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Advances in the structure and preparation of electromagnetic shielding composites with carbon-based filler polymers

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Abstract. The growth of electronic equipment applications, such as aerospace weaponry, wireless base stations, and 5G communication technologies, has led to serious electromagnetic interference (EMI) and pollution problems. These issues can cause equipment overload and damage. Carbon-based fillers are commonly used as conductive fillers in EMI shielding materials due to their low cost, lightweight and flexible nature, high thermal and electrical conductivity, corrosion resistance, and ease of processing and molding. This paper aims to provide theoretical support for future research by identifying the major trends in the recent advancements of carbon-based filler polymer composites for electromagnetic shielding from the years 2020 to 2024, specifically in filler selection, multifunctional interface design, and preparation methods. Furthermore, this paper discusses the shielding principle of EMI shielding materials, the influence of different kinds of carbon-based fillers on the EMI shielding effect of polymer matrix composites, and the challenges and progress of their preparation methods. In the end, it concludes by proposing future research directions to overcome current technological barriers and further develop EMI shielding materials for industrial applications.

Keywords: Carbon-based Fillers, Electromagnetic Shielding Mechanisms, Composites, Structural Design, Polymer Preparation Methods

1. Introduction

Rapid advances in electronics and communications technology have aggravated concerns regarding the effects of electromagnetic radiation on human health and sensitive equipment [1, 2]. Effective electromagnetic shielding materials are essential to mitigate electromagnetic wave energy and ensure the reliability of sophisticated electronic components [3]. Despite their effectiveness, conventional metallic shielding materials are limited by their high density, high cost and processing complexity. Alternatively, polymer-based conductive composites (CPCs) have gained attention due to their light weight, chemical resistance, ease of processing, and stable shielding properties.

Carbon-based fillers play a key role in polymer-based conductive composites, which is mainly reflected in the improvement of electrical conductivity, electromagnetic shielding properties, mechanical properties and so on. There are a wide variety of carbon-based fillers, mainly including carbon fibers, carbon nanotubes, graphene, and MXene, which have different characteristics in terms of mechanical properties, electrical conductivity, and thermal conductivity [4], and thus allow for the selection of the most suitable filler according to the requirements of a specific application.

This paper reviews recent progress in carbon based filler polymer composites for electromagnetic shielding between 2020 and 2024. Specifically, it includes filler selection, multifunctional interface design and preparation methods, aiming to provide theoretical support for future research by identifying key development trends and addressing potential challenges. This work is expected to promote the development of both light, high electromagnetic interference (EMI) shielding efficacy and low-cost EMI shielding materials, with wide-ranging implications for high-power, high-density and highly integrated technologies.

2. Electromagnetic Shielding Mechanism

EMI shielding utilizes the electrical or magnetic loss of conductive or magnetic materials to reflect or absorb incident electromagnetic waves, thereby limiting their propagation and protecting the interfered object. According to Schelkunoff plane wave transmission line theory, there are three main mechanisms encountered by electromagnetic waves in shielding materials: reflection loss, absorption loss, and multiple reflection loss, as shown in Figure 1 [5]. When an incident electromagnetic wave is projected onto a material's surface, a portion of the wave reflects at the surface, resulting in reflection loss. The remainder of the electromagnetic wave is then transferred into the material, accompanied by magneto-thermal conversion, which dissipates the electromagnetic wave, a process known as absorption loss. Electromagnetic waves will be reflected several times in the material's internal interface, resulting in continuous attenuation, also known as multiple reflection loss. Eventually, unabsorbed electromagnetic waves will pass through the shielding material [6].

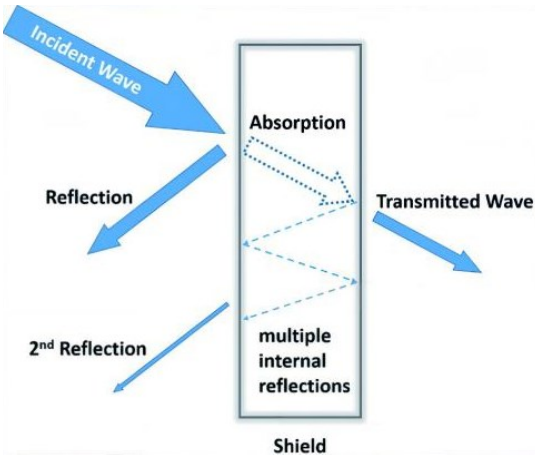

Figure 1. Schematic Diagram of Electromagnetic Shielding Mechanism

The consumable materials can be categorized into electrical losses, electromagnetic losses, or a mixture of both. The consumption of electrically depleted materials is based on two fundamental principles: electromagnetic natural resonance loss and hysteresis loss. Lossy materials are typically affected by dielectric electrons, ionic polarization, and interface polarization, which cause the attenuation of electromagnetic waves. They can be classified into two types: dielectric loss and resistive loss. The dielectric-type wave-attracting material experiences dielectric electrode relaxation loss, while the resistive loss-type wave-attracting material experiences a resistive loss effect. Currently, there are more studies on loss-type attracting wave materials in terms of electromagnetic shielding, and the basic principle of electromagnetic wave propagation is also more complex. Electromagnetic shielding materials function through two mechanisms: reflection loss and dielectric loss [7].

3. Types and Properties of Carbon-based Fillers

This section examines various types of carbon-based filters, analyzing their structural features and their impact on electromagnetic shielding performance. The analysis provides guidance for designing efficient electromagnetic shielding materials.

3.1. Carbon Fiber (CF)

Carbon fiber is a one-dimensional structural carbon material, which is a raw material fiber that will contain more than 90% carbon [8]. Carbon fiber, based on its light weight, high strength and excellent thermal stability, has received much attention in recent years for its dual functions of load bearing and electromagnetic shielding [9-12]. Carbon fiber composite EMI shielding film has excellent EMI shielding performance in the X-band range of ~32 dB, and with a thickness of only less than 1 mm, it has great potential for both commercial and military communication technology applications. Li et al [13] Carbon fibers were coated and encapsulated with magnetic nanoparticles in polyimide (PI) resin used as a shielding material, as shown in Figure 2. The composite material exhibits excellent EMI shielding efficiency of ~32 dB in the X-band at a thickness of 0.16 mm. And an EMI shielding efficiency of ~62 dB can be achieved by stacking 6 layers to achieve a nanocomposite film that shields 99.999937% of incident electromagnetic waves with a thickness of only 0.98 mm. Given its excellent overall performance, this ultra-thin and low-cost biomass-derived film has high EMI shielding prospects in practical applications.

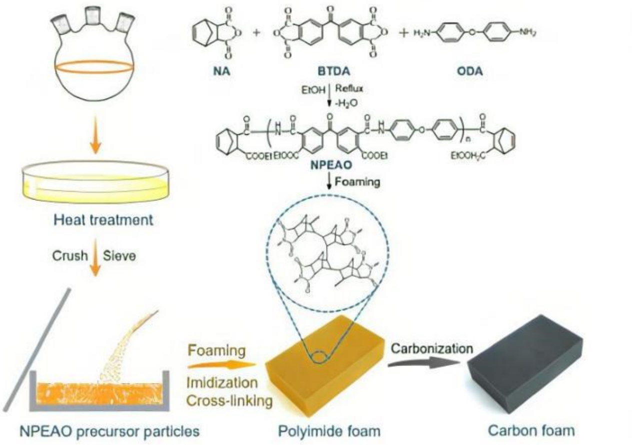

Figure 2. Schematic Diagram of the Preparation of Carbon Foam Derived from Thermoset PI Foam [13]

Wang et al. adopted a two-step pyrolysis strategy to decompose the resin matrix and remove carbon residues without damaging the internal structure of the carbon fibers, followed by surface modification to facilitate the reconstruction of resin composites with regenerated carbon fiber reinforced phenolic resin. The composite material achieved a maximum EMI shielding efficiency of 70.5 dB in the X-band [14].

3.2. Carbon Nanotubes

Carbon nanotubes (CNTs), with their unique hollow tubular structure, are anisotropic nanomaterials with high modulus and strength, combining outstanding advantages such as excellent electrical conductivity, high strength, large aspect ratio (usually >1000), and a specific surface area [15, 16]. Since the conductivity of carbon nanotubes is much higher than that of polymers, by modulating the structure and interfaces, the demand for higher conductivity, uniform dispersion, and high electromagnetic shielding effectiveness can be met with lower filler volume, which is effective in terms of electrical conductivity, wave-absorbing shielding, and mechanical strength [17, 18].

Tao et al. [19] used highly elastic carbon nanotube foam to prepare heat-resistant carbon nanotube/polyimide films, designing a sandwich foam with carbon nanotube foam in the core and carbon nanotube/polyimide films in the outer layer. The test results show that by adjusting the film thickness, the compressive strength of the foam reaches 88.2 kPa, the electrical conductivity is 675 S/m, and the electromagnetic shielding effectiveness in X-band is 88.3 dB.

The doping of conductive filler carbon nanotubes (CNTs) into thermoplastic polyimide (TPI) resin not only enhances its toughness, but also confers good electrical conductivity and anti-electromagnetic interference, enabling it to operate stably for long periods of time in extreme environments, such as high-vacuum, high-temperature, and high-pressure, etc. Zhang et al. utilizing graphene crosslinked carbon nanotubes, prepared a thermally conductive, electrically conductive and elastic TPI composite as shown in Figure 3 [20]. The welding effect of graphene transformed the discontinuous carbon nanotube sponge into a continuous 3D network. Under compression, the deformation of the 3D network leads to close collision and contact of the nanotubes in the TPI composites, which facilitates electron transport between adjacent carbon nanotubes, resulting in good electrical conductivity.

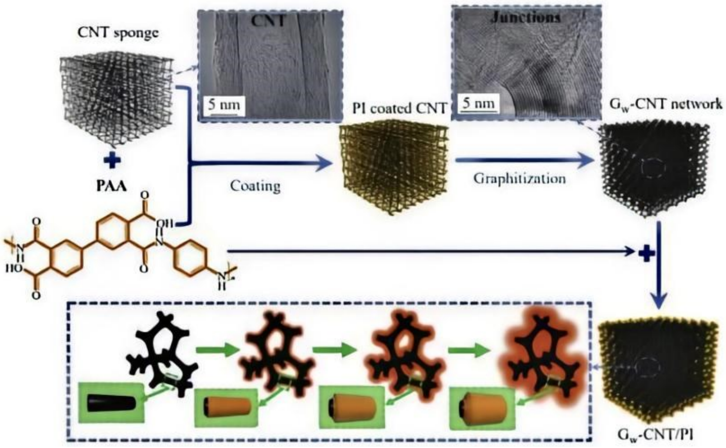

Figure 3. Diagram of the Preparation of Three-dimensional Elastic Nanotube Sponge/Polyimide (Gw-CNT/PI) Composites [20]

The shielding ability and conductivity values of CNTs, however, are influenced to a greater extent by the processing techniques used. The larger the L/D ratio of CNT, the better the conductivity, but the more difficult it is to process. When the L/D ratio is too large, the CNT is difficult to disperse in the polymer matrix, resulting in a decrease in the volume resistivity of the conductive material instead [21]. In order to obtain better shielding properties, melt blending is usually chosen to ensure uniform dispersion of fillers. Unlike other techniques, filler materials like carbon nanotubes do not rupture during melt mixing, which gives the composites better electrical conductivity and thus enhanced electromagnetic shielding properties.

3.3. Graphene

Graphene is a hexagonal honeycomb-shaped two-dimensional carbon nanomaterial composed of sp2 hybridized orbitals, which has the advantages of a large specific surface area, good electrical conductivity, and high mechanical strength, and has been widely used in the fields of electromagnetic shielding, flexible sensors, and electronics [22]. The free motion of graphene makes graphene ultra-highly conductive, which enables high-strength polymer composites at relatively low graphene loadings. At the same time, it has large π-bonds that can run through the entire layer of polyatoms and excellent dielectric dissipation [23].

Shahabadi et al [24] prepared WPU/LMG nanocomposites via a facile aqueous-phase method using lignin non-covalently modified graphene (LMG), which was combined with waterborne polyurethane (WPU). The results showed that the WPU/LMG nanocomposites possessed excellent electrical

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conductivity properties with conductivity up to 0.276 S/m. Ding [25] introduced a graphene foam (GF) composite by chemical vapour deposition, containing a flexible polymer matrix PDMS and magnetic Fe3O nanoparticles. The ternary composite has an absolute EMI SE of up to 71 dB owing to the abundant internal interfaces, the high conductivity of the 3D interconnected graphene, and the magnetic loss of Fe3O.

The graphene with two-dimensional sheet structure has excellent electrical conductivity, which be applied to electromagnetic interference shielding materials, but low-cost and high-quality graphene is still difficult to be produced on a large scale due to the complexity of the production technology as well as high cost and other problems. Therefore, the search for a simpler, more economical and efficient production technology is the key to the solution to the problem of large-scale production of graphene.

3.4. MXenes

MXene is an emerging 2D transition metal carbide/nitride with excellent electrical conductivity, high mechanical stability, a variety of functional groups and large interlayer space with significant energy storage capacity[26-27]. MXene is easily dispersed in water due to its hydrophilic groups. Its unique properties make MXene a valuable material. When MXene is compounded with elastomers, the nanocomposites can impart functionality. Especially in electromagnetic interference shielding and sensing, MXene shows extraordinary performance, among which Ti3C2Tx is one of the most common MXene materials [28].

MXene 2D nanomaterials were homogeneously dispersed within non-polar polymers to impart excellent conductive and EMI shielding properties to polymeric materials.Jiao [29] used electrostatic interactions to combine MXene with PS microspheres of controllable particle sizes to form core-shell structured MXene@PS conductive masterbatch, which was then melt-composited with PP to prepare the multifunctional polypropylene composite MXene@PS/PP. The results showed that the composite exhibited an average EMI shielding performance of more than 21.87 dB over the frequency range of 8.2-40.0 GHz at an MXene content of 13.3 wt%.

Tang et al. [30] prepared MXene/polypyrrole (PPy)@bacterial cellulose (BC) thin films (MPB) with high conductivity by vacuum filtration method. The results showed that the electrical conductivity and X-band electromagnetic shielding effectiveness (EMI SE) reached the maximum values of 1209 S/cm and 63.89 dB at the ratio of MXene and PPy@BC of 3:1, respectively.

The high conductivity of the MXene layer makes it useful for the fabrication of polymer matrix composites for EMI shielding. In addition, the nanoscale thickness of MXene makes these composites ideal for flexibility and low-thickness EMI shielding applications.

4. Construction of the Multi-interface Architecture

This section explores various methods for constructing multi-interface structures, such as vacuum-assisted filtration, melt blending, solution mixing, in-situ polymerization, foam processing, and layer-by-layer casting. The aim is to provide technical support and references for preparing composite materials with excellent electromagnetic shielding performance.

4.1. Vacuum-assisted Filtration

Vacuum-assisted moulding is a commonly used method for preparing thin films The basic principle is that under vacuum conditions, by using a filter membrane or template, nanoparticles or molecules in solution are filtered or deposited on the substrate surface under negative pressure to form the desired thin films or nanostructures.

Shi [31] prepared MXene/WPU composite films by vacuum-assisted filtration. The MXene dispersion was mixed with 32 wt% solid WPU solution, stirred for 30 min and vacuum filtered on a sand core funnel. The filter film was then dried and removed to obtain MXene/WPU composite films with different MXene contents (40 wt%, 60 wt%, and 75 wt%). When the proportion of MXene was 60%, the composite achieved the best EMI shielding quality of 24 dB in the 8.2-12.4 GHz band.

The composites prepared by this method have high densities and controllable morphology, with fast preparation speeds and the ability to prepare large-area films. However, it is not applicable to some heat-sensitive fillers and polymers due to the high pressure and temperature required in the preparation process.

4.2. Melt Blending Method

The melt blending method is a method of preparing composites by fully mixing fillers and polymers in a molten state and then by extrusion or injection moulding. In the process, polymer molecules diffuse and mix with each other at high temperature to form a uniform composite structure.

Hu et al. [32] used the melt blending method to homogenize MCB with polyvinyl chloride (PVC) in different proportions at high temperature. Subsequently, the mixture was extruded, granulated and injection moulded to obtain polyvinyl chloride/carbon black (PVC/CB) composites. The results showed that the electromagnetic shielding performance of PVC/MCB composites prepared by the melt blending method was significantly better than that of PVC/MCB composites prepared by other methods.

This method is suitable for large-scale production owing to the simplicity of operation and low process cost, but may lead to filler agglomeration and insufficient interfacial adhesion due to the compatibility limitations of fillers and polymers in the molten state.

4.3. Solution Blending Method

Solution blending is used to prepare composites by mixing carbon-based fillers with polymer prepolymer dissolved in a solvent, followed by solvent evaporation or coagulation precipitation. Jin et al, [33] prepared VMQ/GFBT/MWCNT nanocomposites with three-dimensional spatial conductive networks by mixing vinylmethylsilicone rubber (VMQ), graphene nanoplatelets/Fe3O4@BaTiO3 hybrids (GFBTs), and MWCNTs using a solution blending method. It was shown that the maximum electromagnetic shielding effectiveness of the composites reached 33.3 dB in the range of 1.0-20.0 GHz.

This method is a simple and convenient preparation process, which can better control the dispersion of the filler, but the selection of solvent and the evaporation process may cause pollution to the environment.

4.4. In-situ Polymerisation

In-situ polymerisation is a method of preparing composites by carrying out a polymerisation reaction at the filler surface or filler/polymer interface, where the polymer feedstock is polymerised directly onto the filler surface or filler/polymer interface. Kazakova MA et al.[34] proposed a modified polyethylene-based multi-wall carbon nano-hydrocone tubes (MVCNT) based on in-situ polymerisation of ethylene to produce modified polyethylene-based multi-wall carbon nano-hydrocone tubes (MVCNT). It is shown that for Co/MWCNT-PE composites, a very high reflection loss (RL)-55 dB can be displayed when the filler amount is 12% and 1.7%, respectively.

This method can achieve chemical bonding between filler and polymer and improve the mechanical properties and interfacial stability of composites, but the selection of filler and polymer is more demanding.

4.5. Layer-by-Layer Pouring Method

The layer-by-layer casting method involves dispersing carbon-based fillers in a polymer matrix by doping layer by layer. This technique allows precise control of the distribution and content of the filler, resulting in a better hierarchical structure.

Sheng et al. [35] prepared Fe3O4@reduced graphene oxide (rGO)/MWCNT/waterborne polyurethane (WPU) multilayer composites by a layer-by-layer casting process, and the reflectance coefficient R of the composites was only 0.27 when the electromagnetic shielding effectiveness of the composites was 35.9 dB, and the composites achieved good co-ordination of the impedance match and the shielding efficiency by adjusting the rGO content in Fe3O4@rGO. This composite achieves a good coordination between impedance matching and shielding efficiency by adjusting the content of rGO in Fe3O4@rGO, which proves that the dual absorption loss can be achieved by regulating the directional arrangement of the conductive/conductive fillers in the matrix.

The composites prepared by this method have better interfacial bonding and relatively homogeneous dispersion, but the preparation process is cumbersome and the production efficiency is low.

5. Conclusion

The currently developed carbon-based filler polymer-based electromagnetic interference (EMI) shielding polymer composites are characterized by light weight, high strength, corrosion resistance, easy processing, and EMI shielding performance, which are gradually replacing the traditional metal-based EMI shielding materials. In addition, this composite material has been able to basically meet the needs of aerospace weaponry, wireless base stations and 5G communication technology and other areas of application. However, although carbon material as a conductive filler in polymer substrates can achieve efficient EMI shielding effect, there are some problems in composites such as easy agglomeration of carbon material and uneven dispersion due to the fact that carbon material belongs to the inorganic phase while polymer belongs to the organic phase. Therefore, the homogeneous dispersion and stability of carbon fillers in the polymer matrix still need to be improved to ensure their optimal effect in composites. The next research needs to break through the bottleneck in terms of interfacial design aspects (e.g., interfacial layer introduction, surface modification) to construct an efficient conductive network, and lightweight and multifunctional compatibility to provide a more reliable theoretical basis for the preparation and development of polymer-based EMI shielding composites.

Additionally, the processing process and cost of the existing carbon-based filler polymer-based EMI shielding composites need to be further improved to increase the production efficiency and reduce the manufacturing cost. Therefore, future research should focus on improving the dispersion and interfacial compatibility of carbon fillers, optimizing the processing of composites, exploring the application of novel functional materials, and developing high-performance and low-cost polymer-based EMI shielding composites. Through continuous innovation and research, the development of this field can be further pushed forward to make a greater contribution to combating electromagnetic interference and improving the performance of electronic devices.

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