Polymer-based EMI shielding composites with 3D conductive networks: A mini-review

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Abstract
High-frequency electromagnetic waves and electronic products can bring great convenience to people’s life, but lead to a series of electromagnetic interference (EMI) problems, such as great potential dangers to the normal operation of electronic components and human safety. Therefore, the research of EMI shielding materials has attracted extensive attention by the scholars. Among them, polymer-based EMI shielding materials with light weight, high specific strength, and stable properties have become the current mainstream. The construction of 3D conductive networks has proved to be an effective method for the preparation of polymer-based EMI shielding materials with excellent shielding effectiveness (SE). In this paper, the shielding mechanism of polymer-based EMI shielding materials with 3D conductive networks is briefly introduced, with emphasis on the preparation methods and latest research progress of polymer-based EMI shielding materials with different 3D conductive networks. The key scientific and technical problems to be solved in the field of polymer-based EMI shielding materials are also put forward. Finally, the development trend and application prospects of polymer-based EMI shielding materials are prospected.

KEYWORDS
3D conductive networks, conductive polymer composites, polymer-based EMI shielding materials

1 | INTRODUCTION
With the advent of 5G era, the applications of high-frequency electromagnetic waves and electronic products are gradually broadened, which brings great convenience to people’s life, and leads to a series of electromagnetic interference (EMI) problems. It not only seriously threatens the normal operation of electronic components, but...
also causes potential security risks to human beings.\textsuperscript{1–3} Therefore, the research and development of EMI shielding materials is becoming increasingly urgent.\textsuperscript{4–6}

Polymer-based conductive composites have gradually replaced the traditional metal-based shielding materials because of their advantages, such as low density, high-specific strength, stable chemical properties, and easy processing.\textsuperscript{7–9} The relatively satisfactory EMI shielding performance can be achieved by simply adding magnetic or highly conductive fillers. However, it needs a large amount of fillers, which inevitably lead to the problems of processing difficulty and low mechanical properties, and the shielding effectiveness (SE) is still difficult to meet the demands of superior shielding performances.\textsuperscript{10,11} The researchers have attempted to improve the EMI SE of polymer-based composites to some extent with the same filler loading by surface functionalization of fillers or external induction, but the improvement effect is still limited and far lower than the expected.\textsuperscript{12–} Research shows that the construction of 3D conductive networks can realize the uniform dispersion of conductive fillers to effectively connect the conductive fillers to conduct electrons, and the abundant interfaces are conducive to further enhance the attenuation of electromagnetic waves, achieving remarkable EMI SE for polymer-based composites with ultralow loading of conductive fillers.\textsuperscript{13–16}

In this paper, the shielding mechanism and preparation methods of polymer-based EMI shielding materials in recent years, as well as the latest research progress in the field of EMI shielding are reviewed. The key scientific and technical problems that need to be solved in the field of polymer-based EMI shielding materials with 3D conductive networks are also proposed. Finally, the development trend and application prospects of polymer-based EMI shielding materials are prospected.

2 | SHIELING MECHANISM OF POLYMER-BASED EMI SHIELING MATERIALS WITH 3D CONDUCTIVE NETWORKS

EMI shielding refers to a shield that blocks the propagation of electromagnetic waves in the transmission path. The attenuation degree of electromagnetic waves is expressed by SE. According to Schelkunoff’s theory,\textsuperscript{17} when the electromagnetic waves propagate to the surface of the shielding material, some electromagnetic waves are directly reflected (SE\textsubscript{R}) due to impedance mismatch. The rest of electromagnetic waves are absorbed by the materials as absorption loss (SE\textsubscript{A}), and the parts attenuated by multiple reflection are multiple reflection loss (SE\textsubscript{M}). Usually, SE\textsubscript{M} can be ignored when SE\textsubscript{A} is higher than 10 dB.\textsuperscript{12} The total SE (SE\textsubscript{T}) is shown as Equation (1):

\[
SE_T = SE_R + SE_A
\]

\(SE_A\) originates from the interaction of electric or magnetic dipoles in a conductor material with the electromagnetic field. The SE\textsubscript{A} in the shield has no concern with the type of electromagnetic waves, but greatly depends on the thickness, frequency, conductivity, and permeability of the shield. The greater the thickness, frequency, conductivity, and permeability, the greater SE\textsubscript{A} of the EMI shielding. SE\textsubscript{R} is caused by the mismatch between the spatial impedance and the inherent impedance of the shielding layer. It results from the interaction between the conductive particles in the conductor materials and the electromagnetic field related to the permeability and conductivity. The higher conductivity or the lower permeability results in the greater SE\textsubscript{R} of the materials.

The types of shielding materials are usually determined by the S parameters (S\textsubscript{11}, S\textsubscript{21}, S\textsubscript{22}, S\textsubscript{12}) obtained by the vector network analyzer. S\textsubscript{ij} represents electromagnetic waves transfer from port j to port i. According to these S parameters, the reflectance rate (R), absorption rate (A), and transmittance rate (T) can be calculated in Equation (2), Equation (3), and Equation (4), respectively:

\[
R = |S_{11}|^2 = |S_{22}|^2
\]

\[
T = |S_{12}|^2 = |S_{21}|^2
\]

\[
A = 1 - R - T
\]

Usually, when A > R, the materials are absorbing-type shielding materials. On the contrary, they are reflecting-type shielding materials.

When the electromagnetic waves act on the 3D conductive networks, some of electromagnetic waves are reflected due to impedance mismatch.\textsuperscript{18} During the penetration of remaining electromagnetic waves within the 3D conductive networks, the 3D conductive networks generate strong eddy current loss and other electrical losses and convert the electromagnetic waves into heat energy due to their superior conductivity. At the same time, the unique 3D conductive networks possess a large specific surface area and abundant interfaces. Due to the dipole polarization and dielectric loss caused by the interfaces, the 3D conductive networks further promote the conversion of electromagnetic waves into heat energy. A large number of capacitive structures can be formed regionally between the fillers with different electrical conductivities, as well as between the fillers and matrix, leading to the accumulation, rearrangement of space charges, and interfacial polarization in the alternating electromagnetic fields.
Moreover, the transmission path of electromagnetic waves is prolonged by multiple reflection and scattering in the 3D conductive networks, further enhancing the reabsorption and dissipation of electromagnetic waves in the conduction process of 3D conductive networks. In addition, the crossing, jumping, and migration of electrons in 3D conductive networks also play an important role in the attenuation of electromagnetic energy, which strengthens the loss of electromagnetic waves. Therefore, only a small part of electromagnetic waves can pass through the shielding materials, resulting in the absorption-dominated shielding mechanism (Figure 1).19–22

3 | PREPARATION METHODS OF POLYMER-BASED EMI SHIELDING MATERIALS WITH 3D CONDUCTIVE NETWORKS

By constructing 3D conductive networks among the conductive fillers within the polymer matrix, polymer-based EMI shielding composites with light weight, outstanding conductivity, excellent EMI shielding performances, and outstanding mechanical properties can be prepared with a pretty low loading of conductive fillers. The preparation methods of polymer-based EMI shielding composites mainly include the template method, freeze-drying method, hydrothermal method, and foaming method.23–27

3.1 | Template method

In most template methods, 3D conductive networks are constructed based on the open-celled foams with 3D cellular structures as templates, which are graphitized at high temperature or filled with conductive fillers.28 This method has attracted extensive attention because of their advantages, such as high flexibility, simple preparation conditions, and easily controlled structure design. Guo et al.29 took the polyurethane (PU) sponge as template, and prepared the carbon foam/epoxy nanocomposites by the impregnation of lignin-resorcinol-glyoxal prepolymer after high temperature graphitization and filling with epoxy resin. The results showed that when the loading of carbon foam was 6 wt%, the $\sigma$ and EMI SE of epoxy nanocomposites could reach 216 S/m and 34 dB, respectively. Chen et al.31 took the nickel foams as template, and prepared the light and flexible graphene foam after growth of graphene on the nickel foam and impregnation of polydimethylsiloxane (PDMS) (Figure 2A). When the loading of graphene was only 0.8 wt%, the graphene foam with a density of only 0.06 g/cm$^3$ presented an EMI SE of 30 dB and a specific SE
(SSE = SE/density) of 500 dB·cm$^3$/g, which exceeded the shielding performances of many metal-based and carbon-based shielding materials. Moreover, the excellent flexibility also endowed the EMI SE of the graphene foam with extraordinary stability, and there was no obvious decline of shielding performances after bending for 1000 times. In the previous research of our group, Gu et al.\textsuperscript{30} used alumina honeycomb as template for the deposition of GO, and prepared the rGO foams with honeycomb structures by further thermal reduction. The results showed that when the loading of rGO honeycomb was 1.2 wt%, the EMI SE of rGO foam/epoxy nanocomposites reached up to 38 dB, which was 6.3 times higher than that of the blended rGO/epoxy nanocomposites (6 dB). Gu et al.\textsuperscript{31} obtained the highly conductive PU@PDA@Ag sponge using PU sponge as template to in-situ deposit Ag nanoparticles. With 29 wt% loading of Ag nanoparticles, PU@PDA@Ag sponge exhibited remarkable EMI SE of 84 dB and SSE of 2625 dB·cm$^3$/g, respectively (Figure 2B). In recent years, biomass materials and their derivatives (such as wood, sugarcane, cotton, bread, corrugated cardboard, paper tissue, etc.) have shown great potential in the field of EMI shielding owing to their advantages of rich sources, low cost, nontoxic, and natural 3D networks.\textsuperscript{32-38} Jia et al.\textsuperscript{39} used the corrugated cardboard as template, and prepared carbon foam/epoxy composites by thermally annealing and impregnation of epoxy resin. Different types of corrugated board can be used to adjust the EMI SE of carbon foam/epoxy composites in the range of 46–82 dB and SSE in the range of 325–1171 dB·cm$^3$/g. The disadvantage of this method is that conductive fillers tend to fall off from the template especially upon external stress or strain, resulting in the structural defects.

3.2 Freeze-drying method

In the freeze-drying method, the conductive fillers in suspension are usually assembled and frozen to form ice crystals or solid organic molecules. After sublimating the solvent of the reaction system at low temperature and pressure, the 3D conductive networks are constructed by maintaining the original dispersion state of the fillers by the interaction between the adjacent conductive fillers. This method can be carried out in aqueous phase, and is easy to control the composition and structures of 3D conductive networks. Wang et al.\textsuperscript{40} prepared the CNTs/polyimide (PI) foams by freeze-drying the mixture of CNTs with polyamic acid followed by thermal imide. When the loading of CNTs was 56 wt%, the EMI SE of
CNTs/PI foam reached 41 dB. Owing to the excellent flame retardancy, the CNTs/PI foams still displayed a high EMI SE up to 35 dB after burning upon alcohol lamp. Xu et al.\textsuperscript{41} reported that the MXene/polyvinyl alcohol (PVA) hybrid foams were prepared by the facile freeze-drying method (Figure 3A–J). The MXene/PVA hybrid foams with the MXene loading of 66 wt% presented high EMI SE of 28 dB and SSE of 5136 dB cm$^3$/g, respectively. Wu et al.\textsuperscript{20} obtained the superelastic MXene/PDMS foams via freeze-drying the mixture of MXene with sodium alginate followed by impregnation of PDMS. With the MXene loading of 6.1 wt%, the MXene/PDMS foams exhibited a high EMI SE of 54 dB. Moreover, the combination of flexible matrix and 3D conductive networks endowed the MXene/PDMS foams with excellent stability. Even after 500 cyclic compression tests, the EMI SE was still 48 dB. In the previous research work of our group, Gu et al.\textsuperscript{42} used the freeze-drying method and vacuum-assisted impregnation processes to obtain the PANI/multi-walled carbon nanotubes (MWCNTs)/graphene aerogel/epoxy nanocomposites. With the synergistic effect of graphene and MWCNTs, the EMI SE of PANI/MWCNTs/graphene aerogel/epoxy nanocomposites with 4.61 wt% PANI/MWCNTs/graphene aerogel of loading reached the optimal EMI SE of 42 dB.
when the loading ratio of GO to MWCNTs was 3:1. Gu et al. also prepared the copper nanowires/graphene aerogel (CUNWS-TAGA) via the freeze-drying method (Figure 3K–O). However, it is difficult to realize the large-scale production via the freeze-drying method due to the long time running and harsh terms with low temperature and low pressure.

3.3 Hydrothermal method

In the hydrothermal method, the reaction system is heated and pressurized to improve the activity of reactants and promote the self-assembly or lapping between the conductive fillers, thus constructing the 3D conductive networks. The required equipment and reaction conditions are relatively simple and easy to control. It is a common method for assembling nanofillers into 3D structures. Zhao et al. assembled highly conductive and superelastic graphene/CNTs hybrid foams by the hydrothermal method, and then further impregnated with PDMS to prepare the graphene/CNTs/PDMS nanocomposites. At the high temperature and pressure, the graphene and carbon nanotubes were assembled together to form the highly developed 3D porous networks. The results showed that 0.21 wt% graphene/0.07 wt% CNTs/PDMS nanocomposites presented a \( \sigma \) of 1.2 S/cm and an EMI SE of 31 dB, respectively. Chen et al. prepared the phenolic resin/GO foams by hydrothermal method, and then obtained the graphene/carbon foam/epoxy nanocomposites via annealing phenolic resin/GO foams followed by impregnation of epoxy resin. Thanks to the synergistic effect of 2D graphene nanosheets and carbon fiber, the \( \sigma \) and EMI SE of 0.33 wt% graphene/carbon foam/epoxy nanocomposites reached 73 S/m and 35 dB, respectively. Zhao et al. prepared the MXene/graphene aerogel/epoxy nanocomposites by the vacuum-assisted impregnation process. A large number of highly conductive MXene nanosheets were coated on the surface of graphene skeleton, forming an efficient conductive network. It would produce strong electrical loss, reflection, and scattering in the porous structures, which was conducive to promoting the dissipation of electromagnetic waves. With only 0.74 vol% of MXene, the MXene/graphene aerogel/epoxy nanocomposites showed an ultrahigh \( \sigma \) of 695.9 S/m and an outstanding EMI SE of 50 dB, respectively (Figure 4A).

In the previous research of our group, Gu et al. assembled \( \text{Fe}_2\text{O}_4 \)/graphene aerogel by the hydrothermal method and impregnated with epoxy resin to prepare the \( \text{Fe}_3\text{O}_4 \)/graphene aerogel/epoxy nanocomposites. Owing to the magnetic loss of \( \text{Fe}_3\text{O}_4 \) and enhanced dielectric loss, the EMI SE of the 1.5 wt% \( \text{Fe}_3\text{O}_4 \)/1.2 wt% graphene aerogel/epoxy nanocomposites reached 35 dB, which was 30% higher than that of the graphene aerogel/epoxy nanocomposite (27 dB), while \( \text{SE}_{\text{R}} \) was almost unchanged. Gu et al. prepared the MXene/C hybrid foam/epoxy nanocomposites by the hydrothermal assembly of MXene/C hybrid foam and impregnation of epoxy resin (Figure 4B). With 4.25 wt% MXene/C hybrid foam loading, the MXene/C hybrid foam/epoxy nanocomposites presented a \( \sigma \) of 184 S/m and an excellent EMI SE of 46 dB, respectively. The disadvantage of this method is that the material size is limited by the reaction container.

3.4 Foaming method

The foaming method mainly introduces a large amount of gas into the composites by the physical or chemical processes to form the mutually penetrated or closed cellular structures. In the porous structures, the matrix/conductive filler composite phase acts as the continuous phase to construct the 3D conductive networks, while the gas acts as the dispersed phase. The supercritical carbon dioxide foaming method resembles a novel physical foaming method, which is favored by scholars with the combined advantages of easy processing, low cost, and environmental protection. Kuang et al. prepared the lightweight and degradable MWCNTs/L-poly(l-lactic acid) (PLLA) nanocomposites by the supercritical carbon dioxide foaming method. The results showed that the closed cellular structures endowed the MWCNTs/PLLA nanocomposites with higher EMI SE and SSE compared to the blended MWCNTs/PLLA nanocomposites due to the stronger electrical loss and reflection of electromagnetic waves. When the MWCNT loading was 10 wt%, the EMI SE and SSE of MWCNTs/PLLA nanocomposites were 23 dB and 77 dB cm\(^{-2}\) g\(^{-1}\), respectively. Li et al. prepared the MWCNTs/epoxy composites foams by the supercritical carbon dioxide foaming method. The SSE of 1 wt% MWCNTs/epoxy composites foams increased from 5.2 to 21.3 dB cm\(^{-2}\) g\(^{-1}\) owing to the reaggregation and orientation of MWCNTs during the foaming processes. Fan et al. applied the functionalized nano-carbon black (F-NCB) as modifier and nucleating agent, and obtained the F-NCB/silver nanosheets (AGNS)/epoxy foams with an expansion rate of up to 20% by the supercritical carbon dioxide foaming method (Figure 5A). The EMI SE and SSE of epoxy foams reached 43 dB and 334.59 dB cm\(^{-2}\) g\(^{-1}\), respectively, with the AGNS loading of 20.0 wt%. Yang et al. prepared the flexible Ag@insulating glass bead/\( \text{Fe}_2\text{O}_4 @\text{MWCNTs/silicone rubber (SR)} \) composite foams by the supercritical carbon dioxide foaming technique (Figure 5M–O). When the Ag loading was 0.51 vol%, the EMI SE of Ag@insulating glass bead/\( \text{Fe}_2\text{O}_4 @\text{MWCNTs/SR} \) composite foams with a thickness of 0.7 mm reached 31 dB. The flexibility of the
SR and construction of 3D conductive networks provided the Ag@insulating glass bead/Fe₃O₄@MWCNTs/SR composite foams with superior stability, so that the EMI SE remained unchanged even after folding for 1000 times. The disadvantages of this method are that the penetration depth of carbon dioxide is limited and it is difficult to control the morphologies of internal cells.

3.5 Other methods

In addition to the above methods, other methods have also been used to construct the 3D conductive networks with outstanding $\sigma$. Chen et al.⁵² prepared the rGO/Fe₃O₄/cellulose films with 3D conductive networks by the solvent evaporation method (Figure 6A–C). When the loadings of rGO and Fe₃O₄ were 15 wt% and 8 wt%, respectively, the EMI SE of rGO/Fe₃O₄/cellulose films reached 24 dB. Shi et al.⁵³ reported a novel liquid–liquid interface self-assembly method to prepare the MXene aerogel (Figure 6D–F). The corresponding MXene aerogel/epoxy composites showed an EMI SE of 35 dB with only 0.40 vol% MXene. Yu et al.⁵⁴ prepared the CNTs/PE nanocomposites with segregated structures through hot compression molding after assembling PE microspheres and CNTs by electrostatic interaction.
The unique segregated structures realized the tight connection between the adjacent CNTs and formed the highly conductive networks to attenuate electromagnetic waves. With only 2 wt% CNTs, the EMI SE of CNTs/PE nanocomposites with 2 wt% CNT loadings reached 34 dB.

In order to highlight the advantages of 3D conductive networks, Table 1 summarizes the loading of fillers, thickness, s and EMI SE of the polymer-based shielding materials with various 3D conductive networks. Thanks to the efforts done by many researchers, a large number of lightweight and high-performance shielding materials with unique 3D structures have been prepared. These materials far exceed the basic commercial application requirements (> 20 dB), and some of them have met the military and aerospace indexes (> 60 dB), laying a solid foundation for the development of EMI shielding field in the future.
There are many kinds of polymer-based shielding materials with 3D conductive networks, which can be divided into mainly four categories according to their configurations, including polymer-based shielding materials with randomly distributed 3D conductive networks, anisotropic 3D conductive networks, segregated structures, and 3D multi-interface networks.

The general 3D conductive networks consist of randomly distributed conductive fillers overlapped with each other. Among them, the 3D conductive networks of the conductive fillers and the cross-linking networks of the polymer matrix run through each other to form the stable and randomly distributed 3D conductive networks. Shen et al.63 prepared the porous graphene/polyetherimide (PEI) hybrid foams by the phase separation method.
The construction of 3D conductive networks significantly improved the shielding performances of the graphene/PEI hybrid foams. When the graphene loading was 10 wt%, the EMI SE and SSE of graphene/PEI hybrid foams reached 22 dB and 44 dB cm⁻³/g, respectively. In view of the easy agglomeration and harsh gelation conditions of nanofillers, the cellular structures assembled with polymers are favored by their advantages of simple preparation process and easy performance control. Liu et al. prepared the hyperelastic graphene foams with randomly distributed 3D conductive networks by bridging graphene nanosheets after the thermal imidization of PI (Figure 7A–D). The 3D cross-linking networks were able to restore the initial shape at 90% deformation, and the developed conductive networks also endowed the graphene foams with the outstanding σ of 1000 S/m and remarkable EMI SE of 83 dB. The introduction of magnetic particles is a common way to improve the impedance matching characteristics, which is beneficial to reduce the reflection of electromagnetic waves. Zheng et al. introduced Fe₃O₄ nanoparticles to prepare the porous graphene@Fe₃O₄/PEI hybrid foams. The introduction of Fe₃O₄ nanoparticles significantly improved the absorption of electromagnetic waves for the graphene@Fe₃O₄/PEI hybrid foams by magnetic loss and dielectric loss. When the loading of graphene@Fe₃O₄ was 10 wt%, the SSE of graphene@Fe₃O₄/PEI hybrid foams in the X-band reached 41.5 dB cm⁻³/g, and the reflection loss was only 0.5 dB due to the improvement of the impedance matching, indicating that most of the electromagnetic waves were dissipated inside the foams. In the previous research of our group, Gu et al. introduced Fe₃O₄ nanoparticles into disordered porous 3D Fe₃O₄–CNT/graphene foams (Figure 7E–G). When the loading of graphene and Fe₃O₄–CNT were 0.24 wt% and 2.76 wt%, respectively, the EMI SE of 3D Fe₃O₄–CNT/graphene foam/epoxy nanocomposites reached 36 dB. Compared with the nanocomposites without Fe₃O₄ nanoparticles, the nanocomposites containing Fe₃O₄ nanoparticles exhibited an improved EMI SE by 16% with an unchanged SER.

| Nanocomposites | Filler content wt% | Thickness mm | Conductivity S/m | EMI SE dB | Frequency GHz | Refs |
|----------------|--------------------|--------------|------------------|-----------|------------|------|
| Ag NWs/PI      | 4.5                | 0.029        | –                | 35        | 8–12       | 55   |
| Graphene/PDMS  | Bulk               | 0.3          | 3600             | 83        | 2–18       | 23   |
| MXene@NR       | 6.71#              | 0.246        | 1400             | 54        | 8.2–12.4   | 56   |
| CNTs/Ni@CNTs/PVDF | 22               | 0.5          | 257              | 51        | 18–27      | 57   |
| rGO/Fe₂O₃–cellulose | 8           | 0.16         | < 0.1            | 20.4      | 8.2–12.4   | 58   |
| rGO/PU         | 10                 | 60           | –                | 40        | 8–12       | 59   |
| CNTs/PDMS      | 1.74#              | 2            | 66               | 43        | 8.2–12.4   | 60   |
| CNTs/PI        | 67                 | 2            | 17.1             | 41        | 8.2–12.4   | 40   |
| MXene/PDMS     | 6.1                | 2            | 2211             | 71        | 8.2–12.4   | 20   |
| MWCNTs/WPU     | 76.2               | 4.5          | 45               | 51        | 8.2–12.4   | 61   |
| MXene/PVA      | 0.15#              | 5            | 8.3 × 10⁻⁶       | 28        | 8.2–12.4   | 41   |
| rGO/PI         | Bulk               | 2            | 1000             | 83        | 8–12       | 62   |
| Graphene@Fe₂O₄/PEI | 10              | 2.5          | –                | 18        | 8–12       | 63   |
| Ag@HGMs/Fe₂O₃@MWCNTs/SR | 0.51#        | 2            | 279              | 59        | 8.2–12.4   | 51   |
| NCB/AgNS/epoxy foam | 20                | 2            | 89               | 51        | 8.2–12.4   | 50   |
| CNT sponge/epoxy | 1.34             | 2            | 516              | 40        | 8–12       | 15   |
| rGO foam/epoxy  | 1.2                | 3            | 40               | 38        | 8.2–12.4   | 30   |
| MXene/C foam/epoxy | 4.25            | 2            | 184              | 46        | 8.2–12.4   | 10   |
| MXene/rGO aerogel/epoxy | 0.74#      | 2            | 696              | 50        | 8.2–12.4   | 22   |
| Fe₂O₃/rGO aerogel/epoxy | 2.7             | 3            | 27.5             | 35        | 8.2–12.4   | 63   |
| Fe₂O₃–CNTs/rGO foam/epoxy | 3          | 3            | 15.3             | 36        | 8.2–12.4   | 64   |
| MXene aerogel/epoxy | 0.40#          | 2            | 416.6            | 34.5      | 8.2–12.4   | 53   |
| MXene@PS       | 6.2                | 2            | 1081             | 62        | 8.2–12.4   | 65   |
| rGO@PS         | 3.47#              | 2.5          | 43.5             | 41        | 8.2–12.4   | 66   |

Note: # means the volume fraction; – indicates that the values are not available.
Compared with the simply blended polymer-based shielding materials, the randomly distributed 3D conductive networks form a more developed conductive path, which results in the significant increase in $\sigma$ and EMI SE at a lower filler loading. However, the conductive fillers with random distribution fail to make full utilization of their ability to attenuate electromagnetic waves, and the shielding performances can be further improved through structural optimization and other alternative methods.

### 4.2 Polymer-based shielding materials with anisotropic 3D conductive networks

With the control of external conditions, the ordered arrangement of conductive fillers in a single direction or multiple directions can be obtained. Simultaneously, the anisotropic 3D conductive networks can be constructed by the self-assembly of conductive fillers during the arrangement process. The ordered arrangement of conductive fillers can further reduce the percolating threshold of 3D
conductive networks with anisotropy. It shows higher conductivity along the arrangement direction, which is beneficial to improving the shielding performances. Li et al. prepared the anisotropic graphene aerogels by a directional freezing-freeze drying process. The radial EMI SE of the 0.8 wt% ordered graphene aerogel/epoxy nanocomposites reached 33 dB, which was 22% higher than that of the disordered graphene aerogel/epoxy nanocomposites (27 dB).

Zeng et al. reported that the anisotropic MWCNTs/WPU foams were obtained by directional freezing-freeze drying process. When the MWCNTs loading was 76.2 wt%, the MWCNTs/WPU foam presented outstanding radial EMI SE of 50 dB and SSE of 1148 dB⋅cm3/g, respectively (Figure 8A–H). The bidirectional freezing technique can further reduce the percolating threshold and improve the EMI SE of 3D conductive networks. Zeng et al. obtained the cellulose nanofibers (CNFs)/silver nanowire (Ag NW) aerogels by the bidirectional freezing method. The lamellar structure significantly enhanced the multiple reflection and absorption of electromagnetic waves. When the Ag NW loading was 50 wt%, the EMI SE and SSE/t of CNF/Ag NW aerogels at an extremely low density of 6.2 mg/cm3 reached remarkable 70 dB and 178 235 dB⋅cm2/g, respectively, exceeding the shielding performances of many other reported foams (Figure 8I–P). In addition, many biomass materials with natural 3D networks, such as wood and sugarcane, are also good raw materials for the preparation of shielding materials with anisotropic 3D conductive networks. Shen and Feng prepared the anisotropic wood-derived carbon foam using wood as template by high temperature annealing followed by filling of epoxy resin to prepare the wood-derived carbon foam/epoxy nanocomposites. With 7 vol% loading of wood-derived carbon foam, the σ and EMI SE of the wood-derived carbon foam/epoxy nanocomposites were 12.5 S/m and 28 dB, respectively.

The shielding performances of polymer-based shielding materials can be further enhanced by adjusting the microstructures of 3D conductive networks. The anisotropic 3D conductive networks are not only beneficial to the reduction of percolating threshold, but also generate more efficient filler/polymer interfaces to enhance the reflection and reabsorption of electromagnetic waves between the ordered structures, leading to the improved attenuation of electromagnetic waves. However, the ordered arrangement of conductive fillers tends to weaken the lap joint of the conductive fillers in the vertical direction, thus reducing the conductivity and mechanical properties in the vertical direction.
4.3 | Polymer-based shielding materials with segregated structures

The polymer-based shielding materials with segregated structures are mainly prepared by coating conductive fillers on the surface of polymer microspheres through electrostatic action or hydrogen bonding and followed by hot pressing molding, such as the segregated rGO@PS, MXene@PS, and CNTs@PE composites. In the segregated structures, the conductive fillers are selectively distributed in the interfaces between the polymer microspheres, forming an efficient 3D conductive network owing to the higher local content of conductive fillers. This results in the super high $\sigma$ and EMI SE at pretty low filler loading. Wu et al. first prepared the rGO@PS nanocomposites with segregated structures by the electrostatic self-assembly of GO on the surface of PS microspheres, followed by the chemical reduction of GO and hot pressing of rGO@PS microspheres. The unique segregated structure endowed the rGO@PS nanocomposites with a 3D highly conductive network. The percolating threshold was only 0.15 vol%, and the rGO@PS nanocomposites presented an outstanding $\sigma$ of 1024.8 S/m when the rGO loading was 4.8 vol%. Li’s group and Yu’s group conducted in-depth studies in the field of EMI shielding. Li et al. assembled GO nanosheets on the surface of PS microspheres and prepared rGO@PS nanocomposites with segregated structures by hot pressing. When the rGO loading was 3.47 vol%, the rGO@PS nanocomposites exhibited superior $\sigma$ of 43.5 S/m and EMI SE of 45 dB, respectively (Figure 9A–D). Yu et al. prepared the flexible MXene@natural rubber (NR) films with segregated structures by electrostatic self-assembly of MXene on the surface of NR microspheres followed by the vacuum filtration technique. The unique segregated structures not only provided the MXene@NR films with efficient electronic conduction capacity but also effectively conducted the external stress. The obtained MXene@NR films with a loading of 6.71 vol% exhibited high $\sigma$ of 1400 S/m and EMI SE of 54 dB, respectively. Compared with the pure NR film, the tensile strength and elastic modulus of the MXene@NR films were increased by 700% and 15 000%, respectively. Yu et al. obtained the MXene@PS nanocomposites with segregated structure by electrostatically self-assembly of MXene nanosheets and PS microspheres, followed with hot pressing (Figure 9E–J). When the MXene loading was 1.90 vol%, the $\sigma$ and EMI SE of MXene@PS nanocomposites reached 1081 S/m and 62 dB, respectively. In addition, the introduction of magnetic particles is also a common method to improve the absorption ability of composites to electromagnetic waves. Li et al. prepared the CNTs/RGF/PC nanocomposites by hot pressing molding, after Fe$_3$O$_4$ nanoparticles were deposited on the surface of GO nanosheets (RGF) and assembled with polycarbonate (PC) particles and CNTs. With the synergistic effect of RGF and CNTs, the 4 wt% CNTs/5 wt% RGF/PC nanocomposites showed excellent EMI SE of 44 dB and $SE_{A1}/SE_{E1}$ up to 90%.

This unique segregated structure is easy to adjust the distribution of conductive fillers between the adjacent polymer particles in order to achieve the precise design of 3D conductive networks. However, as a large number of polymer particles are separated by the conductive fillers, it will take disadvantages to the stress transfer in the composites, leading to the decreased mechanical properties of the composites. Therefore, ultra-high pressure and suitable temperature are often required to obtain the desirable mechanical properties.

4.4 | Polymer-based shielding materials with 3D multi-interface networks

The shielding materials with double 3D thresholding networks are composed of two or more kinds of conductive fillers, and the 3D conductive networks constructed by each conductive filler penetrate each other. The shielding performances of double 3D thresholding networks can be enhanced by combining the advantages of the dual conductive networks owing to the synergistic effect of the conductive fillers. Nguyen et al. prepared the Ni/graphene/PDMS foams with 3D multi-interface networks by loading graphene foam with Fe$_3$O$_4$@MXene hybrid nanosheets and impregnation of PDMS (Figure 10A). With the superior $\sigma$ of the inner graphene foam and the outer Fe$_3$O$_4$@MXene foam, 11.35 wt% Fe$_3$O$_4$@MXene/0.61 wt% graphene/PDMS foams reached the outstanding EMI SE of 80 dB in the X-band and 77 dB in Ka-band (26.5–40 GHz), respectively. Weng et al. obtained AgNW/MXene/melamine formaldehyde (MF) sponges with 3D multi-interface networks by impregnation coating of AgNW and MXene nanosheets in MF sponges (Figure 10B). Based on the synergistic effect of the double 3D thresholding networks constructed by 1D AgNW and 2D MXene, AgNW/MXene/MF sponges with a pretty low density of 49.5 mg/cm$^3$ exhibited excellent EMI SE of 52.6 dB. In the previous research of our group, Liang et al. obtained the GNPs/rGO foams with double 3D thresholding networks by the assembly of graphene microplates (GNPs) and graphene oxide followed by the thermal reduction (Figure 10C). Due to the synergistic effect of GNPs and rGO, the $\sigma$ and EMI SE of GNPs/rGO foam/epoxy nanocomposites with double 3D thresholding networks reached 179.2 S/m and 51 dB, with the rGO and GNPs loadings of 0.1 wt% and 20.4 wt%, respectively. Song et al. prepared the rGO/MXene honeycombs with double
3D thresholding networks by growing GO honeycombs on the alumina honeycombs followed by electrostatic adsorption of MXene and thermal reduction. The rGO/MXene honeycombs with double 3D thresholding networks combined the advantages of superior conductivity of both rGO and MXene (Figure 10D). With only 1.2 wt% rGO and 3.3 wt% MXene, rGO/MXene honeycomb/epoxy nanocomposites presented superior $\sigma$ of 387.1 S/m and EMI SE of 55 dB, respectively.

The double 3D thresholding networks can realize the electron conduction potential of each conductive filler, thus enhancing the conductivity of the material and making up for the deficiency of a single conductive filler. This significantly improves the attenuation of electromagnetic waves. However, the double 3D thresholding networks require that each conductive filler should have a good affinity and cause no serious influence on the formation of the other conductive networks.
SUMMARY AND OUTLOOK

In this paper, the fabrication methods and shielding mechanisms of 3D conductive networks, as well as the latest research progress for polymer-based EMI shielding materials in recent years are reviewed. Although the researchers have made great progress in the preparation of shielding materials with 3D conductive networks with low density and outstanding EMI SE, most 3D conductive networks are not suitable for large-scale production due to the harsh preparation conditions, such as high temperature and pressure, the use of toxic substances, high cost, and complex processes. The template method can build a variety of 3D conductive networks based on the developed 3D cellular structures of the template, but the process of removing the template will cause certain damage to 3D conductive networks. The freeze-drying method is easy to control the composition and structures of the 3D conductive networks, but the preparation process takes a long time and the material size is limited. The equipment and reaction conditions of hydrothermal method are relatively simple, but the use of toxic chemical reagents is required. The foaming method has the advantages of low cost and simple preparation process, but the internal cell size is difficult to control uniformly. Therefore, the exploration and development of new preparation and processing methods, simplifying the process, adjusting the product size, and reducing the cost are vitally important for the development of shielding materials with 3D conductive networks.

Although the randomly distributed 3D conductive networks can achieve remarkable EMI SE with a pretty low filler loading, it still needs to further utilize the potential of conductive filler through structural optimization and other methods. The anisotropic 3D conductive networks can reduce the percolating threshold of materials, but it often reduces the mechanical properties in the vertical direction. The segregated structure is easy to control the loading and distribution of conductive fillers, but the polymer matrix is separated by the selectively distributed conductive fillers, which will lead to a decrease in mechanical properties of the composites. The 3D multi-interface net-
works can make up for the deficiency of a single conductive filler and improve the loss ability of electromagnetic waves for the EMI shielding materials. However, the formaldehyde sponge has certain requirements for the affinity of all conductive fillers, which requires better connectivity. Therefore, in order to give full play to the loss ability of electromagnetic waves for the conductive fillers, the reasonable and efficient structural design, synchronous improvement of the mechanical and shielding properties are crucial for the preparation of next-generation polymer-based EMI shielding materials with high performances.

Therefore, the advanced preparation and processing methods (such as 3D printing and microfluidic technology, etc.) should be developed, and the precise design of microstructure optimization based on the 3D conductive networks should be developed to realize the collaborative optimization of various physicochemical properties of the shielding materials. The theoretical models between the material microstructures and electromagnetic shielding performances should be established. Moreover, the relationship between the materials and the electromagnetic waves should be explored to further understand the inherent mechanism of EMI shielding. It will greatly promote the development of the next-generation polymer-based EMI shielding materials with high performances. It is believed that the shielding materials with 3D conductive networks will make great contributions to the development of aerospace, electronic communication, medical treatment, artificial intelligence, and environmental protection in the near future.

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

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