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Review

The Synergistic Enhancement of Electromagnetic Interference Shielding Polymer Composites using Carbon Nanofiber-Metal Oxide Hybrid Fillers

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ABSTRACT. In this modern era, humans cannot be separated from utilizing electronic devices. The unwanted noise or disturbance problem in electronic devices is majorly caused by Electromagnetic Interference (EMI). Recent technology and materials are being developed to manufacture EMI shielding composites. Carbon nanomaterial-based composites have become a very attractive material because of their lightweight, incredible mechanical, thermal, and conductivity properties. Carbon nanofibers (CNFs) and metal oxides are promising combination materials that reveal unique properties of both components to enhance shielding effectiveness. This combination of CNFs and metal oxides results in a synergistic effect, where the conductive CNFs enhances the reflection of EMI, and the metal oxides contribute to the absorption and attenuation of the waves. In this review article, the authors focus on the extensive discussion about the synthesis method of producing CNFs, some studies that combine CNFs and metal oxide, and some reports about using CNFs-metal oxide as a hybrid filler in the EMI shielding composites.

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INTRODUCTION

As technology advances rapidly, electronic devices have become crucial in every aspect of human life. The rapid advancement of technology in the innovation of various electrical and electronic devices has created environmental radiation pollution, such as electromagnetic interference, radio frequency interference, and electronic noise. The unwanted electromagnetic waves produced by electronic devices can interfere with the normal functioning of nearby electronic devices (or components). This phenomenon is called electromagnetic interference (EMI) (Sankaran *et al.*, 2018). Therefore, these EMI signals radiate from other equipment or from the component experiencing the interference (Bora *et al.*, 2021).

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EMI happens when uncontrollable electromagnetic energy is transmitted between electronic devices via radiated or conducted pathways. It depends on the type of coupling or transmission path used. Radiated interference transfers energy through the air, while conducted interference transmits it through cables (Wang *et al*., 2018). EMI from one device can disrupt and impact the performance of other electronic appliances, devices, and systems, reducing their efficiency, causing data corruption, and device malfunctions (Kruželák *et al*., 2021; Hamouda and Amneenah, 2024). The EMI issue could lead to severe damage to communication systems (Solkin, 2021), disruption of aircraft control systems (Leuchter *et al*., 2021), automotive safety systems (Mitilineos *et al*., 2021), medical devices (Chan *et al*., 2020; Rahimpour *et al*., 2021), and electronic devices (Shan *et al*., 2022). Prolonged exposure to these interferences can also impact human health, potentially leading to symptoms such as headaches, irritability, insomnia, fatigue, or difficulty concentrating, which might contribute to more serious health issues (Panahi-Sarmad *et al*., 2022).

This EMI problem can be solved by developing materials capable of shielding electronic devices and systems. These materials are often designed with circuits or filters and metal coatings to enhance the device's shielding (Crespo *et al*., 2014). Metals remain the dominant material in commercial EMI shielding applications. Highly conductive metals such as copper, aluminum, and steel are often used due to their effectiveness in blocking and reflecting electromagnetic waves. However, lightweight alternative materials are also needed. Researchers have developed composites, metal alloys, and foams to meet this demand, which can be enhanced by incorporating carbon-based fillers, metals, or other conductive fillers. While these materials offer reduced mass and design flexibility, their shielding effectiveness (SE) is generally lower than that of metals, and they often lack sufficient mechanical strength for structural applications. Therefore, innovation is needed to develop and study advanced materials that offer a combination of high shielding effectiveness, strong mechanical properties, and lightweight (Pandey *et al*., 2020). This review discusses the potential use of carbon nanofibers (CNFs) and metal oxides as hybrid fillers in composites for electromagnetic interference (EMI) shielding applications. This discussion covers the fundamentals of EMI shielding, several methods for synthesizing carbon nanofibers (CNFs), methods for preparing CNFs and metal oxides as hybrid fillers, and their use by incorporating them with polymer composites in EMI shielding applications. The goal is to explore the material systems that exhibit high EMI shielding effectiveness, high electrical conductivity, good mechanical strength, and lightweight properties.

FUNDAMENTALS OF EMI SHIELDING

As outlined in the introduction, EMI refers to interference caused by electromagnetic signals that disrupt the performance of a transmission line, device, or system (Hassija *et al*., 2021). Minimizing EMI involves reducing or removing interference using techniques like shielding and filtering, which lower the intensity of magnetic and electric fields (Wang *et al*., 2018). The mechanism of EMI protection is primarily composed of reflection, absorption, and multiple reflections (Aswathi *et al*., 2019). The first step is the reflection process, which causes the mobile charge carriers to interact with the electromagnetic waves. Metals are commonly used as a material for EMI shielding because they provide free electrons that are able to interact with electromagnetic waves (Jagatheesan *et al*., 2014). EMI shielding mainly occurs through reflection in materials with high conductivity. Although conductivity is not essential for EMI shielding, it enhances the material's ability to reflect electromagnetic waves. The second step mechanism is absorption, which is influenced by the presence of electric or magnetic dipoles interacting with electromagnetic radiation. There are two necessities for material: with a high dielectric constant (for producing electric dipoles) and high magnetic permeability (for producing magnetic dipoles through absorption) (Wei *et al*., 2019). The third mechanism is multiple reflections. This process occurs at material surfaces or interfaces consisting of sandwich layers or composites with fillers (Xie *et al*., 2019). Figure 1 displays the schematic illustration of the EMI shielding mechanism process.

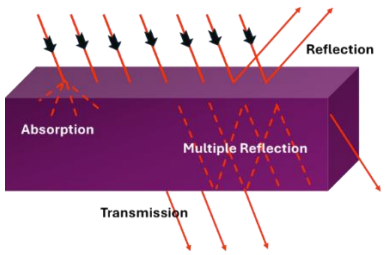

Figure 1. Illustration of the EMI shielding mechanism process.

Losses resulting from the EMI shielding mechanism process (reflection, absorption, or multiple reflections) are typically measured in decibels (dB). Their total losses are referred to as the shielding effectiveness (SE). Among these, absorption loss goes up proportionally with the thickness of the shielding material (Chung, 2001). EMI SE refers to a material's ability to block microwave (MW) and radiowave (RW) radiation by acting as a barrier preventing penetration. It is crucial to distinguish EMI shielding from magnetic shielding, which reaches low-frequency magnetic fields up to 60 Hz. Since all electronic devices emit micro- and radio-wave radiation, effective EMI shielding is essential for minimizing interference (Orasugh and Ray, 2023). Shielding effectiveness values are measured based on several parameters using experimental vector network analyzer techniques. Equations 1 – 5 are the equations for analyzing the total SE and its absorption and reflection components (Yazdi *et al*., 2023).

$$SE_T = 10\ log\frac{1}{T} \quad (1)$$

$$SE_R = 10\ log\frac{1}{(1-R)} \quad (2)$$

$$SE_A = 10\ log\frac{(1-R)}{T} \quad (3)$$

$$R = |S_{11}|^2 \quad (4)$$

$$T = |S_{21}|^2 \quad (5)$$

R is the reflection coefficient, and T is the transmission coefficient. S_{11} indicates the voltage reflection coefficient at the input port when the output ports are matched. S_{21} indicates the reverse voltage gain. S_{22} is the voltage reflection coefficient at the output port when the input ports are matched. SET stands for total shielding effectiveness, while SE_A and SE_R stand for absorption and reflection shielding effectiveness. The effective EMI shielding components need excellent electrical, magnetic, or dielectric properties. The shielding effectiveness is highly influenced by the material's electrical conductivity and dielectric behavior. Materials with higher conductivity typically exhibit greater permittivity and lower impedance, which enhances their ability to reflect electromagnetic waves efficiently (Li *et al*., 2024).

CARBON NANOFIBERS AS FUNCTIONAL MATERIALS

Metal-based materials, such as copper, nickel, aluminum, silver, and gold, are applicable for EMI shielding applications due to their high efficiency in reflecting electromagnetic waves. This effectiveness is primarily due to the abundance of free electrons, which results in excellent electrical conductivity (Pandey *et al*., 2020). Materials with high dielectric constants generate electric dipoles, such as $BaTiO_3$ (barium titanate), Fe_2O_3 (ferric oxide), and ZrO_2 (zirconium dioxide). These high-permeability materials contribute to magnetic dipoles, resulting in ideal absorption, such as Fe_3O_4 (magnetite), ferrite, and superpermalloy. However, the use of metals in modern devices is limited by their high weight density, lack of flexibility, susceptibility to corrosion, and processing complexity (Kruželák *et al*., 2021). Minimizing the weight of EMI shielding is crucial in aerospace applications. These challenges can be addressed by using lightweight materials, such as carbon-based hybrid polymer composites, or by developing interfacial structural designs that integrate carbon-based materials with lightweight metal components (Morales *et al*., 2023). These efforts have been directed towards developing hybrid polymer composites to address the shortcomings of metal-based EMI shielding materials. Hybrid polymer composites are increasingly viewed as highly attractive EMI shielding materials due to several key advantages, such as lightweight, enhanced EMI shielding performance (Chandra *et al*., 2019), tunable properties (Sinha *et al*., 2020), improved electrical and mechanical properties (Ali *et al*., 2024), cost-effectiveness (Ismail *et al*., 2022), high flexibility and ease of processing (Lee *et al*., 2022), versatility (Jawaid *et al*., 2022), and flexible functionality (Atinafu *et al*., 2021).

Carbon nanomaterial-based composites are widely used as conductive fillers in various applications, including EMI shielding, due to their high aspect ratio. Carbon nanomaterials consist of various allotropes, such as zero-dimensional (0D) (quantum dots), one-dimensional (1D) (carbon nanotubes (CNTs) and carbon nanofibers (CNFs)), two-dimensional (2D) (graphene), and three-dimensional (3D) (fullerene) (Kumar *et al*., 2021). These carbon nanomaterials are conductive due to their delocalized π-conjugated electronic structure. This results in good electronic properties, such as high electron affinity, low ionization potential, and high conductivity. To improve specific performance, modifications or doping can be done with supporting fillers (Park *et al*., 2020). Carbon

nanofibers (CNFs) often show advantages over other carbon allotropes regarding EMI shielding. Carbon nanofibers offer simpler synthesis methods, including chemical vapor deposition (CVD) or electrospinning, than those required for graphene or carbon nanotubes (Zhou *et al*., 2020). CNFs tend to have fewer aggregation or entanglement issues during synthesis. This is due to their higher surface area, approximately 20 to 300 m^2/g, making them easier to handle and process into composites (Santos *et al*., 2024). Precursor materials and processes for CNFs production are often lower in cost, contributing to cost-effective and scalable synthesis. This allows for larger-scale production (Ruiz-Cornejo *et al*., 2020). CNFs are easier to integrate into various polymers due to their flexibility and compatibility with different processing techniques, such as melting, blending, and sonication (Feng *et al*., 2014).

Structurally, CNFs consist of modified graphene layers. These nanofibers can have three structural forms: tubular, herringbone, or platelet (Shoukat and Khan, 2022). CNFs consist of irregular linear filaments that are sp2 hybridized and have an aspect ratio of up to 100:1. The orientation of the carbon layers within CNFs can affect their mechanical properties (Yadav *et al*., 2020). The sp2 hybridization of carbon atoms in carbon nanofibers (CNFs) also plays a crucial role in determining their unique physical, mechanical, electrical, and thermal properties (Maher *et al*., 2025). The CNFs structure consists of strong carbon-carbon bonds chemically stable within the graphene layers. Each carbon atom forms three sigma bonds with its surrounding atoms, creating a planar hexagonal arrangement with strong in-plane covalent bonds (Razeghi, 2019). This bonding provides high mechanical strength, stiffness, and flexibility because the planar hexagonal carbon structure can be slightly bent without breaking. Each carbon atom in the sp^2 hybridized structure retains one unhybridized p orbital, which overlaps with adjacent p orbitals to create a delocalized π electron cloud that extends above and below the graphene-like layer (Kumar *et al*., 2022; Urade *et al*., 2023). This delocalized π electron system allows CNFs to conduct electricity efficiently, making them highly conductive for EMI shielding composites, sensors, and energy storage devices. CNFs are synthesized using several primary methods with modified techniques in controlling the fibers' properties, size, and structure (Ruiz-Cornejo *et al*., 2020).

Electrospinning

The electrospinning technique was first demonstrated in 1747 by Abbé Nollet and later patented by John Cooley and William Morton in the early 1900s. Besides, the synthesis of nanofibers by the electrospinning method was first developed by Anton Formhals in the late 1934 (Keirouz *et al*., 2023). Electrospinning has become the most popular method to produce CNFs because of its efficiency, facile, and potential opportunities for the commercial-scale production of CNFs (Wang *et al*., 2023). Electrospinning becomes a distinct method that utilizes an electrostatic field to produce ultrafine fibers. This technique involves a simple setup with mainly components: a fiber collector, a high-voltage power supply, a syringe pump, and a spinneret. This method begins with preparing the polymer, which is dissolved in a volatile solvent. The polymer is expelled from a syringe at a controlled rate using a syringe pump.

The applied electrostatic force makes a gap of positive and negative charges within the liquid. It happens with charges matching the needle's polarity moving toward the surface. This leads to the production of charged polymer drops at the needle's tip. The intensity of the electrostatic field will expand as the surface charge density on the droplet increases. It intensifies charge repulsion on the liquid's surface. This expansion of the droplet's surface area reduces the repulsion, causing the droplet to form a Taylor cone. Eventually, the electrostatic repulsion exceeds surface tension. It makes producing a jet that swiftly moves toward the collector. During this movement, the polymer solution stretches and undergoes a whipping motion, while the solvent evaporates, creating ultrafine fibers (Bonakdar and Rodrigue, 2024). Figure 2 displays the illustration of the electrospinning process scheme.

Polymers commonly used as precursors for synthesizing CNFs through the electrospinning method include polyacrylonitrile (PAN), polyvinylpyrrolidone (PVP), polystyrene (PS), polyimide (PI), polyvinylidene fluoride (PVDF), polyvinyl alcohol (PVA), poly(methyl methacrylate) (PMMA), pitch, lignin, and cellulose (Keshavarz *et al*., 2022). Key factors that influence the electrospinning process are polymer solution characteristics, synthesis conditions, and environmental (ambient) factors (Haider *et al*., 2018). The polymer solution determines the morphology and diameter of electrospun fibers. The key factors for polymer solutions are molecular weight, viscosity, polymer concentration, conductivity, and surface tension. Low polymer concentrations result in beaded nanofibers due to chain breakage, while increased concentration and viscosity lead to uniform fibers. Optimal viscosity is needed for proper fiber formation, with low viscosity causing beads and high viscosity making extrusion difficult. Surface tension impacts jet stability, and lower tension aids fiber formation. Polymer solution

conductivity affects diameter distribution, leading to smaller nanofibers. The solvent should completely dissolve the polymer and have a medium boiling point. Key factors of processing conditions consist of needle tip-to-collector distance, voltage, flow rate, and needle diameter that will influence nanofiber morphology. The collector functions as an electrode, impacting the fiber accumulation. The distance between the needle tip and the collector has an impact on the fiber diameter and morphology. Voltage will control the electric field strength, as the flow rate affects the jet speed. Environment (ambient) factors could be caused by humidity and temperature, which impact the fiber size. Humidity can affect the solidification process based on polymer properties (Ibrahim and Klingner, 2020; Mailley *et al*., 2021; Guo *et al*., 2022; Tayebi-Khorrami *et al*., 2024).

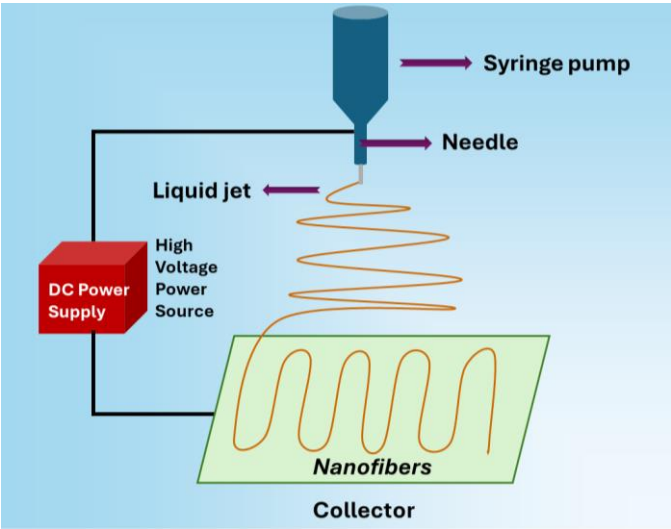

Figure 2. Illustration of the electrospinning scheme.

Polyacrylonitrile (PAN) is commonly used for nanofiber production because of its straightforward carbonization process. Qanati *et al.* (2022) studied the electrospinning synthesis of CNFs using a precursor polymer of polyacrylonitrile (PAN). PAN was dissolved in dimethylformamide (DMF) at 8% concentration, and various voltages (10, 12, 14, 16, 18 kV) were applied. The characteristics of PAN fibers were examined at different concentrations (6, 8, 10, 12, 14, 16% w/w) in DMF to optimize the solution for electrospinning. The process involved the distance of the collector and needle tip (13 cm), flow rate (0.8 ml/hour), and total duration (3 hours). The needle size used in electrospinning CNFs typically varies, commonly ranging from 25-gauge to 18-gauge (Hwang *et al*., 2021; Albetran, 2023).

After electrospinning completely, the fibers were stabilized by heating at 300 °C and carbonized in a quartz chamber under argon flow, with final temperatures ranging from 800 to 1600 °C. The results indicated that at 12 kV, the electrical force was inadequate to hold surface tension, resulting in short fiber nanoparticles (with an average diameter of around 820 nm). At the voltages of 14 kV and 16 kV, fibers were produced without impurities (lower average diameters 324 nm and 221 nm). At voltages up to 16 kV, excessive jet whipping and unstable formation of fiber are observed. At the voltage of 10 kV, it did not produce any jets. The optimal voltage range for uniform, bead-free fibers was found to be between 14 and 16 kV. For solutions with 6%, 8%, and 10% (w/w) concentrations, the average fiber diameters were 205 nm, 271 nm, and 446 nm, respectively. At 12% and 14% (w/w), micro-sized fibers with average diameters of 1245 and 963 nm were obtained. The electrical force could not overcome surface tension at a 16% (w/w) concentration, resulting in significant agglomeration of nanoparticles (Qanati *et al*., 2022). The electrospun PAN nanofibers exhibited different colors at each stage: white after synthesis, brown after stabilization at a temperature range of 250-300°C, and black after carbonization at 700°C (Ospanali *et al*., 2020).

Electrospinning is a flexible technique for fabricating materials with specialized functionalized morphologies suitable for various applications. The resulting nanofibers offer superior features, such as ultrafine diameters, high aspect ratios, large specific surface areas, and the ability to incorporate diverse material compositions. Furthermore, by fine-tuning the electrospinning parameters, the nanofibers' surface characteristics and internal structure can be precisely controlled, tailoring their physical and chemical properties. Therefore, incorporating

electrospinned nanofibers with tunable features into EMI shielding materials has great potential for achieving enhanced performance (Li *et al*., 2024). Carbon nanofibers produced by electrospinning offer greater flexibility and efficiency for large-scale nanofiber fabrication. Furthermore, these nanofibers' diameter, length, and structural stability can be precisely controlled by adjusting the process parameters, enabling environmentally friendly production (Yang *et al*., 2023).

Chemical Vapor Deposition

The first report on CNFs synthesis using the Chemical Vapor Deposition (CVD) method was made by researchers Oberlin et al. in 1975. This marked the beginning of rapid progress in carbon nanomaterial research (Wang *et al*., 2019). The CVD process typically involves using a furnace chamber where all gases are first evacuated to prevent oxidation from atmospheric oxygen. Carbon precursor gases are mainly hydrocarbons, along with the flow of inert gas (argon or helium). The furnace is heated to decompose the gases, causing them to react with a metal catalyst and deposit carbon onto the substrate. Residual hydrogen is then removed from the chamber. The decomposition of vapors occurs at temperatures between 600 and 1200 °C, where hydrocarbons break down into hydrogen and carbon. The resulting carbon will dissolve into the metal catalyst, and hydrogen will evaporate (Saputri *et al*., 2020).

The CVD offers the benefit of controlling the diameter, alignment of the fiber, and crystallinity by precisely adjusting the synthesis conditions. CNFs are produced through catalytic CVD using hydrocarbons (propane, benzene, ethylene, acetylene, or carbon monoxide). The carbon will grow over a metal surface (Ni, Fe, Co, Au) or using a metal alloy catalyst. The catalyst will interact with the introduced gas phase, and the compound will deposit on a substrate. The reaction occurs at temperatures between 500 – 1500 °C (Lee and Hyun, 2016; Manawi *et al*., 2018; Lv *et al*., 2021). Parameters of the synthesis of CNFs using the CVD process are catalysts, selective hydrocarbon, selective catalysts, temperature, and time for reaction (Ghaemi *et al*., 2018). Figure 3 shows the illustration of the CVD process.

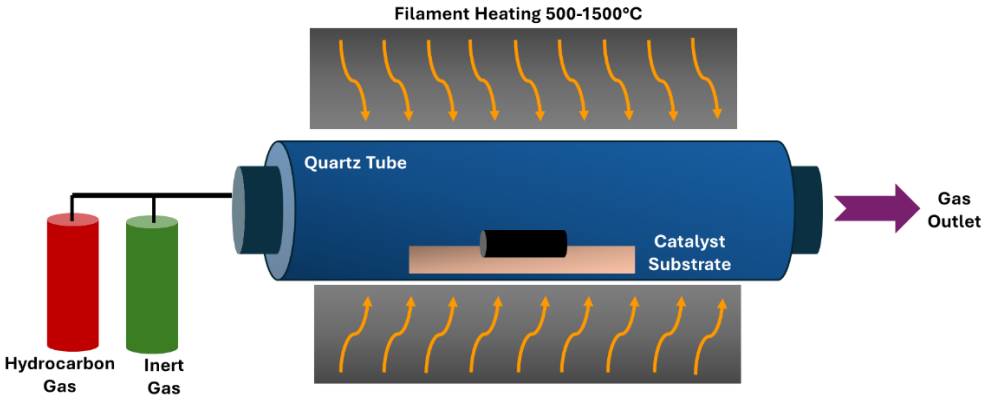

Figure 3. Illustration of the CVD process.

Kim *et al*. (2024) synthesized CNFs through a CVD process with a Ni-Pd alloy catalyst at 750 °C in an atmospheric-pressure CVD system. The catalyst, coated onto a substrate, was oxidized at 500 °C for 10 minutes to remove impurities. Ethylene was used as the carbon source, and the catalyst was activated in a 40 volume% H_2 atmosphere at 750 °C and 1 atm. During synthesis, ethylene was introduced at a flow rate of 100 sccm, while Ar and H_2 were continuously supplied. After 10 minutes of CNFs growth, the ethylene and H_2 flows were halted, and the reactor was cooled under an Ar atmosphere. The obtained CNFs exhibited rounded tips without a catalyst, indicating a base-growth mechanism. The fibers showed an average diameter of about 9 nm and a length of approximately 10 μm. This represents an improvement compared to other herringbone-structured CNFs, which typically have diameters of around 50 nm (Kim *et al*., 2024).

Keshavarz *et al*. (2022) found that non-spinning methods like CVD yield CNFs with superior mechanical strength and electrical conductivity. Unfortunately, it has high synthesis costs. In contrast, carbonization methods like electrospinning are more cost-effective and less toxic, but result in lower graphitization, weaker electrical and mechanical properties, and more structural defects. However, these defects can be advantageous for biomolecule attachment, making them beneficial for biomedical applications (Keshavarz *et al*., 2022)

Template-Based Synthesis

Template synthesis is a technique for producing a nanomaterial that utilizes a template material, adjusting the material's structure, morphology, and size. It is straightforward to execute and not highly sensitive to preparation conditions, but choosing the correct template is essential. The method influences particle formation by regulating crystal nucleation and growth to shape the particles (Tijjani *et al*., 2022). Wang *et al.* (2010) synthesized mesoporous carbon nanofibers using anodized aluminum oxide (AAO) templates. The CNFs was prepared using an evaporation-assisted self-assembly method. A solution of F127 and resol-ethanol was used to infiltrate the AAO templates, followed by heating at 100 °C for 24 hours to polymerize through thermal initiation using the phenolic resins. The carbonization was processed in nitrogen at 350 °C or 700 °C for 3 hours. After removing the AAO templates with HF, the results were classified as polymer nanofibers (PNF) at 350 °C or carbon nanofibers (CNFs) at 700 °C (Wang *et al*., 2010).

The chemical vapor deposition (CVD) method for synthesizing CNFs is well-regarded for its high-temperature process, enhancing graphitization and producing superior electrical conductivity. However, electrospinning, particularly when used to produce hollow nanofibers, can also yield high conductivity, resulting from the expanded surface area and the ordered arrangement of polymer chains as the fibers form. Compared to template-based synthesis, which typically involves complex steps such as template removal and limited control over large-scale uniformity, CVD and electrospinning offer more scalable and controllable approaches. However, template-based methods can precisely control nanostructure dimensions and morphology, which may be advantageous in tailoring specific electromagnetic properties. Table 1 compares the electrospinning, CVD, and template-based methods (Jeon *et al*., 2025).

Table 1. Comparison between different CNFs preparation methods.

Synthesis Method of CNFs	Comparison				
	High Carbon Content	**Structure and Morphology**	**Nanoscale Diameters (nm)**	**Pros**	**Cons**
Electrospinning	More than 90%	Aligned electrospun fibers	10 – 500	Simple and scalable process that functions without the use of complex catalysts, while allowing precise control over morphology.	Limited to certain morphology types and requires additional post-processing steps
CVD	More than 95% with purification	Tubular structure	10 – 100	Delivers excellent electrical conductivity (10 – 100 S/cm) through the use of graphitized fibers.	Involves high production costs and faces scalability challenges due to expensive equipment and operational demands.
Template-based synthesis	More than 90%	Uniform and porous fibers	5 – 200	Offers precise control over structure with adjustable pore size and surface area.	Template removal makes the process labor-intensive and time-consuming.

PREPARATION METHOD OF CARBON NANOFIBERS (CNFs)-METAL OXIDE AS HYBRID MATERIAL

Hybrid fillers consist of organic (typically carbon-based) and inorganic components. By tailoring the properties of these components, fillers can be optimized to enhance EMI protection by improving their absorption and reflection capabilities. Organic components contribute to filler weight reduction and improve their electrical properties, while inorganic components generally play a role in fine-tuning the magnetic properties for more effective protection (Zecchi et al., 2024). Hybridizing CNFs with metal oxides is a promising approach to

improving EMI protection. This combination complements each other's properties to achieve superior shielding performance.

A conducting polymer composite must demonstrate an EMI shielding effectiveness (SE) of at least –15 dB to be considered commercially viable for EMI shielding. The x-band frequency range (8.2 to 12.4 GHz) is especially crucial for EMI shielding applications of electronic devices (Choudhury et al., 2024). By integrating various types of fillers, hybrid fillers can capitalize on their respective advantages while overcoming their limitations. They can be engineered to possess tailored magnetic or electrical properties based on the specific application. (Zecchi et al., 2024). Various polymers and copolymers, have all been proposed as potential components for high-performance EMI shielding materials, such as polyacrylonitrile (PAN), polyaniline (PANI), polyvinylidene fluoride (PVDF), polydimethylsiloxane (PDMS), polyurethane (PU), polyimide (PI), polyamide (PA), polycarbonate (PC), and polypropylene (PP), ethylene vinyl acetate copolymer (EVA), carboxy-terminated butadiene acrylonitrile (CTBN), poly(ethylene glycol) diacrylate (PEGDA), poly(ethylene-alt-maleic anhydride) (PEMA), poly(L-lactide) (PLLA), polyolefin elastomer (POE), polylactic acid (PLA), , polyetherimide (PEI), polyvinyl alcohol (PVA), polyvinyl chloride (PVC), polybenzoxazine (PBO), polydopamine (PDA), polyethylene terephthalate (PET), ethylene-co-methyl acrylate (EMA), polytriazole (PTA), polypyrrole (PPy), ethylene octene copolymer (EOC), polyethylene (PE) (Kumar et al., 2021).

The development of materials for EMI shielding applications requires a thorough understanding of the various potential materials that can meet the required properties or complement each other. In this review, we focus on using carbon nanofibers (CNFs) and various metal oxides as hybrid fillers in polymers for EMI shielding. We review recent studies exploring methods for producing CNFs-metal oxide combinations and their application as hybrid filler-polymer composites.

A key strategy for improving the properties and performance of composite materials is the uniform incorporation of fillers into the matrix. Incorporating hybrid fillers offers a promising approach to improving material properties through synergistic interactions. Hybrid fillers provide advantages in improved interfacial properties and high strength in multi-filler polymer composites, thereby enhancing their electric field resistance. The dual filler system affects the electric field resistance by shielding weak dielectric interfaces. The combination of fillers creates a synergistic effect, maximizing the advantages of each filler within a single composite. These hybrid structures are designed to optimize filler interfaces in polymer composites and nanocomposites (Afolabi and Ndou, 2024). The following are some previous reports on incorporating CNFs and metal oxides using various synthesis methods.

Mamun *et al*. (2023) worked to combine Fe_3O_4 and PAN-electrospun CNFs for oxygen reduction reaction catalysts. Various Fe_3O_4 and PAN were prepared with concentrations: 25 wt.% Fe_3O_4 and 12 wt.% PAN (2.1:1 ratio), 30 wt.% Fe_3O_4 and 12 wt.% PAN (2.5:1 ratio), and 40 wt.% Fe_3O_4 and 10 wt.% PAN (4:1 ratio). Nanofiber mats were produced via needleless electrospinning onto a nonwoven polypropylene substrate. Key electrospinning parameters included an 80 kV voltage, 0.9 mm nozzle diameter, 150 mm/s carriage speed, and specific distances of 240 mm (mass-to-substrate) and 50 mm (electrode-to-substrate). The process occurred at 22 °C with 32% humidity for 20 minutes. The mats underwent oxidative stabilization at 280 °C. Carbonization was performed at 800 °C with a heating rate of 10 K/min and a nitrogen flow rate of 100 mL/min. The study revealed that the nanofiber mat with 25 wt% Fe_3O_4 had an elevated yield of 55.4%, with stabilization yields around 89.5% and carbonization yields around 61.9%, respectively. Laser scanning microscopy (CLSM) images exhibited that the 30 wt.% Fe_3O_4 nanofibers produced beads, while the 25 wt.% and 40 wt.% Fe_3O_4 samples had larger membrane areas. SEM images demonstrated that the 30 wt.% Fe_3O_4 nanofibers had more visible fibers than the membrane areas, with thinner fibers observed in the 25 wt.% sample (average diameter of 118 nm) compared to thicker fibers in the 30 wt.% sample (297 nm). Energy Dispersive X-Ray Spectroscopy (EDS) mapping also showed the emergence of iron on the nanofiber surface. The effective incorporation of active centers into electrospun membranes, while maintaining high and selective ORR catalytic activity, enhances their potential for use in electrochemical devices (Mamun *et al*., 2023).

Lee *et al*. (2021) developed a method to fabricate metal oxides such as NiO, ZrO_3, TiO_2, and SnO_2 on carbon nanofibers (CNFs) using electrospinning and sol-gel techniques. PAN and PVP were dissolved in DMF to create electrospun fibers, with PVP providing binding sites for inorganic materials and PAN serving as a carbon precursor. The step was initially done by depositing metal oxide nanoparticles on the fibers and immersing them in alkoxide solutions, heated to 280 °C, stabilized for 1 hour, and then carbonized at 800 °C for 1 hour under a nitrogen/hydrogen mixture. The electrospun fibers were collected, dried, and heat-treated, resulting in CNFs

modified with metal oxides. Various alkoxides were also used as precursors for metal oxides with CNFs. The resulting diameter of CNFs expanded from 80 nm up to 200 – 450 nm. XRD patterns confirmed the crystalline TiO_2 (rutile phase), while Raman spectra showed graphite (1600 cm^{-1}) and disorder bands (1370 cm^{-1}) and detected TiO_2 modes at 436 and 612 cm^{-1}. After heat treatment, TiO_2 and other metal oxides (NiO, ZrO_3, and SnO_2) were successfully formed on the CNFs' surface. The hybrid material CNFs@TiO_2 was applied as an electrode in lithium-ion batteries to demonstrate mechanical and electrochemical properties. The result shows an improvement in the reversible 600 mAh/g capacity after 100 cycles (Lee *et al*., 2021).

Electrospinning has become the most popular method for combining carbon nanofibers with metal oxides due to its one-step efficiency, where metal oxide precursors are simply blended with PAN/ PVP solutions, as seen in materials like CNFs-Zinc Oxide (ZnO) (Nekounam *et al*., 2023), CNFs-Cobalt Oxide (Co_3O_4) (Kim G. and Kim B., 2022), and CNFs-Barium Titanate ($BaTiO_3$) (Sahoo and Panda, 2012). Although the combination of carbon nanofibers and metal oxides is mostly carried out using the electrospinning method, many other approaches can still be used to modify CNFs and metal oxides. Das *et al*. (2015) explored a mild-temperature, nonchemical approach for encapsulating magnetite (Fe_3O_4) nanoparticles within hollow-core CNFs through ultrasonication-assisted capillary imbibition and diffusion. This technique results in densely packed nanoparticles inside CNFs cavities via solvent evaporation. By fine-tuning the process conditions, they successfully created magnetite-filled CNFs, which combines the conductive properties of the fibers with the magnetic characteristics of the nanoparticles. After sonication, the suspension was dried at 120°C to concentrate the nanoparticles inside the CNFs cavities and excess nanoparticles on the outside were removed by washing again using sonication (Das *et al*., 2015). Ajmal *et al*. (2023) described the technique of coating carbon nanotubes (CNTs) with Fe_3O_4 using microwave-assisted irradiation. This process begins with functionalizing the CNTs, which are then dissolved in a 0.5 M NaOH solution and xylene, mixed for 20 minutes of sonication. At the same time, a 0.13 M Fe^{2+} solution and a 0.25 M Fe^{3+} solution (with a ratio of 2:1) are sonicated for 20 minutes. The ion solution is gradually added to the CNT slurry in a microwave reactor, where 1000 W microwave radiation is applied for 2 hours at 180°C. The slurry was repeatedly washed after centrifugation and dried overnight at 60 °C (Ajmal *et al*., 2023).

Ni'maturrohmah et al. (2018) also synthesized copper modified with graphene oxide using an electrochemical method. Graphene oxide and NaCl were dissolved in deionized water. Electrolysis was performed in an electrochemical cell at 3V and 5V for 30 minutes. It functions as a copper plate as the anode and a carbon rod as the cathode. The result showed yellow-black precipitation that dried at 100 °C for 1 hour to obtain the final material (Ni'maturrohmah *et al*., 2018). Saraswati *et al*. (2017) reported using the submerged arc discharge method of carbon-modified iron oxide synthesis. Two electrodes were used: a carbon anode decorated with a mixture of graphite, iron oxide (synthesized using the electrolysis method), and silica binder in a 3:1:1 mass ratio, and a sharp-tipped graphite cathode. The mixture was sonicated for 480 seconds, molded into a rod, and heated at 180 ºC for 6 hours. The electrodes were placed in a 600-mL beaker with 300 mL of 50% ethanol, and a current of 10 A (13.75 V) was applied. The nanoparticles were then collected after arcing, and the ethanol was evaporated (Saraswati *et al*., 2017).

THE UTILIZATION OF CARBON NANOFIBER-METAL OXIDE AS HYBRID FILLER FOR EMI SHIELDING POLYMER COMPOSITES

The utilization of CNFs and metal oxides as hybrid fillers in EMI shielding composites has attracted attention due to their outstanding functionalization properties. CNFs can potentially offer high mechanical strength, thermal stability, and electrical conductivity, while metal oxides such as ZnO, TiO_2, and Fe_3O_4 provide high magnetic permeability and dielectric properties that can potentially enhance electromagnetic wave absorption (Anjaneyalu *et al*., 2019; X.-Y. Wang *et al*., 2022; Guadagno *et al*., 2023). Combining these materials creates a synergistic effect, enhancing the composite's performance in absorbing and reflecting electromagnetic radiation waves (Cao *et al*., 2024). In EMI shielding applications, hybrid fillers can effectively disrupt the transmission of electromagnetic radiation by dissipating energy as heat or reflecting it away from the core component (Isari *et al*., 2024).

The conductive CNFs network provides a pathway for electric current, while the metal oxide increases the composite's magnetic permeability and dielectric constant, enhancing its shielding effectiveness (Lyu *et al*., 2024). While electromagnetic wave absorption is the primary shielding mechanism in CNFs-metal oxide composites, surface reflection also plays a crucial role. When incident electromagnetic waves strike the CNFs surface, some

energy is reflected, leading to initial attenuation. The remaining waves penetrate deeper into the material, undergoing multiple internal reflections, generating heat that is absorbed and dissipated by the CNFs-metal oxide hybrid structure. As the waves travel through the fibrous matrix, their intensity is further attenuated, resulting in an overall increase in shielding efficiency (Li *et al*., 2024). An illustration of the EMI shielding mechanism of CNFs-metal oxide is shown in Figure 4.

As previously described, electrospinning has been widely used to fabricate CNFs–metal oxide hybrids due to its efficiency in producing uniform nanofibers with controlled diameters. By optimizing the ratio of CNFs to metal oxide, as well as the processing conditions, these hybrid fillers can be engineered to achieve high-performance EMI shielding composites. This composite has potential applications in various fields, including electronics, aerospace, and automotive industries, where EMI shielding is critical (Mishra *et al*., 2018).

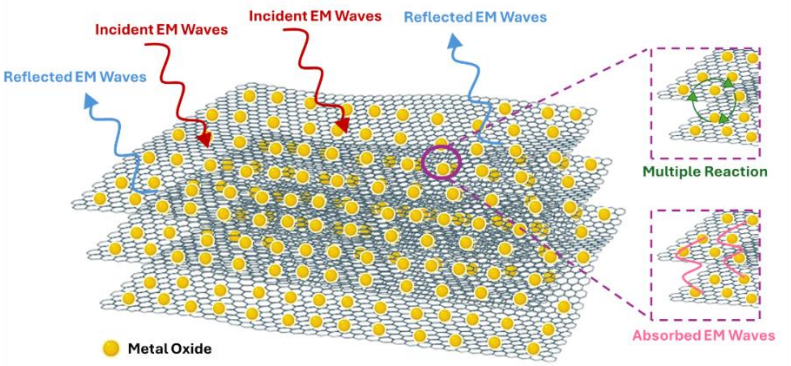

Figure 4. The illustration of the EMI shielding mechanism in a CNFs-metal oxide hybrid filler.

Saraswati *et al*. (2025) explore the study about the development of epoxy resin combined with polyimide (ER–PI) as hybrid composites reinforced with aminated CNFs. The CNFs were modified through an amination process involving ethylenediamine, sulfuric acid, and sodium nitrite. These aminated CNFs were then incorporated into the polymer matrix, resulting in composites with enhanced mechanical properties and electromagnetic interference (EMI) shielding effectiveness. As illustrated in Figure 5, the CNFs fillers are dispersed within the epoxy resin–polyimide matrix, but their distribution also results in the formation of voids. These voids allow electromagnetic waves to escape more easily, reducing the material's ability to trap and attenuate them effectively.

In contrast, the uniform dispersion of aminated CNFs within the polymer matrix minimizes the formation of such voids. It possibly enhanced electromagnetic wave attenuation through multiple reflections and absorption, improving absorption-based shielding performance. This research highlights that the aminated CNFs improved dispersion within the matrix and established stronger covalent bonds, leading to significant improvements in hardness, tensile strength, and EMI shielding efficiency compared to composites with unmodified CNFs. The specimen composite and the tensile load result are given in Figure 6 (Saraswati *et al*., 2025).

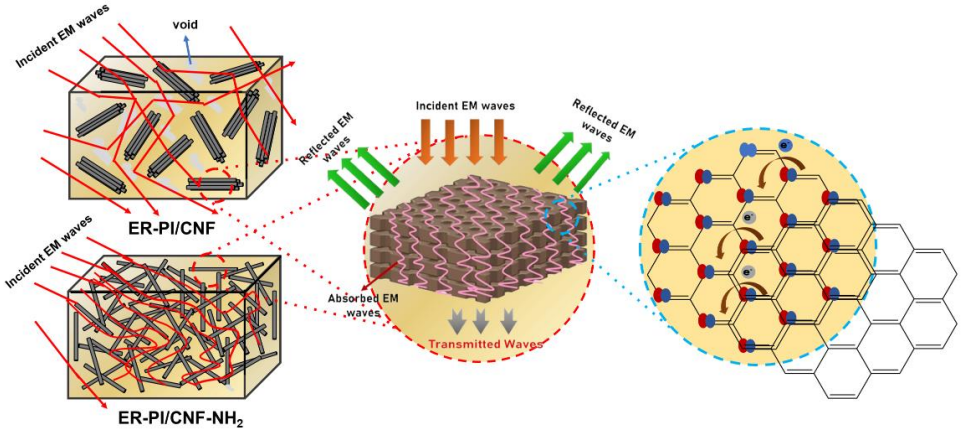

Figure 5. The illustration of CNFs and aminated CNFs filler interaction in epoxy resin-polyimide composite during absorbing the electromagnetic wave (Saraswati *et al*., 2025).

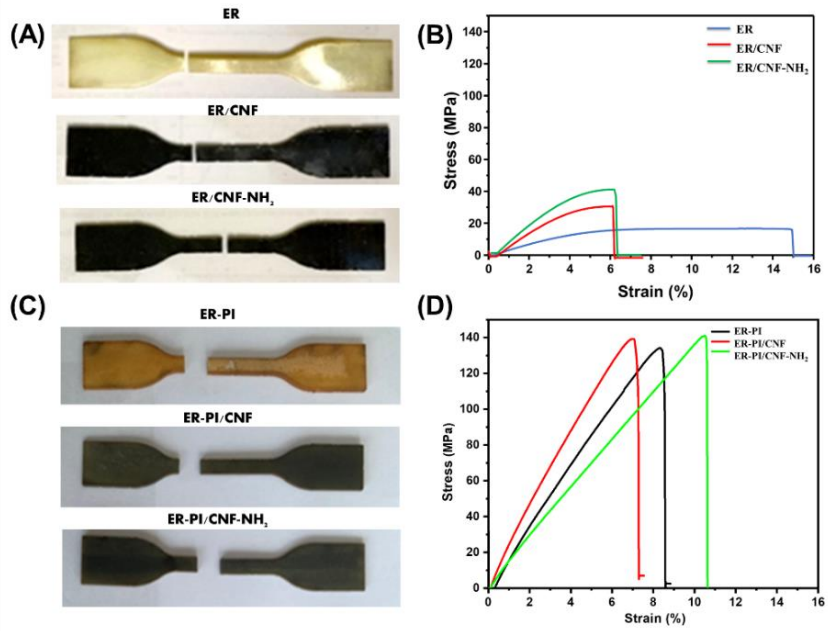

Figure 6. The specimen composite of epoxy resin-polyimide reinforced by CNFs and aminated CNFs hybrid filler (a, c), and the tensile load vs displacement curves of composites (b, d) (Saraswati *et al*., 2025).

Wu *et al*. (2015) described a functionalization process using the co-precipitation method of carbon nanofibers coated with magnetic iron oxide nanoparticles. The prepared Fe_3O_4-CNFs was sonicated in acetone for 15 minutes (with a concentration of 20 mg/mL). Epoxy resin was mixed and sonicated for 1 hour. The amount of hardener was also added. The mixture was then cast into a rubber mold for curing. The hybrid Fe_3O_4@CNFs was treated with permanent magnets (with a magnetic field of 50 mT) to achieve an aligned material during the curing process.

Furthermore, the hybrid Fe_3O_4@CNFs improves the mechanical toughening efficiency and fracture energy of the epoxy nanocomposites up to 150% compared to the unmodified epoxy polymer. It is due to the nanofiber hybrid's interfacial debonding, crack bridging, pull-out, and rupture of the Fe_3O_4-CNFs void growth in the epoxy polymer. The hybrid Fe_3O_4-CNFs aligned to the crack surface impacts the increasing interaction with the higher crack tip, resulting in more effective toughening (Wu *et al*., 2015).

Deeraj *et al*. (2021) studied zirconia-embedded carbon nanofibers (CNFs) prepared via electrospinning. The EMI shielding performance of CNFs was characterized using Ku band, a critical frequency range for satellite and aircraft communication. The study investigated the influence of layer number (thickness) on total shielding effectiveness to clarify the role of absorbance in EMI shielding. Zirconia-embedded carbon nanofiber/epoxy (ZrO_2/CNFs/epoxy) laminates were prepared using a simple hand layup method to demonstrate practical applicability further. Pure epoxy, being non-conductive and insulating, exhibits a shielding effectiveness (SE) below 5 dB, making it unsuitable for EMI shielding on its own. In contrast, ZrO_2/CNFs/epoxy laminates achieved total EMI SE values between 16.7 and 18.8 dB. This improvement underscores the synergistic contribution of CNFs and epoxy in enhancing EMI shielding performance. Notably, zirconia-loaded samples reached an EMI SE of -94 dB at a thickness of 0.72 mm, corresponding to a specific shielding effectiveness of about -130 dB/mm, which is remarkably high. When compared with other materials such as flexible graphite (129 dB/mm), graphene foam (84 dB/mm), graphene aerogel (12 dB/mm), and CNFs mats (63 dB/mm), the ZrO_2/CNFs/epoxy composites developed in this work show comparable or even superior performance (Deeraj *et al*., 2021).

Several studies have explored using hybrid fillers or materials by combining electrospun carbon nanofibers (CNFs) with metal oxides. Table 2 compares the performance of several CNFs-metal oxide in polymer matrices and their EMI shielding. Based on these studies, it can be concluded that incorporating magnetic materials with CNFs significantly enhances EMI shielding effectiveness compared to using CNFs alone. This improvement is attributed to the synergistic interaction between the CNFs and metal oxides, which complement each other's properties.

Table 2. Comparison of the EMI shielding performance of various CNFs-metal oxide composites.

CNFs-Metal Oxide Hybrid Filler Material	Polymer Matrices	Conductivity	Magnetic Properties	EMI Shielding Effectiveness (SE^T)	Ref.
CNFs-Fe_3O_4	Polydimethylsiloxane (PMDS)	7.10 S/cm	Ms = 7.5 emu/g; Mr 2.62 emu/g; Hc = 240 Oe	55.47 dB with 0.7 mm thickness	(Bayat et al., 2014)
CNFs-ZrO_2	Epoxy	0.011 $(\Omega. cm)^{-1}$	-	94 dB with less than 0.8 mm thickness	(Deeraj et al., 2021)
CNFs-$BaTiO_3$	Polydimethylsiloxane (PMDS)	0.32 S/cm	-	81 dB with 0.24 mm thickness	(Sharma and James, 2024)
CNFs-$CoFe_2O_4$-Cu	-	-	Ms = 24 emu/g; Mr = 0.63 emu/g; Hc = 156.5 Oe	15.30 dB with 100 nm thickness	(Yousefi et al., 2025)
CNFs-$W_{18}O_{49}$-Ag	-	-	-	100.9 dB with 110 µm thickness	(H. Wang et al., 2022)

CONCLUSION

The hybridization of carbon nanofiber and metal oxide will make a potential material as the hybrid filler for EMI shielding composites. CNFs offers high electrical conductivity, mechanical strength, and thermal stability, while metal oxides contribute high magnetic permeability and dielectric properties that possibly increase the absorption of electromagnetic waves. The synergistic effect will enhance the composite's ability to absorb and reflect electromagnetic waves as the EMI shielding composite material.

CONFLICT OF INTEREST

There is no conflict of interest in this article.

AUTHOR CONTRIBUTION

DDS: Conceptualization, Manuscript Review Drafting and Editing, Project Administration; TES: Conceptualization, Manuscript Review Drafting and Editing, Project Administration, Supervision, Validation; WWR: Manuscript Review Drafting and Editing, Validation, Supervision.

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