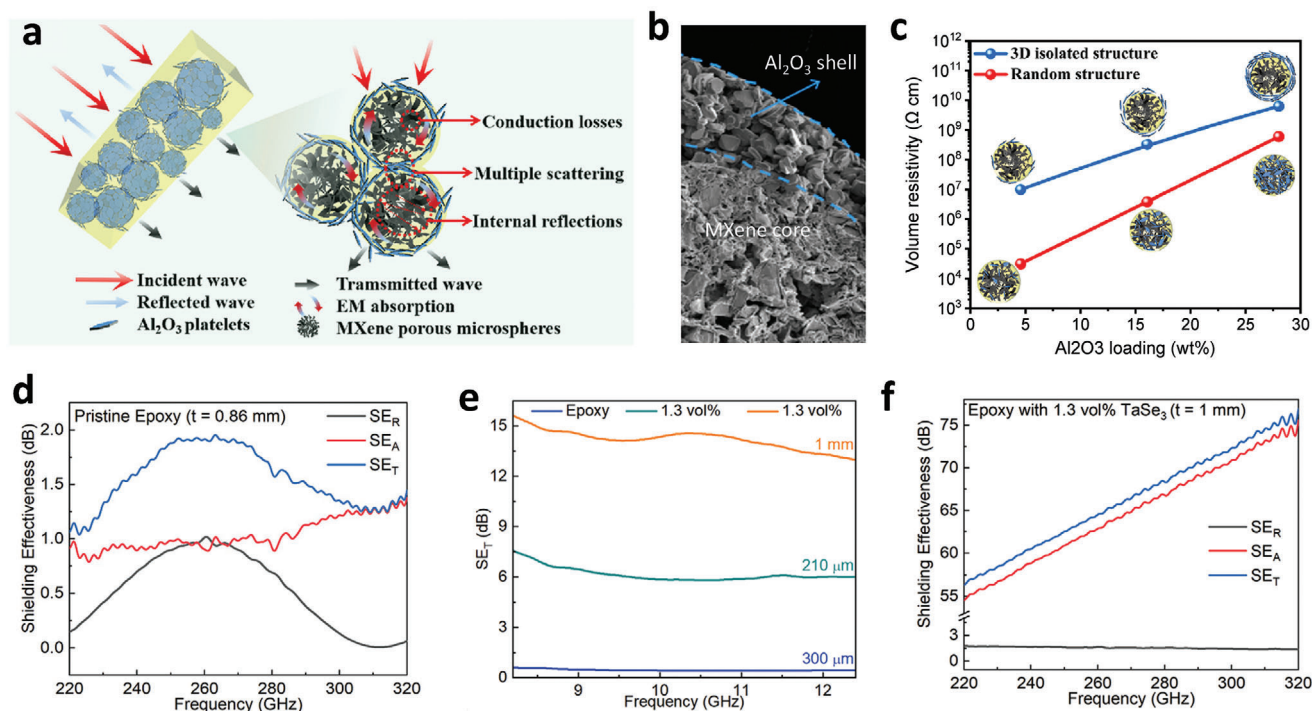


Figure 6. (a) Schematic showing the working mechanism of the MXene/Al₂O₃/epoxy composite with 3D isolated structure. (b) SEM of the Al₂O₃-isolated MXene filler. (c) Electrical resistivity of the obtained MXene/Al₂O₃/epoxy composite. Reproduced with permission.^[49] Copyright 2024, Elsevier. (d) EMI SE of pristine epoxy. (e,f) EMI shielding performance of epoxy/TaSe₃ composite. Reproduced with permission.^[16] Copyright 2021, Wiley-VCH.

Following the design concept described above, recent developments have led to the creation of several electrically insulating composites with enhanced EMI shielding performance. For example, an isolation filler network was created by combining MXene aerogel microspheres with Al₂O₃ platelets, in which the conducting MXene porous microspheres are isolated by the Al₂O₃ platelets. After the filler skeleton was impregnated with epoxy resin, only localized conducting region for EMI shielding can be formed within the fabricated composites. With such isolated filler network, the resulting composites exhibit an ultrahigh electrical resistivity of $1.2 \times 10^9 \Omega \text{ cm}$, a high thermal conductivity of 2.1 W mK^{-1} , and an EMI SE of 22.3 dB at 8.6 GHz (Figure 6a–c).^[49] In another study, Balandin et al. used the high-aspect-ratio quasi-1D van der Waals materials like TaSe₃ nanowires as functional fillers to fabricate insulating EMI shielding composites.^[21] By keeping the filler content below the percolation threshold, the electrical resistivity of the composites can be maintained at levels higher than $10^7 \Omega \text{ cm}$. Meanwhile, the composites still present a moderate EMI shielding performance in the gigahertz range (8.2–12.4 GHz). Specifically, the UV-light-cured polymer/TaSe₃ composite exhibits an EMI SE of $\sim 10 \text{ dB}$, while the epoxy/TaSe₃ and sodium alginate/TaSe₃ composites show an EMI SE of $\sim 15 \text{ dB}$ (Figure 6d,e). The EMI shielding performance further improves with increasing frequency, achieving levels between 55 and 75 dB as the frequency ranges from 220 to 320 GHz (Figure 6f). While the authors attributed the X-band EMI shielding performance to interactions involving electric field-free carriers and the ultrahigh sub-terahertz shielding performance to a scattering effect, the detailed EMI shielding

mechanisms are still not thoroughly elucidated. Further research is crucial for an understanding of the electrical and magnetic properties of the quasi-1D van der Waals TaSe₃ materials, as well as for developing methods to control their size. This knowledge will help further reveal the underlying EMI shielding mechanisms and enable improvements in the shielding performance of the composites.

3. Perspectives

EMI shielding materials are widely used as a key component in electronic systems. While high electrical conductivity is beneficial for EMI shielding performance, it can also potentially cause short circuits between components, leading to equipment damage. Particularly in modern electronic devices, which often feature high power and complex designs (e.g., high-density component layouts), insulation design of EMI shielding materials becomes increasingly important and challenging. Therefore, efficient EMI shielding materials with high intrinsic insulation can undoubtedly provide more reliable electrical isolation between components, ensuring greater application flexibility and safer operation when used within complex electronic systems.

Although recent years have witnessed the development of less conductive and insulating EMI shielding materials, research into intrinsically insulating EMI shielding materials is still in the initial stages. Several significant challenges confront this field. Achieving optimal electrical insulation while maintaining effective EMI shielding using current strategies is a primary challenge. Most of the reported intrinsically insulating EMI shielding

Table 1. Summarized design strategies and performance of reported less conductive and electrically insulating EMI shielding materials.

Design strategy	Material	Electrical resistivity [Ω cm]	EMI SE [dB]	Thickness [mm]	Features	Refs.
Multilayer structure	TPU/MWCNTs/BN composite	10^{10} - 10^{11}	53.61	2	Advantages: simple; high shielding effectiveness;	[21]
	GnP@PDMS/GF composite	0.35×10^{10}	50.13	/	high insulation	[22]
	(BNNS/ANF)-(AgNWs/ANF) composite	4.7×10^{13}	70	0.095	Current limitations: poor in-plane insulation; potential failure risk	[50]
	BNNS/LMPA/ER composite	10^8 - 10^{11}	14-20	/		[51]
	AlN/BN/MWCNT/PP composite	6.61×10^{13}	27.68	/		[52]
	CF@Fe ₂ O ₃ /(BN/SR) composite	6.2×10^{14}	37.7	2		[19]
	SiR/GNPs/BN composite	10^{12} - 10^{13}	40.67	2		[20]
	MXene/PDMS/BN composite	2.9×10^{12}	35.2	1.241		[23]
	Pure PAM hydrogel	/	≈ 20	/	Advantages: intrinsic insulation; high tunability; isotropic insulation	[17]
Dielectric systems (polar systems)	Pure PVA hydrogel	/	≈ 17	2	Current limitations: limited shielding effectiveness	[29]
	PPy/PEG/PVA hydrogel	1.3×10^5	28	2		[35]
	Pure PVA hydrogel	$\approx 1.3 \times 10^4$	≈ 19	2		[53]
	SA/PAM/PGO hydrogel	5×10^3	21.1	2		[54]
	PVA-PEG-SA hydrogel	1.8×10^5	≈ 21	2		[55]
	PAA hydrogel	/	18.6	1.6		[56]
Disrupted percolation network	PVDF@MWCNT/BN composite	0.12×10^{14}	8.68	2	Advantages: intrinsic high resistivity; low reflection;	[15]
	PMMA-CNT composite	1.3×10^{15}	≈ 11	0.57	isotropic insulation	[14]
	MXene/Al ₂ O ₃ /EP composite	1.2×10^9	22.3	/		[49]
	TaSe ₃ /SA composite	$> 10^7$	15.0	0.077	Current limitations: limited shielding effectiveness	[16]
	TaSe ₃ /UVP composite	$> 10^7$	11.0	0.13		
	TaSe ₃ /epoxy composite	$> 10^7$	≈ 14.0	1		

materials still exhibit relatively low EMI SE (Table 1). For materials possessing extremely high intrinsic resistivity, it is particularly difficult to achieve ultrahigh EMI SE without the contribution of reflection and sufficient conduction loss of electromagnetic waves. Developing EMI shielding materials based on dielectric loss mechanisms has been proven highly effective. However, the electromagnetic energy attenuated by these mechanisms is typically converted into heat, and the insulating designs, such as creating external insulating coatings and disrupting the filler network, can sometimes impede heat dissipation. This issue can be especially problematic in high-power electronic applications, where the accumulated heat can severely affect device performance. Durability and stability are additional key concerns, since EMI shielding materials must be capable of withstanding diverse environmental conditions, such as moisture exposure, temperature fluctuations, and mechanical stress, which can potentially

degrade the insulating and EMI shielding properties over time. At present, most existing strategies still struggle to meet the complex demands of real-world applications. Moreover, the production of insulating EMI shielding materials with intricately designed compositions and microstructures presents challenges in maintaining consistent performance and scalability, which underscores the need for advancements in manufacturing technology to deliver cost-effective, scalable, and reproducible methods. Innovating and exploring new ideas to address these challenges are still highly needed to further the development of next-generation EMI shielding materials.

Currently, the future of electrically insulating EMI shielding materials remains uncertain, with no significant breakthroughs achieved in this area so far. Nevertheless, this field still holds substantial opportunities. Continued in-depth research could also contribute to a deeper understanding of how compositional and

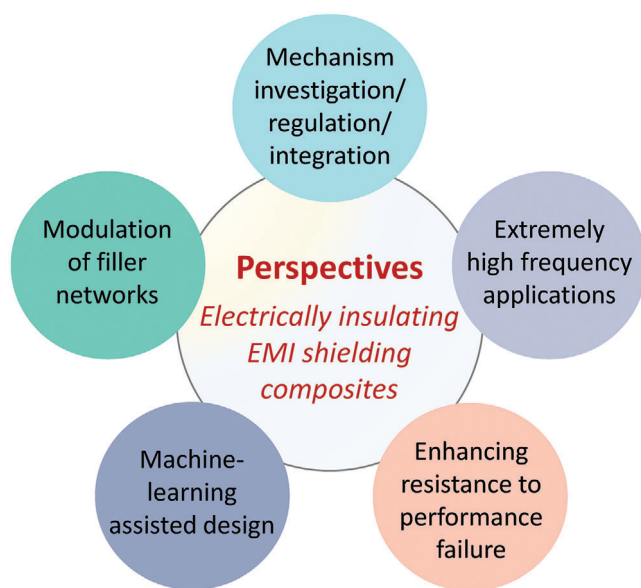


Figure 7. Outlooks toward achieving electrically insulating EMI shielding.

structural designs influence interactions between the materials and electromagnetic waves, facilitating the design of innovative high-performance EMI shielding materials. It is particularly noteworthy that polymer composites may offer unique advantages in achieving effective insulating electromagnetic shielding due to their high processability and customizable composition, structure, and properties. Here, we have several perspectives to share on designing electrically insulating EMI shielding composite materials (Figure 7).

1) The conducting behaviors of composites can be significantly affected by the orientation of the conducting fillers/components within the polymer matrix. Thus, composites containing highly aligned fillers can be rationally designed and fabricated to exhibit anisotropic electrical properties, enabling high intrinsic electrical insulation along a specific direction. It has been demonstrated that infiltrating well-aligned wood vessels with melted metals can result in a composite that exhibits distinct anisotropic conducting properties.^[57] Perpendicular to the vessel direction, the electrical conduction can be largely inhibited due to the lack of contact between the parallel metal fillers, leading to a conductivity as low as $4.8 \times 10^{-7} \text{ S m}^{-1}$. Meanwhile, the metal filler along the vessel direction provides a continuous electron pathway to ensure a high conductivity of $5.4 \times 10^4 \text{ S m}^{-1}$, which is sufficient for effective EMI shielding. However, to date, there has been limited research on utilizing such materials for insulating EMI shielding. In addition, conducting filler networks with high anisotropy can also be constructed using the ice template-assisted assembling processes.^[58] For instance, the bidirectional freeze-casting method has proven effective in creating long-range ordered (centimeter level) lamellar-structured filler networks, which could lead to distinct anisotropic conducting properties.^[59–61] However, significant efforts are still required to further enhance the degree of anisotropy in the constructed anisotropic filler

networks, which is crucial for achieving high insulation along the perpendicular direction, although this presents considerable challenges.

- 2) Another possible solution is to use ink-based 3D printing technique combined with freeze/supercritical-drying treatments to create cost-effective porous filler networks. To date, there have been tremendous successful examples in processing various candidate functional fillers (e.g., graphene, CNTs, MXenes, and metal nanomaterials for EMI shielding, boron nitride and other ceramic fillers for electrical insulation) into printable inks, which can be used to print 3D filler networks via the ink-extrusion 3D printing processes and elaborately designed post-treatments.^[62–65] The printed networks can exhibit predesigned structures due to the high manufacturing flexibility of 3D printing technique. For example, sandwiched filler networks, which consist of a core EMI shielding layer and external insulating and functional layers, can be created using multi-material 3D printing. The layers can be partially merged during the printing process, ensuring high structural integrity of the multilayer filler network. After infiltration with polymers, it is expected that EMI shielding composites exhibiting the desired conducting/insulating properties can be created. In addition, by fine-tuning the ink rheology, more complex filler networks can be printed, enabling precise control over electron transfer and EMI shielding behaviors within the resulting composites.
- 3) Most of the previously reported strategies for the electrically insulating EMI shielding have typically focused on a single working mechanism, which often results in limited attenuation capability and moderate EMI SE. It is thus necessary to further enrich the non-conductivity-dependent and synergistic attenuation mechanisms in a single EMI shielding material to enhance the ultimate EMI shielding performance while retaining the high insulation. For example, the disrupted percolation networks can be introduced into a polar composite system, which may enable a synergistic effect of multiple dielectric loss mechanisms to attenuate the incident radiation more efficiently, thereby enhancing the overall EMI SE. Other possible approaches include creating heterogeneous interfaces, doping fillers with heteroatoms, and introducing polar functional groups and defects, all of which can enhance dielectric losses to more effectively attenuate electromagnetic energy.^[66,67] For some fillers like MXenes, their defective features, surface moieties, electronic conductive properties, and interactions with the polymer matrices can all be simultaneously adjusted, which may open new opportunities for designing more efficient disrupted percolation networks within EMI shielding composites.

In fact, the design of dielectric systems follows a similar principle to that of designing electromagnetic wave absorption materials, which also require appropriate insulation characteristics for impedance matching at the incident surface and strong wave attenuation capability. Therefore, many documented design strategies for absorbing materials can offer valuable insights for the development of intrinsically insulating EMI shielding materials.^[26,68–72] For instance, depositing magnetic particles on the surfaces of conductive components within the composite can impede electron hopping, thereby reducing conductivity. Simultaneously, the presence of

magnetic particles facilitates magnetic loss, significantly enhancing the composite's capability to attenuate electromagnetic waves via a synergistic working mechanism.^[73]

In addition, more efforts are needed to enhance the understanding of the composition-structure-property relationships and the working mechanism of designed EMI shielding systems. Current explanations of EMI shielding mechanisms are largely based on empirical observations. Identifying the primary contributions of specific mechanisms of an EMI shielding is challenging, as all working mechanisms (e.g., dielectric loss mechanisms) can contribute to the ultimate EMI SE, but their individual contributions are difficult to be quantified. Continued research is crucial to thoroughly investigate the working mechanisms and precisely identify the contributions of various loss mechanisms, which could be greatly aided by the development of advanced characterization techniques.^[26] Gaining deeper insights into these aspects will undoubtedly aid in developing more efficient insulating EMI shielding materials with innovative mechanisms.

- 4) Currently, research on the electrically insulating EMI shielding materials primarily focuses on the gigahertz frequency band (wavelength range: 3–300 mm), while their potential for extremely high frequency applications (wavelength range: 0.03–3 mm) remains largely unexplored. In particular, in recent years, terahertz wave has garnered significant interests for a wide range of applications, including detection, security screening, medical imaging, and wireless communication, which has boosted the demand for high-performance terahertz absorbing and shielding materials.^[74] For composite materials, their fillers, such as CNTs, graphene, and MXenes, typically range in size from a few nanometers to several micrometers, closely matching the wavelength of terahertz waves. The smaller difference between the size of the fillers and the wavelength of the incident electromagnetic waves can enhance the scattering and attenuation of these waves, thereby contributing to improved shielding and absorption. In addition, at higher frequencies, the intrinsic conductive properties of the dispersed filler becomes more crucial than the formation of continuous conductive network for effective EMI shielding, and the high-frequency electromagnetic waves can also better couple through the capacitance between adjacent, unconnected fillers within the polymer matrix.^[14,75] Thus, it can be expected that developing high-performance insulating EMI shielding materials will have a broad scope when targeting extremely-high frequency electromagnetic waves, which is worth further investigation.
- 5) Operating under harsh conditions requires that the electrically insulating EMI shielding materials are capable of resisting damage and preventing insulation failures from high voltages, severe temperature shocks, and extreme heat or cold.^[76] Additionally, some electronic applications also demand EMI shielding materials with advanced functionalities, such as thermal conductivity for efficient heat dissipation, joule heating for proactive thermal management, high transparency for optical applications, and active modulation capabilities for smart EMI shielding.^[77–81] Addressing these diverse requirements while maintaining favorable EMI shielding and insulating properties calls for innovative approaches in both compositional elements, such as selecting appropri-

ate polymer matrices (e.g., polyimide for high-temperature applications^[82]), and in structural design. Such advancements are crucial for enhancing the durability, effectiveness, and broad applicability of the insulating EMI shielding materials.

- 6) Embracing machine learning-assisted design methods may revolutionize material development in this field, allowing researchers to move beyond traditional trial-and-error approaches. It has been indicated that by analyzing extensive datasets obtained from published reports, experimental results, and numerical simulations, machine learning can develop precise prediction models for electromagnetic properties. Meanwhile, machine learning can also enable the inverse designs. By training the artificial neural networks, machine learning models can be generated for accurately mapping the relationship between design parameters and properties of materials, thereby providing optimized design parameters.^[66] These advantages of machine learning can significantly aid in the optimization of the design of EMI shielding materials prior to manufacturing.

4. Conclusion

The EMI shielding solutions that utilize intrinsically insulating materials can surpass the limitations of conventional approaches, thereby offering enhanced reliability and versatility for electronic applications. Although the exploration of novel electrically insulating EMI shielding materials has only begun to gain attention in recent years, early research advancements have already uncovered vast opportunities and considerable potential across a range of application scenarios. To shape a successful future, it is crucial to not only deepen our understanding of the intricate composition-structure-property relationships and working mechanisms of these materials but also to refine manufacturing techniques. We envision that continued in-depth research could lead to a new wave of innovation in EMI shielding materials, potentially transforming the landscape of EMI shielding solutions.

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Conflict of Interest

The authors declare no conflict of interest.

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- [1] A. Iqbal, T. Hassan, S. M. Naqvi, Y. Gogotsi, C. M. Koo, *Nat. Rev. Electr. Eng.* **2024**, *1*, 180.
- [2] A. Iqbal, F. Shahzad, K. Hantanasirisakul, M.-K. Kim, J. Kwon, J. Hong, H. Kim, D. Kim, Y. Gogotsi, C. M. Koo, *Science* **2020**, *369*, 446.
- [3] G. Wang, S. J. H. Ong, Y. Zhao, Z. J. Xu, G. Ji, *J. Mater. Chem. A* **2020**, *8*, 24368.
- [4] H. Abbasi, M. Antunes, J. I. Velasco, *Prog. Mater. Sci.* **2019**, *103*, 319.
- [5] J. Liu, M.-Y. Yu, Z.-Z. Yu, V. Nicolosi, *Mater. Today* **2023**, *66*, 245.
- [6] Q. Wei, S. Pei, X. Qian, H. Liu, Z. Liu, W. Zhang, T. Zhou, Z. Zhang, X. Zhang, H.-M. Cheng, W. Ren, *Adv. Mater.* **2020**, *32*, 1907411.
- [7] S. Wan, X. Li, Y. Chen, N. Liu, Y. Du, S. Dou, L. Jiang, Q. Cheng, *Science* **2021**, *374*, 96.
- [8] B. Yao, W. Hong, T. Chen, Z. Han, X. Xu, R. Hu, J. Hao, C. Li, H. Li, S. E. Perini, M. T. Lanagan, S. Zhang, Q. Wang, H. Wang, *Adv. Mater.* **2020**, *32*, 1907499.
- [9] X. Shen, J.-K. Kim, *Nano Res.* **2023**, *16*, 1387.
- [10] Y. Liu, Y. Wang, N. Wu, M. Han, W. Liu, J. Liu, Z. Zeng, *Nano-Micro Lett.* **2023**, *15*, 240.
- [11] M. Han, C. E. Shuck, R. Rakhmanov, D. Parchment, B. Anasori, C. M. Koo, G. Friedman, Y. Gogotsi, *ACS Nano* **2020**, *14*, 5008.
- [12] D. D. L. Chung, *Carbon* **2001**, *39*, 279.
- [13] F. Shahzad, M. Alhabeb, C. B. Hatter, B. Anasori, S. Man Hong, C. M. Koo, Y. Gogotsi, *Science* **2016**, *353*, 1137.
- [14] K. Hayashida, Y. Matsuoka, *Carbon* **2015**, *85*, 363.
- [15] P. Zhang, X. Ding, Y. Wang, Y. Gong, K. Zheng, L. Chen, X. Tian, X. Zhang, *Composites, Part A* **2019**, *117*, 56.
- [16] Z. Barani, F. Kargar, Y. Ghafouri, S. Ghosh, K. Godziszewski, S. Baraghani, Y. Yashchysyn, G. Cywiński, S. Rumyantsev, T. T. Salguero, A. A. Balandin, *Adv. Mater.* **2021**, *33*, 2007286.
- [17] Z. Zhao, L. Zhang, H. Wu, *Adv. Mater.* **2022**, *34*, 2205376.
- [18] Y. Xiao, G. Chen, B. Shi, Q. Chang, L. Zhang, H. Wu, *Small* **2024**, *2400756*, <https://doi.org/10.1002/sml.202400756>.
- [19] Y. Guo, H. Qiu, K. Ruan, S. Wang, Y. Zhang, J. Gu, *Compos. Sci. Technol.* **2022**, *219*, 109253.
- [20] C.-P. Feng, S.-S. Wan, W.-C. Wu, L. Bai, R.-Y. Bao, Z.-Y. Liu, M.-B. Yang, J. Chen, W. Yang, *Compos. Sci. Technol.* **2018**, *167*, 456.
- [21] G. Wang, X. Liao, F. Zou, P. Song, W. Tang, J. Yang, G. Li, *Composites Commun.* **2021**, *28*, 100953.
- [22] S. Anand, M. C. Vu, D. Mani, J.-B. Kim, T.-H. Jeong, M. A. Islam, S.-R. Kim, *Chem. Eng. J.* **2023**, *462*, 142017.
- [23] H. Liu, R. Fu, X. Su, B. Wu, H. Wang, Y. Xu, X. Liu, *Composites Commun.* **2021**, *23*, 100593.
- [24] W. Chen, L.-X. Liu, H.-B. Zhang, Z.-Z. Yu, *ACS Nano* **2020**, *14*, 16643.
- [25] X.-X. Wang, Q. Zheng, Y.-J. Zheng, M.-S. Cao, *Carbon* **2023**, *206*, 124.
- [26] M. Qin, L. Zhang, H. Wu, *Adv. Sci.* **2022**, *9*, 2105553.
- [27] D. C. Hogg, F. O. Guiraud, *Nature* **1979**, *279*, 408.
- [28] W.-L. Song, Y.-J. Zhang, K.-L. Zhang, K. Wang, L. Zhang, L.-L. Chen, Y. Huang, M. Chen, H. Lei, H. Chen, D. Fang, *Adv. Sci.* **2020**, *7*, 1902162.
- [29] Y. Yang, N. Wu, B. Li, W. Liu, F. Pan, Z. Zeng, J. Liu, *ACS Nano* **2022**, *16*, 15042.
- [30] D. Lai, X. Chen, G. Wang, X. Xu, Y. Wang, *Carbon* **2022**, *188*, 513.
- [31] Y. Yu, P. Yi, W. Xu, X. Sun, G. Deng, X. Liu, J. Shui, R. Yu, *Nano-Micro Lett.* **2022**, *14*, 77.
- [32] J. Liu, L. McKeon, J. Garcia, S. Pinilla, S. Barwich, M. Möbius, P. Stamenov, J. N. Coleman, V. Nicolosi, *Adv. Mater.* **2022**, *34*, 2106253.
- [33] Y. Zhu, J. Liu, T. Guo, J. J. Wang, X. Tang, V. Nicolosi, *ACS Nano* **2021**, *15*, 1465.
- [34] Y. Yang, B. Li, N. Wu, W. Liu, S. Zhao, C. J. Zhang, J. Liu, Z. Zeng, *ACS Mater. Lett.* **2022**, *4*, 2352.
- [35] S. Lu, B. Ouyang, S. Han, F. Qiao, K. Chen, F. Wu, A. Xie, E. Kan, H. Zeng, *ACS Appl. Nano Mater.* **2022**, *5*, 11407.
- [36] X.-X. Wang, M. Zhang, J.-C. Shu, B. Wen, W.-Q. Cao, M.-S. Cao, *Carbon* **2021**, *184*, 136.
- [37] M.-M. Lu, W.-Q. Cao, H.-L. Shi, X.-Y. Fang, J. Yang, Z.-L. Hou, H.-B. Jin, W.-Z. Wang, J. Yuan, M.-S. Cao, *J. Mater. Chem. A* **2014**, *2*, 10540.
- [38] J. Hong, J. Kwon, A. Iqbal, D. Kim, T. Kwon, P. Sambyal, S. M. Hong, H. G. Yoon, M.-K. Kim, C. M. Koo, *Chem. Eng. J.* **2022**, *438*, 135564.
- [39] X. Hu, Y. Cheng, Z. Wei, Y. Zhan, R. Zhang, H. Xia, X. Jiang, *Chem. Eng. J.* **2024**, *493*, 152533.
- [40] H. J. Chen, P. Y. Lee, C. Y. Chen, S. L. Huang, B. W. Huang, F. J. Dai, C. F. Chau, C. S. Chen, Y. S. Lin, *Sci. Rep.* **2022**, *12*, 10232.
- [41] B. Wen, M.-S. Cao, Z.-L. Hou, W.-L. Song, L. Zhang, M.-M. Lu, H.-B. Jin, X.-Y. Fang, W.-Z. Wang, J. Yuan, *Carbon* **2013**, *65*, 124.
- [42] K. Tian, D. Hu, Q. Wei, Q. Fu, H. Deng, *J. Mater. Sci. Technol.* **2023**, *134*, 106.
- [43] H.-Y. Zhang, J.-Y. Li, Y. Pan, Y.-F. Liu, N. Mahmood, X. Jian, *Rare Met.* **2022**, *41*, 3612.
- [44] R. M. Simon, *Polym.-Plast. Technol. Eng.* **1981**, *17*, 1.
- [45] R. Sun, H.-B. Zhang, J. Liu, X. Xie, R. Yang, Y. Li, S. Hong, Z.-Z. Yu, *Adv. Funct. Mater.* **2017**, *27*, 1702807.
- [46] L. Wang, Z. Ma, Y. Zhang, L. Chen, D. Cao, J. Gu, *SusMat.* **2021**, *1*, 413.
- [47] Y. Shi, L. Chu, H. Wei, Z. Li, F. Qin, Q. Zhang, W. Li, *Cell Rep. Phys. Sci.* **2024**, *5*, 101903.
- [48] K. Hayashida, H. Tanaka, *Adv. Funct. Mater.* **2012**, *22*, 2338.
- [49] F. Guo, Y. Wang, K. Xue, L. Liu, J. Li, Y. Huang, *Compos. Sci. Technol.* **2024**, *248*, 110425.
- [50] Y. Han, K. Ruan, J. Gu, *Nano Res.* **2022**, *15*, 4747.
- [51] P. Zhang, R. Tian, X. Zhang, X. Ding, Y. Wang, C. Xiao, K. Zheng, X. Liu, L. Chen, X. Tian, *Composites, Part B* **2022**, *232*, 109611.
- [52] Y. Zhang, B. Tang, Y. Liu, R. Feng, S. Song, C. Xiong, L. Dong, *Polym. Compos.* **2020**, *41*, 1673.
- [53] Y. Xu, Y. Tan, Y. Xue, M. Pei, D. Zhang, S. Liu, S. Qin, *Polym. Eng. Sci.* **2023**, *63*, 3555.
- [54] X. Hu, Y. Cheng, Z. Wei, R. Zhang, Y. Zhan, H. Xia, *ACS Appl. Electron. Mater.* **2024**, *6*, 1770.
- [55] Y. Xu, M. Pei, J. Du, R. Yang, Y. Pan, D. Zhang, S. Qin, *New J. Chem.* **2023**, *47*, 13721.
- [56] S. Ghaderi, M. Kamkar, A. Ghaffarkhah, M. Amini, A. H. A. Hosein, M. Arjmand, *IEEE Sensors IEEE, Piscataway, NJ* **2021**.
- [57] J. Wan, J. Song, Z. Yang, D. Kirsch, C. Jia, R. Xu, J. Dai, M. Zhu, L. Xu, C. Chen, Y. Wang, Y. Wang, E. Hitz, S. D. Lacey, Y. Li, B. Yang, L. Hu, *Adv. Mater.* **2017**, *29*, 1703331.
- [58] Y. Wu, C. An, Y. Guo, Y. Zong, N. Jiang, Q. Zheng, Z.-Z. Yu, *Nano-Micro Lett.* **2024**, *16*, 118.
- [59] H. Bai, Y. Chen, B. Delattre, A. P. Tomsia, R. O. Ritchie, *Sci. Adv.* **2015**, *1*, e1500849.
- [60] P. Min, X. Li, P. Liu, J. Liu, X.-Q. Jia, X.-P. Li, Z.-Z. Yu, *Adv. Funct. Mater.* **2021**, *31*, 2103703.
- [61] W. Gao, N. Zhao, T. Yu, J. Xi, A. Mao, M. Yuan, H. Bai, C. Gao, *Carbon* **2020**, *157*, 570.
- [62] S. Pinilla, J. Coelho, K. Li, J. Liu, V. Nicolosi, *Nat. Rev. Mater.* **2022**, *7*, 717.
- [63] M. A. S. R. Saadi, A. Maguire, N. T. Pottackal, M. S. H. Thakur, M. M. Ikram, A. J. Hart, P. M. Ajayan, M. M. Rahman, *Adv. Mater.* **2022**, *34*, 2108855.
- [64] J. Feng, B.-L. Su, H. Xia, S. Zhao, C. Gao, L. Wang, O. Ogbeide, J. Feng, T. Hasan, *Chem. Soc. Rev.* **2021**, *50*, 3842.
- [65] H. Tetik, Y. Wang, X. Sun, D. Cao, N. Shah, H. Zhu, F. Qian, D. Lin, *Adv. Funct. Mater.* **2021**, *31*, 2103410.
- [66] Z. Zhao, Y. Qing, L. Kong, H. Xu, X. Fan, J. Yun, L. Zhang, H. Wu, *Adv. Mater.* **2024**, *36*, 2304182.
- [67] X. Zhang, F. Yan, S. Zhang, H. Yuan, C. Zhu, X. Zhang, Y. Chen, *ACS Appl. Mater. Interfaces* **2018**, *10*, 24920.
- [68] H. Lv, Z. Yang, H. Pan, R. Wu, *Prog. Mater. Sci.* **2022**, *127*, 100946.

- [69] Z. Li, L. Zhang, H. Wu, *Small*. **2024**, 20, 2305120.
- [70] J. Wen, G. Chen, S. Hui, Z. Li, J. Yun, X. Fan, L. Zhang, Q. He, X. Liu, H. Wu, *Adv. Powder Mater.* **2024**, 3, 100180.
- [71] B. Zhao, Y. Li, Q. Zeng, L. Wang, J. Ding, R. Zhang, R. Che, *Small*. **2020**, 16, 2003502.
- [72] Y. He, Q. Su, D. Liu, L. Xia, X. Huang, D. Lan, Y. Liu, Y. Huang, B. Zhong, *Chem. Eng. J.* **2024**, 491, 152041.
- [73] D. Tan, Q. Wang, M. Li, L. Song, F. Zhang, Z. Min, H. Wang, Y. Zhu, R. Zhang, D. Lan, B. Fan, *Chem. Eng. J.* **2024**, 492, 152245.
- [74] W. Xie, Q. Tang, J. Xie, Y. Fei, H. Wan, T. Zhao, T. Ding, X. Xiao, Q. Wen, *Nat. Commun.* **2024**, 15, 38.
- [75] P. Theilmann, D.-J. Yun, P. Asbeck, S.-H. Park, *Org. Electron.* **2013**, 14, 1531.
- [76] X. Zhou, Y. Liu, Z. Gao, P. Min, J. Liu, Z.-Z. Yu, V. Nicolosi, H.-B. Zhang, *Adv. Mater.* **2024**, 36, 2310849.
- [77] J. Cheng, C. Li, Y. Xiong, H. Zhang, H. Raza, S. Ullah, J. Wu, G. Zheng, Q. Cao, D. Zhang, Q. Zheng, R. Che, *Nano-Micro Lett.* **2022**, 14, 80.
- [78] M. Han, D. Zhang, C. E. Shuck, B. McBride, T. Zhang, R. Wang, K. Shevchuk, Y. Gogotsi, *Nat. Nanotechnol.* **2023**, 18, 373.
- [79] N. Zhang, Z. Wang, R. Song, Q. Wang, H. Chen, B. Zhang, H. Lv, Z. Wu, D. He, *Sci. Bull.* **2019**, 64, 540.
- [80] Z. Ma, X. Xiang, L. Shao, Y. Zhang, J. Gu, *Angew. Chem., Int. Ed.* **2022**, 61, e202200705.
- [81] B. Shen, W. Zhai, W. Zheng, *Adv. Funct. Mater.* **2014**, 24, 4542.
- [82] Y. Cheng, X. Li, Y. Qin, Y. Fang, G. Liu, Z. Wang, J. Matz, P. Dong, J. Shen, M. Ye, *Sci. Adv.* **2021**, 7, eabj1663.