http://pubs.acs.org/journal/acsodf

Review

Graphene-Based Electrospun Fibrous Materials with Enhanced EMI Shielding: Recent Developments and Future Perspectives

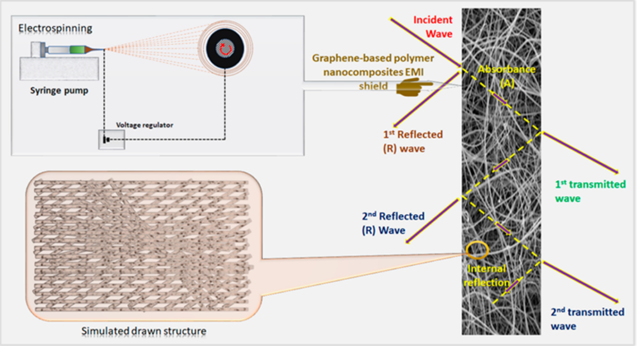
Jonathan Tersur Orasugh and Suprakas Sinha Ray*

Cite This: *ACS Omega* 2022, 7, 33699−33718

Read Online

ACCESS | Metrics & More | Article Recommendations

ABSTRACT: As a result of advancements in electronics/telecommunications, electromagnetic interference (EMI) pollution has gotten worse. Hence, fabrication/investigation of EMI shields having outstanding EMI shielding performance is necessary. Electrospinning (ES) has recently been established in several niches where 1D nanofibers (NFs) fabricated by ES can provide the shielding of EM waves, owing to their exceptional benefits. This review presents the basic correlations of ES technology and EMI shielding. Diverse graphene (GP)-based fibrous materials directly spun via ES as EMI shields are discussed. Electrospun EMI shields as composites through diverse post-treatments are reviewed, and then different factors influencing their EMI shielding characteristics are critically summarized. Finally, deductions and forthcoming outlooks are given. This review provides up to date knowledge on the advancement of the application of graphene-based electrospun fibers/composite materials as EMI shields and the outlook for high-performance electrospun fibers/composite-based EMI shielding materials.



1. INTRODUCTION

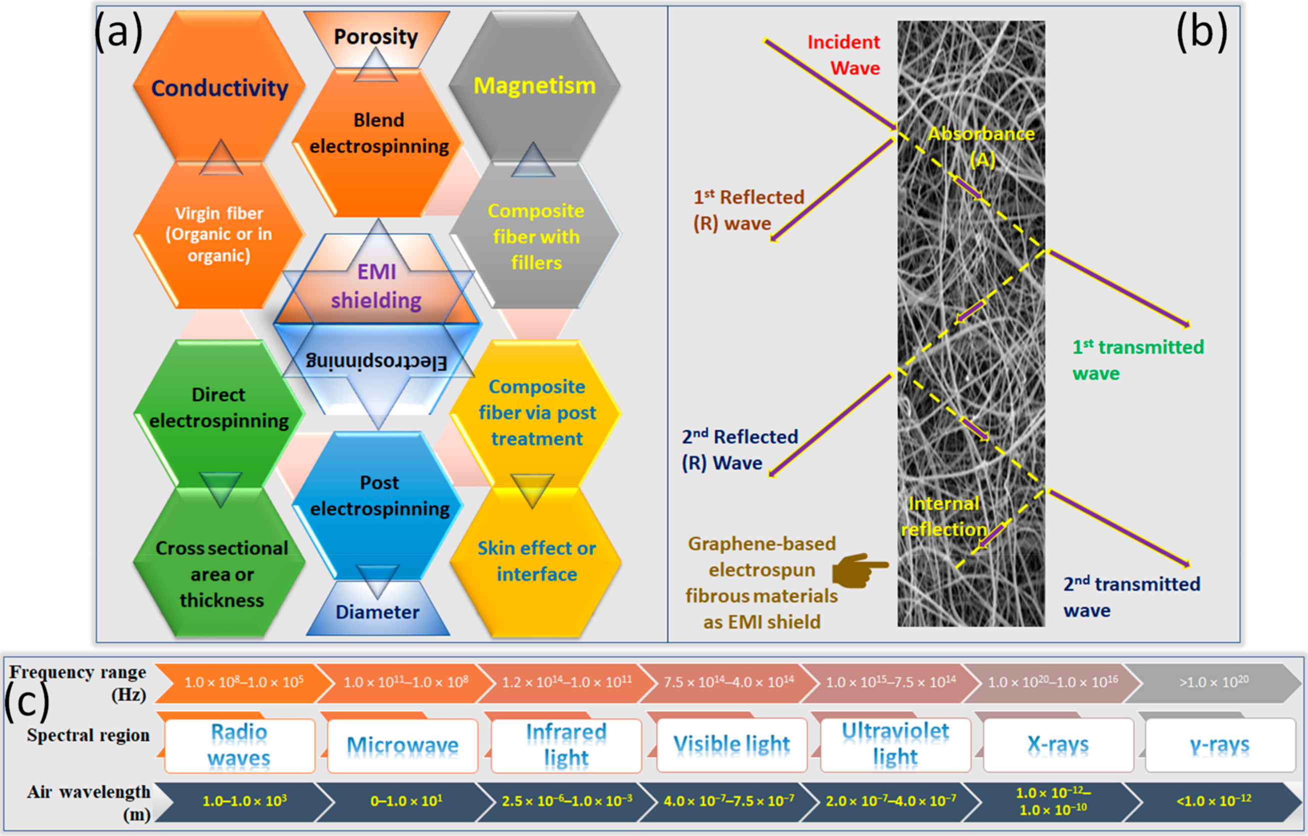
With consideration to electromagnetic waves (EMWs), it is well-known that a change in the electric field (EF) will directly result in a change in the magnetic field because both fields oscillate in a similar direction and are abrupt to one another too, hence, developing EM waves within the fluctuating field. EMWs may propagate in fluid, solid, gas, and vacuum without requiring a transfer medium, which implies they can move wave energy on a plane made of magnetic fields and/or EFs. In the meantime, the EM radiation created by EM waves incredibly affects the outer layers and cannot get back to the propagation beginning once engendered: this impact is referred to as electromagnetic interference (EMI).[1]

Due to fast-progressing information technology innovation, electronic gadgets are assuming significant parts of our everyday life. However, electronic gadgets can likewise progress to issues, such as EMI and many others.[2] The presence of EMI not only causes challenges in customary battle weapons or electronics but also makes these materials experience unusable performance; in addition, it purposes unacceptable results on accuracy of instruments, for example, the unusual activity and estimation blunders of the instruments or divulgence of private archives, subsequently influencing public safety. Additionally, EMW radiation likewise causes harm to humans. For example, the focal sensory system of the human body can be irreparably harmed, due to long haul exposure in an unnecessary electromagnetic radiation climate.[3−5]

The rise of EMI shields tackles a progression of previously mentioned issues brought about by EM waves: these shielding fabrics could conquer the electric/magnetic domains within the locale and viably self-control the spread of EM waves starting with any district and then onto the next. Customary EM wave shields are for the most part fabricated from metal-based components that are generally folded over the gadget to shield it from EMI.[3] Notwithstanding, high-thickness metal-based materials are hard to handle and are easily oxidized and eroded noticeably, which genuinely hinders their utilization in EMI shielding.[4] Moreover, with the improvement of present-day electronics, such as cell phones, PCs, and other small devices under unforgiving conditions, EMI shielding materials progressed with adaptability and low thickness, and the brilliant EMI shielding performance has drawn much consideration lately.[3]

Received: June 8, 2022
Accepted: August 19, 2022
Published: September 12, 2022

https://doi.org/10.1021/acsomega.2c03579
ACS Omega 2022, 7, 33699−33718



Frequency range (Hz)	1.0×10^8–1.0×10^5	1.0×10^{11}–1.0×10^8	1.2×10^{14}–1.0×10^{11}	7.5×10^{14}–4.0×10^{14}	1.0×10^{15}–7.5×10^{14}	1.0×10^{20}–1.0×10^{16}	$>1.0 \times 10^{20}$
Spectral region	Radio waves	Microwave	Infrared light	Visible light	Ultraviolet light	X-rays	γ-rays
Air wavelength (m)	1.0–1.0×10^3	0–1.0×10^1	2.5×10^{-6}–1.0×10^{-3}	4.0×10^{-7}–7.5×10^{-7}	2.0×10^{-7}–4.0×10^{-7}	1.0×10^{-12}–1.0×10^{-10}	$<1.0 \times 10^{-12}$

Figure 1. (a) Overview of graphene-based electrospun fibrous materials as EMI shields, with different corresponding influencing factors. (b) Schematic presentation of EM wave sheltering mechanism of graphene-based polymeric nanocomposites/fibers as well as (c) electromagnetic spectrum wavelength and frequency.

One-dimensional (1D) nanofibers (NFs) have become famous materials lately because of their customizable subatomic designs, low thickness, high porosity, and good mechanical properties.[5] In the meantime, 1D NFs have demonstrated incredible possibilities for planning lightweight, adaptable, and exceptional EMI shielding characteristics.[6,7] NFs are continually being created, and their design advancements are additionally improved bit by bit, for example, for stage partition,[7] stretching technique,[8] self-gathering strategy,[9] electrospinning innovation,[10] synthetic and mechanical division,[10] and in situ polymerization.[11]

Electrospinning is a compelling and generally utilized methodology for setting 1D NFs, inferable from its straightforward, adaptable, climate-amicable, huge scope creation, short creation cycle, and simple to control technique.[3−14] Electrospun NFs, per the literature, are generally employed within the drug delivery niche,[14] organic tissue designing,[15] energy storage,[16] ecological catalysis, as well as adsorption,[17] wireless telecommunications,[18] armed forces,[19] and aviation.[20] Specifically, an enormous number of experiments on the use of electrospun filaments for EMI shielding have been done recently because of their adaptability, light weight, low thickness, and usefulness. Notwithstanding 1D NFs, aerogels and films are likewise normal EMI protecting materials: contrasted with NFs, aerogels have higher porosity and lower thickness; however, their planning interaction is substantially more confounded and energy-consuming.[21] Furthermore, films have basic manufacture measure; however, their little explicit surface region very much restricts their viable utilization in EM wave sheltering.[21]

Although several reviews papers credited to electrospinning and its pertinent utilization are accounted for,[12,22] no important report has been given on the graphene-reinforced polymer electrospun nanocomposite/fiber substrates and their use for EMI shielding. In this regard, we have herewith given a concise discussion on the electrospinning of graphene-based polymer nanocomposites/fibers aimed at EM wave sheltering, zeroing in on various types of electrospun filaments and associated post-treatment of spun fibers for EM wave sheltering, as well as distinctive comparison of impacting factors, as depicted in Figure 1a.

1.1. Fundamentals of Electrospinning (ES).

The electrospinning hypothesis, projected by William Gilbert around 1600, was framed from the development of cone-shaped H_2O in the presence of applied EF.[23] Over the course of the following 300 years, however, the connected compositions and licenses were continually distributed, and electrospinning innovation was not even additionally created because of the absence of sufficient characterization instruments. Until the early 1990s, this innovation was attributed to the creation of electron-magnifying tools to describe materials with nanoscale sizes by Darrell Reneker, Gregory Rutledge, and Wendorff.[24] ES innovation can handle the fiber breadth from the nanometer phase/scale to the micron phase, which is one of the principal approaches to manufacture 1D NFs.

The electrospinning gadget principally incorporates three sections: the spinneret, high-voltage supply of power, and fiber collection gadget. Figure 2 shows a schematic presentation of the electrospinning technique. The polymeric mixture for ES within the needle is moved using the needle siphon, even as a drop is framed at the nozzle/spinner, aided by high surface tension coupled with gravity (in some setups). At the point where the drop is put in the climate of a high electrostatic arena, a gradual increase of the electrostatic repulsion results in extension of the drop, forming a cone shape under the activity of steady force. When the electrostatic voltage at the spinning nozzle approaches a critical point, the repulsion due to electrostatic force balances with the droplets' surface tension, resulting in a Taylor cone.[25] With the steady rise in the static voltage, a fiber is shot out at the edge of the formed Taylor cone and deposited on the collected gadget. In the ES technique, the viscosity/concentration of polymer solution, loading voltage, feed rate, distance for fiber collection, chamber temperature, and/or humidity directly affect the fiber morphologies.[12,26] Until now, many scientific investigations have been committed to investigating the electrospun strands with further developed EM wave sheltering fabrics due to high perviousness, nanoscale diameter, excellent tensile/dynamic characteristics, physical-tailed properties, and abundant skin effect/interfacial contacts aimed at dispelling EM oomph.[26,27]

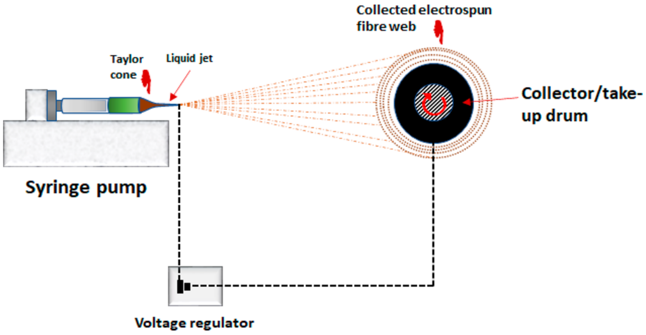


Figure 2. Schematic presentation of the electrospinning technique.

1.2. Introduction to EMI Shielding. EM waves can increase within the void or a given medium. They are categorized as radio waves, microwaves, infrared light, visible light, ultraviolet light X-rays, and γ-rays per their wavelength and/or frequency (Figure 1c). The EMW with varying wavelength and/or frequency can cause diverse influences on peripheral bodies. Whenever EMW strikes the surface of an object, some portion of the EMWs is reflected (Figure 1b), while the residual waves pass through the incident object(s). This might as well be transmitted/reflected at the shield deuce interfaces within, resulting in uninterrupted attenuation.[12]

The SE is generally adopted to quantitatively portray the sheltering impact of EM waves in decibels (dB), as EMI shields having 20 dB (~99% weakening of EM waves) meet business necessities.[1] By estimating the EF strength E_0 without a shielding material and E_S at a similar spot with a shielding material, SE could be determined using eq 1. SE could likewise be determined utilizing eq 2 by estimating the attractive field strength H_0 and H_s without shielding material and a shielding material, individually. Essentially, SE can be determined by eq 3 by estimating the power density P_0 (without an EMI shielding material) and P_s (with an EMI shielding material).[12,28]

$$\mathrm{SE} = 20\log\frac{|E_0|}{|E_s|} \tag{1}$$

$$\mathrm{SE} = 20\log\frac{|H_0|}{|H_s|} \tag{2}$$

$$\mathrm{SE} = 20\log\frac{|P_0|}{|P_s|} \tag{3}$$

$$\mathrm{SE} = \mathrm{SE}_\mathrm{A} + \mathrm{SE}_\mathrm{R} + \mathrm{SE}_\mathrm{M} \tag{4}$$

$$\mathrm{SE}_\mathrm{A} = 8.7t\sqrt{\pi f\mu\sigma} \tag{5}$$

$$\mathrm{SE}_\mathrm{R} = -10\log\left(\frac{\sigma}{16f\varepsilon\mu_\mathrm{r}}\right) \tag{6}$$

Also, the SE of the eventual EM wave shields is determined by utilizing the transmission line theory, and the SE is obtained by combining the reflection loss (SE_R), absorption loss (SE_A), and multiple internal reflections (SE_M) (eq 4). SE_A and SE_R are shown in eqs 5 and 6, individually.[12]

From the equations as described above, f, t, r, μ_r, and σ are the electromagnetic frequency, thickness, distance between the source field and the considered EMI shield(s), the shields' relative permeability, and its relative conductivity, respectively.[12] From the above expressions, it is inferred that the EMI SE is affected by the conductivity, magnetism, thickness, and permeability of the shielding material. The addition of a magnetic/conductive phase to the substratum fabrics can make it retain magnetic as well as electric dipoles which weaken the ohmic resistance (impedance) of shields, resulting in enhanced mismatch to achieve a reflection of the EMWs.[29] In the interim, magnetic dipoles within the shielding fabric may effectively create eddy current loss, natural loss, interfacial polarization, and magnetic loss, consequently resulting in enhanced EMI shielding. Furthermore, SE_M could be insignificant when $\mathrm{SE}_\mathrm{A} \geq 15$ dB.[12,29] The distance needed for the EMW constriction to $1/e$ or 37% inside the EMI shields is referred to as skin depth (δ), which is determined utilizing eq 7.

$$\delta = \frac{1}{\sqrt{\pi f\mu\sigma}} \tag{7}$$

The EMW disseminated by reflection on a superficial level and within the materials is essentially dispersed as heat energy. Additionally, EM waves' reflection within the shield might bring about a decrease in sent signs, which is not identified with the absorption of EM waves within the shield. In shields that are monolithic and isotropic in nature, the absorption (A) is derivatively presented from the reflectance (R) as well as estimated transmittance (T):

$$A = I - R - T \tag{8}$$

The EM wave shielding component incorporates its reflection loss with respect to the shield's surface, the SE_A and the numerous SE_R within the shielding fabric (Figure 1b). With regard to electrospun strands, the specific 1D structural design can build a thick conductive configuration, which can prompt improved reflection of EMWs. Moreover, low impedance could remain because of the high porosity and enormous aspect ratio of electrospun NFs. Accordingly, EM waves could go into the fibrous material, and loss of EM energy will occur within its interfaces among EM waves and the electrospun fibers/strands. This extraordinary impact of damping EM waves and displaying superb shielding impact presents electrospun filaments/fibers as a promising competitors among EMI shields. A graphical representation of electromagnetic wave as well as the A segment of traveling EM wave with in-phase mutually orthogonal E and B is depicted in Figure 3.

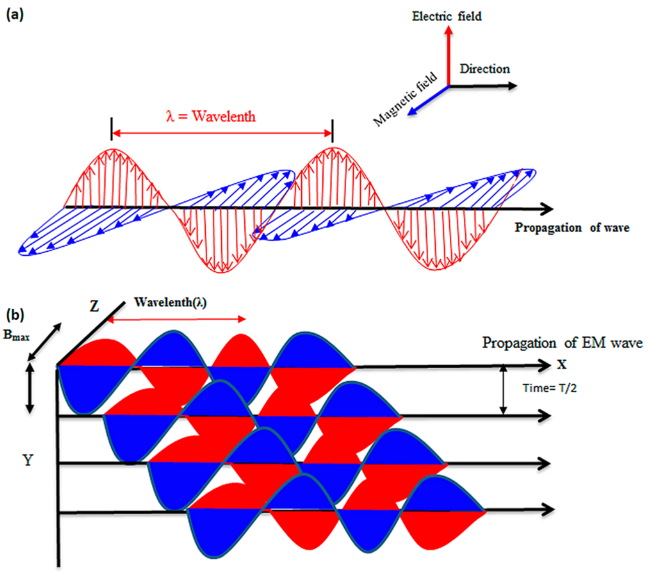


Figure 3. (a) Graphical presentation of electromagnetic wave and (b) A segment of traveling EM wave with in-phase mutually orthogonal E and B. Reproduced with permission from ref 30. Copyright 2022 Elsevier Science Ltd.

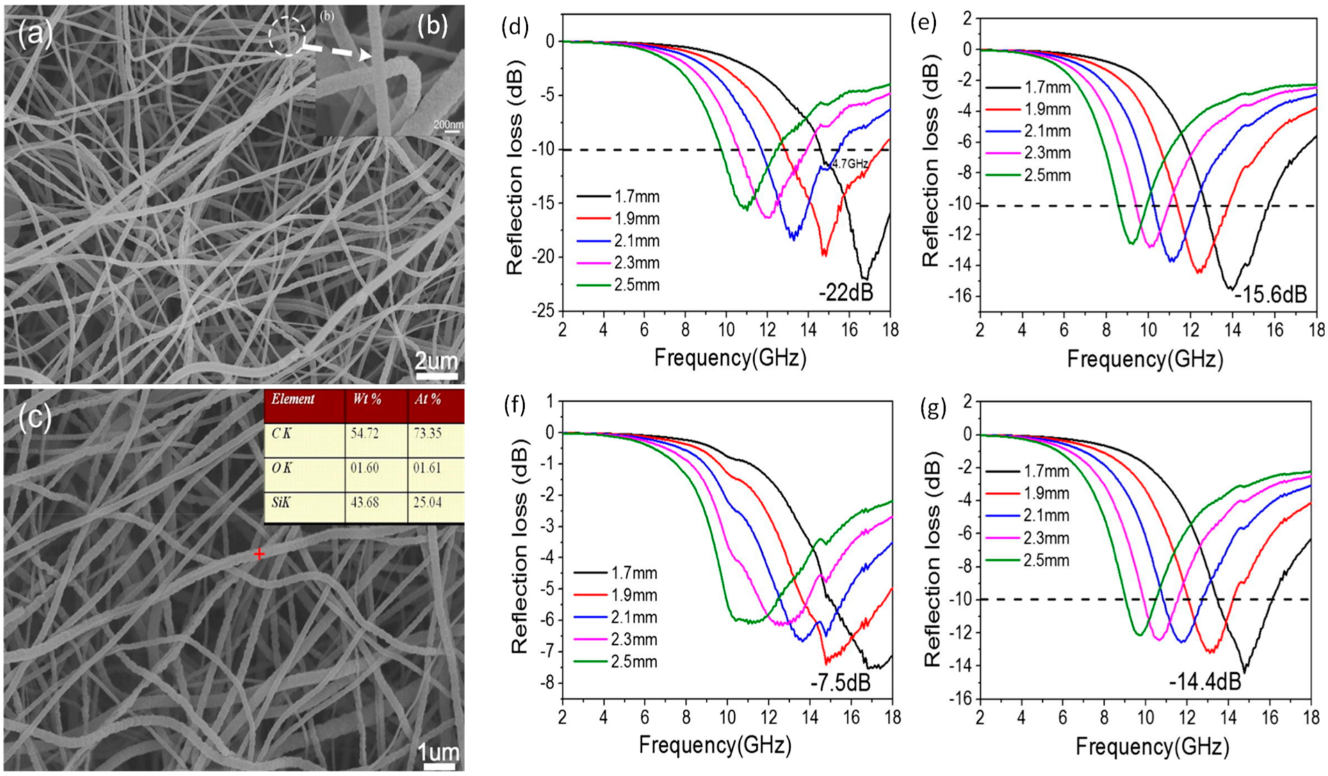
2. GRAPHENE

Graphene is a 2D single-atom-thick plate of sp^2-hybridized carbon atoms with carbon at a chain length of 0.142 nm.[31] It really is the "skinniest material", consisting of a single layer of carbon atoms assembled in an axial honeycomb crystal lattice. This same basic structural component of allotropes of carbon such as graphite, carbon nanotubes (CNTs), as well as fullerenes has always been graphene. Graphite has been around for a great many years. In a pioneering investigation, Novoselov et al.[32] managed to identify monolayers of graphene as well as other 2D crystals for the very first time. Graphene is a versatile material because of its excellent mechanical properties, along with a Young's modulus of 1 TPa,[33] a mean tensile strength of 13,010 GPa, outstanding thermal heat transfer (5000 W/m/K)[34] and charge transport characteristics (250,000 cm^2/V),[32] and good EMI shielding (>20 dB). Such extraordinary characteristics have also sparked tremendous interest in graphene for practical uses such as thermally as well as conductive reinforced nanomaterials, next-generation logic gadgets, sensing, associated electronics, transparent as well as dynamic electrodes for display systems as well as photovoltaic panels, capacitors, and lithium batteries.[35] Hence, graphene is indeed the new stuff for nanostructured materials as well as nanodevice manufacturering in the coming years. Assorted additives have also been attempted for producing nanocomposites with improved characteristics since the revelation of polymer nanocomposites.[36] Inorganic additives including organic montmorillonite-type layered silicate molecules or synthesized clay, nanoparticles, as well as carbon-based fillers such as carbon black, expanded graphite (EG), carbon nanotubes (CNF), and CNTs are examples. Seeing as inorganic fillers possess poor electrical and thermal conductivity, diverse carbon-based additives are becoming more appealing. CNTs, despite their high cost, have proven to be excellent conductive fillers. Graphene's tensile strength is comparable to or slightly higher than that of CNTs.[37] Furthermore, graphene has thermoelectric conductivities significantly higher than those of CNTs.[38] The amazing properties of graphene@polymer nanomaterials included the amalgamation of exceptional mechanical properties, outstanding thermal conductivity, as well as electrical conductivity. As a result, graphene has emerged as a superior padding for polymer nanocomposites. Massive interest in graphene has also resulted in extensive research, and as a result, a large number of articles have been published, as well as outstanding evaluations on graphene and modified graphene-based materials.[39,40]

Several processes for producing graphene sheets have been reported. Graphene is synthesized using thermal chemical vapor deposition (CVD) on metal surfaces and epitaxial growth from silicon carbide (SiC) without the use of complex mechanical or chemical treatments.[41] Plasma-enhanced CVD is another method for producing graphene sheets at a lower temperature than thermal CVD.[42] Physical exfoliation of graphite has generated graphene sheets;[32] this path is indeed recognized also as the "Scotch tape" or "peel-off" technique. From another method, graphene is created by chemically reducing graphene oxide (GO).[43] Among all of these methods, reduced in the presence of graphite to GO is regarded as one of the most cost-effective as well as a viable methods of producing large quantities of graphene sheets.[43] Multiple challenges must be resolved before the full potential of graphene- or graphene-oxide-based composites can be realized. The primary issue mostly in the fabrication of massive quantities of graphene has been that pristine graphene invariably tends to agglomerate to constitute stacked 3D graphitic edifices via $\pi-\pi$ interaction but as a result cannot be processed further owing to its high hydrophobic character. Furthermore, the use of pristine graphene in polymeric composites is difficult because phase-separated nanocomposite forms tend to result in mechanical malfunction at low load conditions than in a neat polymeric matrix.[44] That is also due to the incompatibility of graphene's surface with organic polymeric composites. Other major concerns, including (i) homogeneous distribution of nanosheets in a polymeric matrix and (ii) interfacial adhesion among graphene as well as the symbiont polymer, do have a direct impact on the nanocomposites' characteristics. Controlling the conformational changes, folding, as well as flexing of graphene nanostructures is also critical.

2.1. Advantages and Disadvantages of Graphene-Based Electrospun Polymer Composite.

2.1.1. Advantages of Graphene-Based Electrospun Polymer Composite. The electrochemical stability of intrinsic conducting polymers/graphene composites has been shown to be significantly greater than that of virgin intrinsic conducting polymers in advanced materials applications such as electrodes. The improved mechanical, thermal, and electrical properties of graphene-reinforced polymer-based nanocomposites vary depending on the nanocomposites.[45] Materials based on graphene or polymers have many benefits, including high specific active surface areas, excellent electron transport capabilities, and good capacitance.



Element	Wt %	At %
C K	54.72	73.35
O K	01.60	01.61
SiK	43.68	25.04

Figure 4. Morphology and element analysis (a−c) of the hybrid nanowires. Microwave RL for the samples of (d) S1300, (e) S1400, (f) S1500, and (g) S1600. Reproduced with permission from ref 52. Copyright 2018 Elsevier Science Ltd.

Because of the establishment of a "tortuous path" in the presence of graphene in the nanocomposites, nanocomposites with homogeneously dispersed graphene in the polymer matrix have also demonstrated good barrier characteristics, especially in packaging applications.

Graphene-based polymer nanocomposites have shown excellent performance in terms of EMI shielding. The EMI shielding efficiency of low weight percent graphene-filled polymeric composites can present an EMI shielding performance that is greater than that of the commercial target value of ≥20 dB.[45]

Graphene-based polymer nanocomposites have been proven to outperform neat polymers and conventional graphite-based composites in terms of mechanical characteristics. They also outperform their neat polymer counterparts in terms of thermal stability.

Polymer graphene-based nanocomposites increased electrical conductivity by several orders of magnitude. The development of a conducting network by graphene sheets in the polymer matrix results in a significant improvement in electrical conductivity.[45] Graphene-reinforced polymer composites also outperform their metal-based counterparts in terms of light weightness.

2.1.2. Disadvantages of Graphene-Based Electrospun Polymer Composite. Nanocomposite materials containing graphene/polymer may face great challenges in future applications, especially in scientific and medical fields, due to the cytotoxic effects of graphene−polymer composites.

A major technical problem is dispersing the nanofillers uniformly within polymer matrixes. In the dearth of covalent bonding as well as existing nonbinding interactions such as $\pi-\pi$ interactions and hydrogen bonding, poor interfacial adhesion throughout graphene/polymer nanocomposite emphasizes the significance of platelet surface modification in the reinforcement of the need for constant improvement in the path.

3. CATEGORIES OF GRAPHENE-BASED ES-FABRICATED FIBERS FOR EMI SHIELDS

Electrospinning is an approach to create various types of filaments/fibrous strands, such as virgin filaments with one segment and composite strands with diverse fillers. In this regard, neat polymeric electrospun fibers and their composites utilized for EMI shielding will be presented.

3.1. Graphene-Based Neat ES-Fabricated Fibers for EMI Shields.

Graphene-based polymer electrospun nanocomposite fibers for EMI shielding applications generally can be engineered to possess dielectric tunable behavior as well as electrical conductivity (EC). Such ES of neat fibers (graphene−ceramic fibers and/or graphene−carbon fibers) have been reportedly applied for EMI shielding per the literature.[3,12,24,28,46,47]

3.1.1. Graphene−Ceramic Electrospun NFs. Graphene−ceramic composite NFs such as graphene−SiC, graphene−SiO_2, graphene−Si_3N_4 NFs fabricated via the ES approach have benefits including high Young's modulus, low density, chemical stability, as well as tunable dielectric character, making them an excellent choice for EMI shields within the GHz range of frequency.[48]

Wide band gap semiconductors such as SiC materials can present the impedance match to attain high-efficiency attenuation. Also, the EM loss of the virgin silicon carbide NFs is largely derived from its state loss of available carbon and the polarization loss of nanocrystalline silicon carbide.[49] In the first report of its kind, electrospun SiC (SiC nanocrystals)/rGO core−shell nanowires have been utilized as highly efficient, lightweight EM wave absorbers/shields successfully fabricated via solution electrospinning followed by a postannealing process in a N_2 environment. The EM wave absorption performance of these nanowires was reportedly enhanced proficiently by tuning the included GO solution (1.0 mL) in the ES solutions, which displayed a SE_R of −56.3 dB (optimum), as well as its actual bandwidth absorption, which could be ≤6.2 GHz covering the entire Ku band (12.2−18.0 GHz). These authors established that the explicit core@shell assembly, defects as well as rGO and C surface functionalities, along with the skin-effect/the grain boundaries between the carbon cluster and the SiC nanocrystals, all play a role in the electrospun nanowire's EM wave absorption ability.[50]

Electrospun generated Ti_3C_2Tx/GO (MXene@GO) nanocomposite aerogel microparticles aimed at tunable high-performance EM wave absorption/shielding have been reported.[51] These authors reported that the fabrication of 2D nanomaterials into hybrids is an effective manner to prepare high-performance MA systems, having heterointerfaces that offer innovative loss phenomenon, thereby making up for the limitations of sole materials in EM energy attenuation,[51] with the consideration that, in practical deployments, thin and lightweight microwave-absorbing materials have excellent performance at low-frequency bands that are favorable, and they engineered hybrid aerogels fabricated with GO and Ti_3C_2Tx through quick freeze-assisted electrostatic spinning.[51] The differences in the conductivity GO and Ti_3C_2Tx abundant heterointerfaces and surface groups were generated, even as the Ti_3C_2Tx@GO hybrid exhibited optimized hybrid properties as well as MA.[51] They observed that the inimitable aerogel structure offered light weight along with stretching of the attenuating routes when EM waves were injected even at a relatively small reinforcing filler inclusion of 10.0 wt % at a material thickness of 1.2 mm, while optimized M@GAMS structures exhibited SE_R of −49.1 dB at 14.2 GHz.[51] More notably, the M@GAMS presents effective microwave absorption at the S-band, and the RL reaches −38.3 dB at 2.1 GHz at 5.0 mm thickness.[51] With the modification of such nanocomposite systems using additional active nanoparticles and/or conducting polymers as in blends, outstanding materials can be engineered using a similar path as postulated by the authors above.

A simple approach toward the tunable fabrication of graphite/silicon carbide hybrid nanostrands through ES techniques followed by annealing at elevated temperature has been demonstrated in another study.[52] The hybrid nanowires (Figure 4a−c) synthesized exhibited comparatively wide-ranging EM shielding behavior with lower SE_R of −22 dB (Figure 4d), −15.6 dB (Figure 4e), −7.5 dB (Figure 4f), and −14.4 dB (Figure 4g), corresponding to the reported samples, S1300, S1400, S1500, and S1600, having wider EAB of ∼4.7 GHz and very thin absorber coating thickness of 1.7 mm. The hybrid nanowires' outstanding EM wave absorption capability was attributed to their well-engineered microstructural architecture, chemical composition, and the synergistic impact between graphite and SiC.[52] These authors also suggested that the EM wave shielding performance of the fabricated hybrid nanowires could be boosted with a much lower RL value and a broader EAB by tuning the weight ratios of PCS and PVP in situ.[52] Furthermore, they indicated that using ultralong nanowires as reinforcing fillers in SiO_2, SiC, Si_3N_4, and other ceramics to improve their EM wave absorption performance as well as mechanical qualities would be exciting;[52] we suggest similar systems with the inclusion of graphene for high-performance EM wave shields.

There are reports of lightweight-com-flexible nanocomposites having a multiscale double-unbroken conductive system of TiO_2@SiO_2@PPy and sandwich structure of TiO_2@SiO_2@PPy@rGO, fabricated by electrospun TiO_2/SiO_2 delicate structured NFs.[48] The hybrid system performed as an active dissipative system, resulting in excellent EMI SE accounting for ∼30 dB in the X band as well as outstanding EMI SE of ∼13,829 dB cm^2 g^{-1} (0.089 g cm^{-3}); also, these hybrids sustained decent electrical as well as EMI SE characteristic even after repeated bending, which indicates its satisfactory flexibility being a practicable way of preparing lightweight/flexible hybrids toward high-performance shields for flexible electronics, healthcare, armed forces, devices, etc.[48]

From the available literature, the structural architecture, material defects, and functional groups of graphene and/or other included reinforcing components, along with the skin-effect/filler boundaries between the graphene materials cluster and the adopted matrix, all play vital roles in enhancing EM wave shielding performance of the electrospun graphene-based EMI shields.[50] Reports in this niche of materials are rare or limited, though there is a growing interest in the fabrication of these materials owing to their novel properties.

3.2. Graphene-Based Electrospun Nanocomposite Fibers Having Fillers for EM Wave Shields. Even though we know that electrospun virgin graphene−polymer, graphene−ceramic, etc., nanocomposite fibrous materials present certain advantages in EM wave shields, there still lingers certain challenges to be solved. Categorization of these kinds of nanofibrous systems is constrained by flexibility, transparency, mechanical properties, etc., in some instances, depending on the material entity combinations, thereby significantly limiting their applications in real life. The inclusion of novel active and/or nonactive fillers into the electrospun fibers has shown to be an efficient approach in solving some if not all of the aforementioned challenges along with greatly broadening the choice of raw materials in the case of composites. Conventional reinforcing or nonreinforcing fillers used in electrospun nanocomposites typically includes conductive (thermal or electrical) fillers for enhancing the EC, magnetic fillers for enhancing the magnetic performance, as well as others such as ceramics aimed at adjusting dielectric characteristics of these materials.

3.2.1. Graphene-Based Conductive Fillers Electrospun Composite Fibers. Thermal and electric conductive graphene-based electrospun composite fibrous materials occasionally are categorized into two categories, C-based and metal-based. Carbon nanotubes, GP, as well as transition metal oxides/carbides are among the utmost utilized C-based reinforcing filler materials for nanocomposite fiber fabrication via ES. Furthermore, metal (nano)particles are often explored as reinforcing fillers for enhancing the thermal/EC of electrospun graphene-based nanocomposite fibers. The following subsections present the diverse graphene-based electrospun nanocomposites per the available literature.

3.2.1.1. Graphene−Graphene/Graphite Electrospun Composites. Graphene−graphene electrospun composites are composites containing two or more forms of graphene in their various forms (GO, rGO, graphene (GN), GQDs, rGQDs, etc.). First, we understand that a single carbon atom has six electrons in its orbitals across the $1s^2$, $2s^2$, 2px, and 2py orbitals.[53] The amalgamation of two or more carbon atoms results in the promotion of one electron from the 2s orbital to the 2pz orbital. Graphite formation from carbon atoms results in three sp^2 orbitals created through the hybridization of carbons' $2s^2$, 2px, and 2py atomic orbitals. Carbons' sp^2-hybridized orbitals form the strong in-plane bonds, whereas the 2pz orbital creates the weaker out-of-plane bond between the amalgamated graphite sheets. The splitting apart of the weak bonds between the graphite sheets results in graphene, where the 2pz electrons no longer take part in the bonding and are what give graphene its unique exceptional electrical and optical characteristics. Graphene has been well-known (labeled and marketed) as a wonder material ever since it was first unambiguously synthesized in 2004.

Graphene nanomaterials are produced either by the top-down and/or bottom-up assembly of carbon atoms, which involves the treatment of graphite ore to synthesize graphene through processes such as CVD, chemical synthesis/chemical exfoliation, and mechanical exfoliation, although certain approaches are often amalgamated in some cases.[54]

As one of the most striking EMI shielding reinforcing fillers, GP has the capacity to induce flaws and polarization losses, which could contribute to the attenuation of EM waves. Furthermore, due to its excellent thermal conductivity, electrospun fibrous materials can be used in harsh/high-temperature situations, and GP as reinforcing fillers can improve the conductivity of electrospun nanofibrous materials while also providing flexibility.[55,56]

Reports by Guo et al.[56] on electrospun polystyrene (PS)/graphite nanoplatelets (GNPs) nanocomposite fibers, applied as EMI shields have been published. Their investigation established that the electrospun fibers upon cold-pressing were observed to be stacked, while the hot-pressing technique was utilized to fabricate well-tuned nanocomposites.[45] It was established that the EMI SE values of nanocomposites reached ∼33 dB after the inclusion of 35 wt % GNPs with a thickness of 3 mm, as a result of the creation of a conductive connective architecture as well as stratified structural architecture within the nanocomposites.[45] Also, in another work, Song et al.[57] reported an all-carbon flexible nanocomposite fibrous system for effective application as EMI shields. The authors reported that the inclusion of graphene nanosheet (GN) reinforcing fillers was associated with the polymer fibers in the ES method with the influence of van der Waals forces, as the nanocomposite fibers were carbonized to fabricate CNF−graphene nanosheet−CNF (CNF−GN−CNF) heterojunctions, as depicted in Figure 5a, showing the virgin/pristine surface was smooth, while the amount of CNF−GN−CNF heterojunctions was increased with increasing GN inclusion, as in Figure 5b−d.[57] Figure 5c established that the inclusion of CNF−GN−CNF heterojunctions significantly influences the EC of fires due to the CNF−GN−CNF heterojunctions, which modifies the interfacial interaction of the CNF structural architectures.[57] An EC of 800 S/m was attained by these authors for composite fiber with 17.2 wt % loading; nevertheless, it decreased with the inclusion of 31.9 wt % fiber due to excess GN loading, which resulted in the fracture of fibers and, consequently, the decrease in conductivity.[57] As shown in Figure 5f, CNF−GN networks with a thickness of 0.22−0.27 mm (50−60% thicker than pristine CNF networks) apparently achieved a SE total of 25−28 dB. Flexible all-carbon structure networks, according to scientists, could play an important role in portable, flexible electronic gadgets.[57]

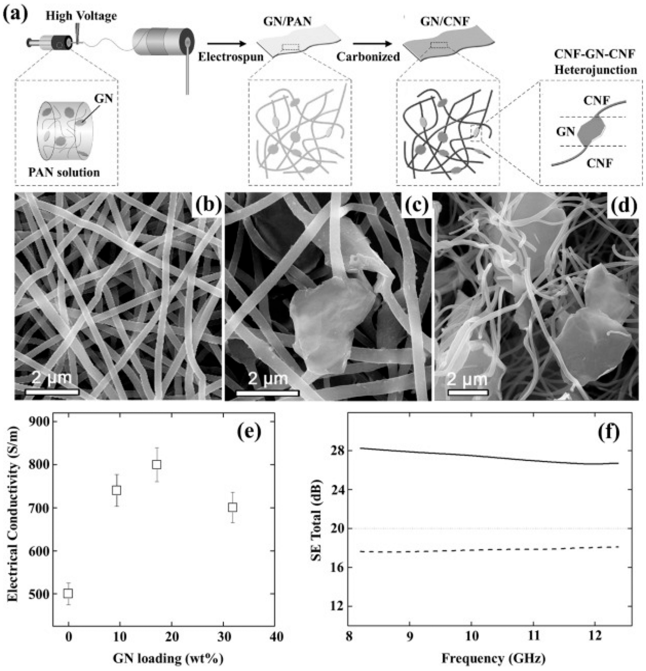


Figure 5. (a) Schematic presentation of the preparation of flexible networks and CNF−GN−CNF heterojunctions via ES. SEM images of virgin CNF networks (b) and GN/CNF composite networks with 17.2 and 31.9 wt % GN (c,d). (e) EC of the samples with different GN loadings. (f) Absolute SE of the GN/CNF nanocomposite networks with 17.2 wt % GN calculated based on its performance in virgin CNF networks. Reproduced from ref 57. Copyright 2014 American Chemical Society.

3.2.1.2. Graphene−Nanometal Electrospun Composites.

The utilization of metal (nano)particles with outstanding magnetic, EC, and nanostructural properties as fillers in the fabrication of EM wave shields via the electrospun technique has recently gained momentum owing to the nanostructural architecture attained via the ES technique. Studies patterning the EMI shielding characteristics of electrospun graphene-based systems with the inclusion of diverse metallic nanoparticles such as Cd, Cu, Co, Fe, Zn, etc. should be investigated because the diverse nanometal particle loading will reveal the noticeable impact on the EC, magnetic, and EMI SE characteristics of the final EM wave shields.[40,58,59]

Graphene offers an exceptionally ultrahigh thermal conductivity in comparison to other well-known thermally conductive fillers. Thermally conductive (Ag/rGO)/polyimide (Ag/rGO)/PI) nanocomposites prepared via electrospinning of an in situ polymerized (Ag/rGO)/polyamide electrospun suspension along with a hot-press procedure has been postulated.[45] Parts (a) and (b) of Figure 6, respectively, show the mass fraction of Ag/rGO and rGO influencing the λ values of the (Ag/rGO)/PI and rGO/PI nanocomposites and infrared thermal images of (Ag/rGO)/PI nanocomposites. The glass transition temperature (T_g), thermal conductivity (λ), as well as THRI (heat resistance index) of the (nano)composites were all enhanced by increasing the Ag/rGO nanofiller inclusion, even as the modeled data relatively agreed with the experimental findings compared to the findings obtained from reported conventional models.[60] The establishment of the importance of the electrospinning process in helping to align rGO/Ag nanofillers to get oriented along the electrospun PI fibers as well embedding of the rGO/Ag nanofillers within

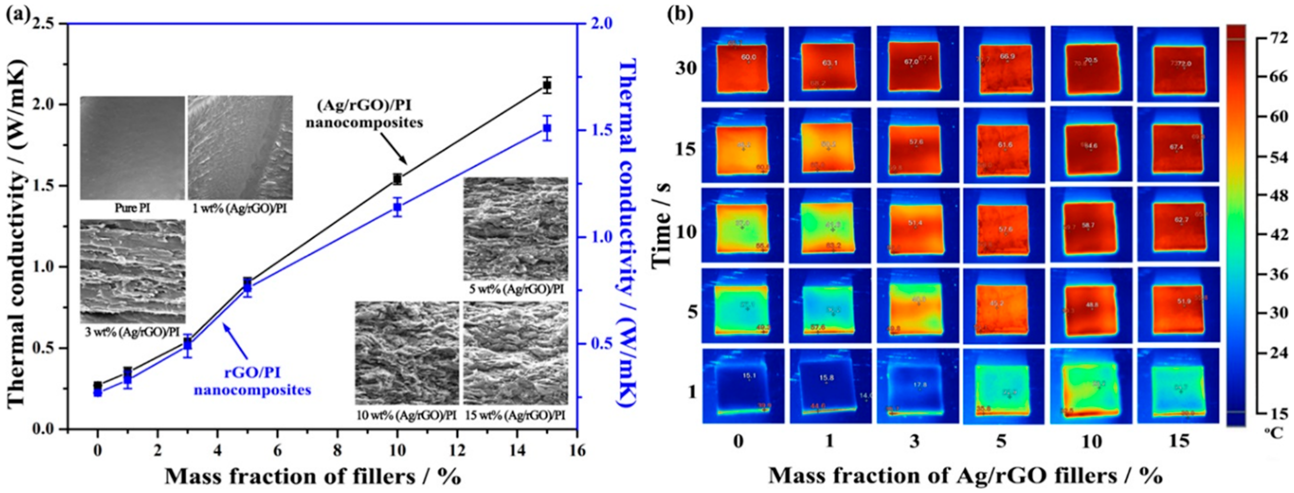


Figure 6. Thermal conductivities of the (Ag/rGO)/PI nanocomposites. (a) Mass fraction of Ag/rGO and rGO influencing the λ values of the (Ag/rGO)/PI and rGO/PI nanocomposites, respectively. (b) Infrared thermal images of (Ag/rGO)/PI nanocomposites. Reproduced from ref 60. Copyright 2019 American Chemical Society.

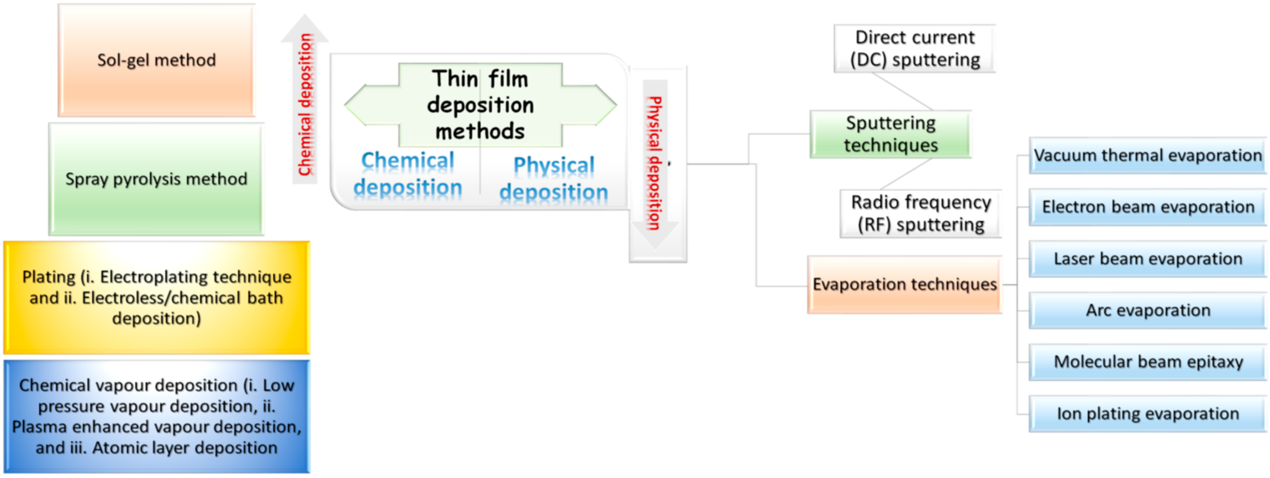


Figure 7. Scheme showing diverse deposition approaches.

or on the electrospun PI fibers was reported by these authors. They also observed that at a Ag to rGO mass ratio of 1:4, when the overall mass% of rGO/Ag nanofillers reached 15 wt %, the significant conductivity of the (rGO/Ag)/PI nanocomposites reached its peak (2.12 W/(m K)), being 8 times as extreme as virgin PI (λ of 0.27 W/(m K)).[60] The enhanced thermal conductivity implies increased EC also, though it has not been directly corrected by these authors. These kinds of formulations, if improved upon, could serve as effective materials having outstanding properties for EMI shields and other state-of-the-art applications.

A flexible PVDF film with alternate oriented graphene/Ni nanochains for EMI shielding and thermal management with excellent EC and thermal conductivity of 76.8 S/m and 8.96 W/mK has been presented in another study. The authors also discovered that by combining high EC and synergetic electromagnetic loss, as well as multilevel electromagnetic multireflection, a film with a thickness of 0.5 mm exhibits a high EMI SE of 43.3 dB, revealing a 98.6% increase over a homogeneous film with a thickness of 21.8 dB, which could be further enhanced to 51.4 dB at a thickness of 0.7 mm.[61]

Other fillers such as conductive transition metal carbides/ceramics that typically hold high stiffness as well as efficient EC than can be used as nanofillers toward the fabrication of composite fibers via ES as EMI shields have also been reported.[62] However, there is a need for in-depth study on these composites with the inclusion of graphene materials which is limited at the moment.

Also, a group of authors has revealed from their study aimed at meeting the requirement in ultrathin portable electronic devices, flexible EMI shields "CNF−GN−CNF" have a thickness of 0.22−0.27 which exhibited a mean EMI SE around 25−28 dB.[57]

4. ELECTROSPUN POLYMER GRAPHENE NANOCOMPOSITE FIBER POST-TREATMENTS FOR EMI SHIELDING

Post-treatment of electrospun polymer graphene nanocomposite fibers is aimed at amalgamating the electrospun fibers with other materials having enhanced functionality. Functionalized electrospun nanocomposite fibrous materials present better EMI shielding characteristics in comparison to virgin electrospun polymeric fibers and/or fibers with nanofillers because of the amalgamation of diverse functional entities. Conventional post-treatment approaches for processing functionalized electrospun fibrous materials could consist of diverse

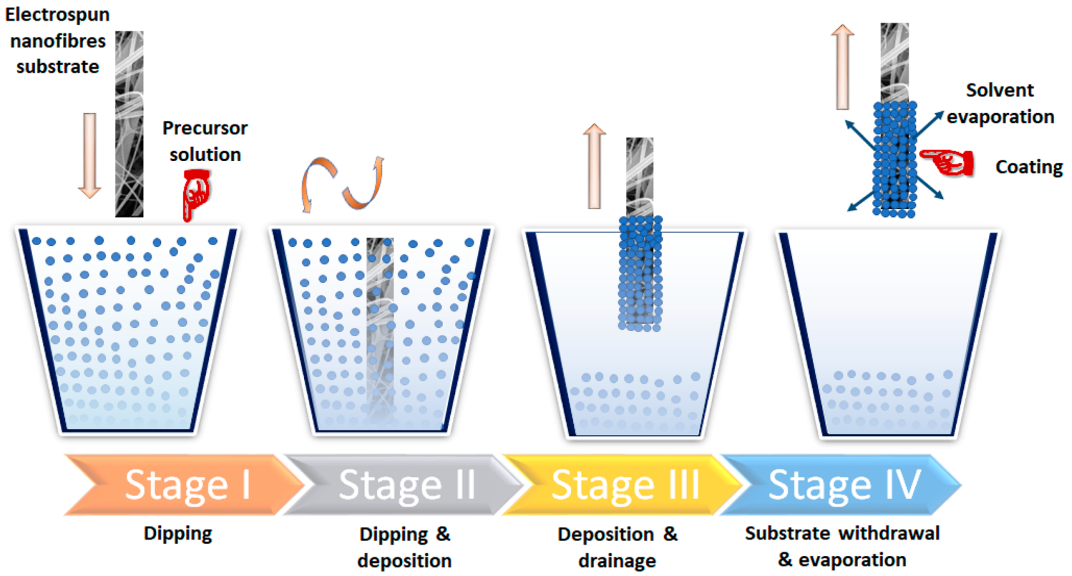


Figure 8. Schematic presentation of the dip-coating method.

deposition approaches (Figure 7), though electroless deposition, spray deposition, dip deposition, as well as vacuum deposition have been mostly utilized.

Thin film deposition is the method of applying a thin film to the surface of a substrate. It also refers to the method of depositing a fine layer of material onto another material as well as on the substratum of previous substrate material or layer upon layer. In this perspective, "thin" refers to overlay thickness within the nanometer range to a few micrometers.

This is a helpful tool for developing optics, such as reflective as well as anti-reflective paints, gadgets, including laminae of insulators, semiconductors, as well as conductors for circuit boards, and packaging, such as aluminum-plated PET membranes.

As shown in Figure 7, deposition approaches are divided into two main types premised on if the methodology is predominantly chemical and otherwise physical. In terms of deposition or coating of graphene layers on the surface of electrospun nanomaterials, methods depending on vapor deposition, sublimation/epitaxy, as well as mechanical exfoliation typically produce single-layer carbon products of high purity. Still, their scalability is now only recently being evidenced, albeit under specified circumstances.

4.1. The Dip Deposition Technique.

Dip deposition is a simplistic approach utilized toward the fabrication of fibrous electrospun nanocomposites where impregnation solution is used to wrap/coat the surface of the fibers via dip deposition. By immersion of the substrate into a solution containing hydrolyzable organic or inorganic compounds and withdrawal at constant speed into an atmosphere containing water vapor, dip-coating is a simple, inexpensive, reliable, and reproducible method for the deposition of wet liquid films. A homogeneous liquid film forms on the substrate's surface when it is removed from the solution. As the solvents are evaporated at room temperature, possible chemical reactions will take place, leaving a thin film of coating on the surface. Usually, the film is hardened/chemically transformed via heat treatment after drying in an atmosphere of water. Figure 8 reports a visual presentation of the dip-coating process. These kinds of fibrous polymeric materials coated with functional coatings through dip deposition are proven effective as a shield against EM waves.

Huang et al. have studied flexible and lightweight electrospun fibrous nanocomposite (TiO_2/SiO_2/PPy/rGo (TSPG) films (thickness of 0.26 mm and density of 0.089 g cm^{-3}) having a multiscale double-incessant structural architecture and sandwich architecture by exploiting dip deposition, displaying outstanding EMI SE.[48] These authors utilized the dip-coating technique, where PPy polymerized onto the surface of the fibers as influenced by $FeCl_3$, as well as GO plates self-assemble on both edges toward achieving functionalized fibers. Their report established that electrospun fibers having conductive coating displayed above average EMI SE properties.[48]

A facile and prevailing technique for the fabrication of elastic and lightweight graphene-based nanocomposites showing high EM wave shielding properties, produced via electrospinning of waterborne polyurethane (WPU) featuring sulfonate functionalities, has been proposed.[63] The authors revealed in their study that GO bilayers wrapped around the WPU fiber matrix displayed excellent networks steered by the electrospun WPU fibers, followed by the use of hydroiodic acid (HI) to reduce the GO to highly reduced GO (rGO), resulting in rGO/WPU nanocomposites, having significantly enhanced EC (~16.8 S/m) along with good EMI SE of ~34 dB within the X band.[63] Yuan et al. reported the electrospinning of a composite fibrous material made of $Fe_3O_4@SiO_2@PPy@GO$ (FSPG) having an innovative core−shell sandwiched amalgamated edifice fashioned via dip-disposition to be applied as EMI shields.[47] They reported the SE of FSPG film as 32 dB and total SE reaching 12,608.4 dB cm^2 g^{-1} at ~0.27 mm thick. The high SE of the hybrid was reported to be ascribed to the magnetic loss of Fe_3O_4 and graphite high λ, resulting in comparatively elevated EC of FSPG composite sheaths of ~0.71 S/cm.[47] The dip deposition process could be adopted as an effective tool for turning graphene-based electrospun fibrous materials toward the attainment of ultrahigh EMI SE systems.

4.2. Electroless Deposition.

This technique is referred to as the cheap deposition approach and could be adopted to formulate an uninterrupted as well as even a glaze on the surface of the spun substrate. Dense electroless deposited layers could boost the interfacial polarization as well as enhance the spun fibrous (nano)composite polarization loss, boosting EMI sheltering characteristics of the material. In an electroless coating, an ionic metal ion is reduced to a substrate by autocatalysis or chemical means without any external power source. This excludes approaches employed to conduct coatings without a current, such as immersion plating (deposits of copper on steel dipped in copper sulfate solution or nickel on steel dipped in chloride and boric acid bath) and homogeneous chemical reduction (silvering). In contrast to immersion plating or silvering, in which the nature of the base material itself acts as a reducing agent without further assistance, autocatalytically deposited electroless metallic coatings rely on a mechanism that is distinct from immersion plating or silvering. The term “electroless plating”, sometimes called “autocatalytic plating”, as shown in Figure 9, is about as old as electroplating. Von Liebig published for the very first time in 1835 the reduction of silver salts by reducing aldehydes. Wurtz[64] described metallic nickel plating with an electroless method from an aqueous solution in the presence of hypophosphite (reducing agent) as a chemical disaster in 1988. Roux confirmed in 1911 that there was evident precipitation of metal in the form of powder. These works, however, were not utilized in any practical applications. In this field, development received little support prior to World War II. By coating a nickel–tungsten alloy in the inner regions of tubes with a citrate solution containing an insoluble anode, Brenner and Riddell produced hypophosphite, a material with unusual reducing properties. Roux’s previous 1950 patent, where the procedure was unstructured and complete, was declared to be separate and different by the U.S. Patent and Trademark Office. On the other hand, Brenner and Riddell’s method was based on a catalytic approach that deposited only on catalytically active surfaces dipped in the plating bath.

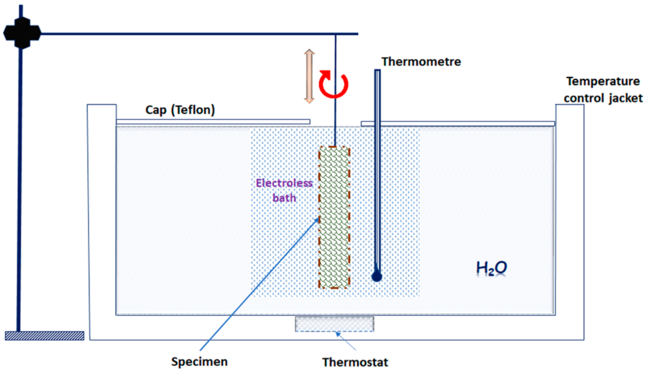


Figure 9. Schematic illustration of equipment used in electroless coating studies depicted in its most basic form.

In a similar vein to Wurtz and Roux’s attempts, Brenner later acknowledged that his invention was an unexpected result. Nevertheless, he noted that the patent was used for U.S. government protection. It is true that in 1963 a widely circulated U.S. Army-based technical study spoke largely about Wurtz and Roux’s discoveries and credited them to Brenner, for whom a patent was issued in 1956. Ultimately, this phenomenon was found to be caused by a chemical reaction involving nickel ions. The rest of the electroless coatings were applied after the electroless nickel deposit was started. A 1970 patent by Bell Laboratories led to the use of thick, pure soft gold plating for semiconductors and circuit boards. Following the invention of the first electroless gold coating bath at Bell Laboratories in 1970, semiconductors and circuit boards were plated with thick, pure soft gold.[65] According to Narcus, the electroless copper deposit was first documented, and the first profitable application was achieved by Cahill and Zeblisky et al.[66] in tartrate baths using formaldehyde as the reducing agent. Improved electroless copper formulations[67] demonstrated a greater rate of plating with highly stable conditions under a wide range of working situations. In 1954, Brenner and Riddell[68] developed electroless cobalt together with nickel deposition. These electroless cobalt deposition technologies have been widely used in the creation of magnetic films. In particular, thin films of cobalt have also been used as data storage devices due to their thinness, high coercivity, and strong remanence. Early in 1970, the practical application of electroless silver was established.[69] There was the development of electroless aluminum,[70] electroless platinum,[71] electroless ruthenium, and electroless rhodium. In early 1984, materials such as plastics, ceramics, polymers, and other nonconducting materials were electrolessly coated before being exposed to electroplating. This event resulted in the formation of the remaining electroless metallic deposits. Meanwhile, electroless coating chemistry has emerged as one of the most promising and important fields of surface development as well as metallic finishes.

This process is yet to be used in graphene-based electrospun (nano)composites but has been used for the fabrication of innovative cellular sheaths having ultrahigh EMI sheltering properties by the deposition of Cu or Ag on electrospun

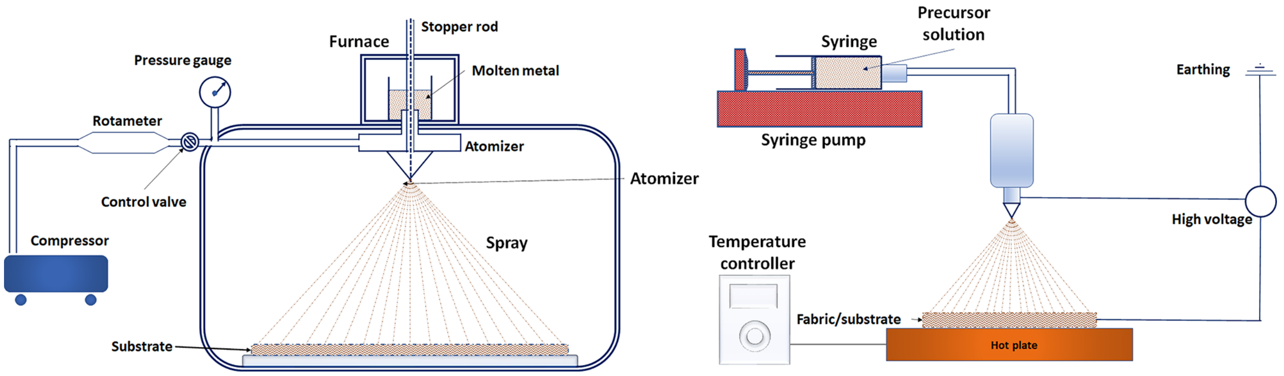


Figure 10. (a) Schematic representation of basic spray deposition technique for the deposition of metallic particles upon a substrate and (b) experimental procedure for ESD illustrated schematically.

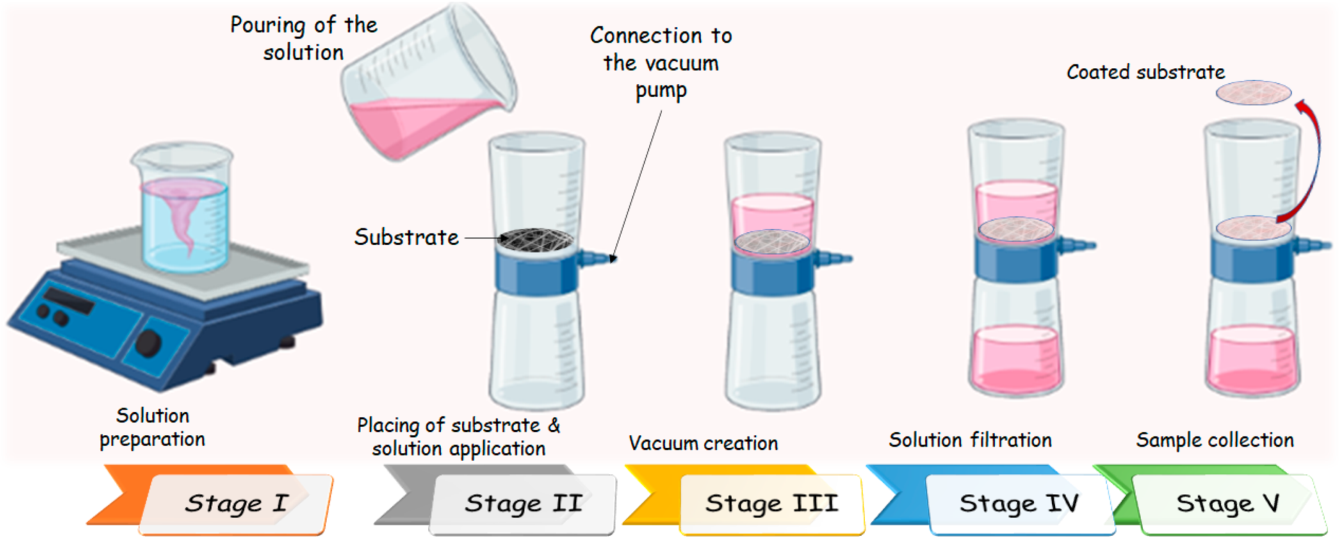


Figure 11. Scheme representing the vacuum filtration deposition technique.

polymeric NFs with the assistance of dopamine.[72] These authors reported the max EMI SE of the membranes to be ∼53 dB at a paper-thin 2.5 μm thickness and a density of 1.6 g cm^{-3}, while the specific SE attained was up to 232,860 dB cm^2 g^{-1}.[72]

Considering the outstanding results obtained with the adoption of this approach in other EMI shields designed per the literature, we believe it will effectively serve as a vital tool in tuning the properties of graphene-based EMI shields for enhanced property performance.

4.3. Spray Deposition. The spray deposition technique is extensively utilized for the deposition of thin-layer metallic layer(s) onto the surface of substrates such as electrospun NFs, where metal (nano)particles are evenly attached to the exterior portion of the electrospun NFs/substrate (Figure 10a). Boundless impact on the EM wave shielding performance of electrospun-coated fibers consisting of diverse metal nanoparticles by this technique with a difference in its volume resistivity has been explored, though largely for composite materials with nanometals without the inclusion of graphene-based materials.[73,74]

Spray deposition could be also performed using the electrostatic approach through electrostatic atomization where a droplet (positively charged) via an applied high voltage to the metallic nozzle is sprayed upon the substrate (Figure 10b); this approach is also referred to as electrostatic spray deposition (ESD). The technique is separated into three steps: the generation of an aerosol from the precursor solution, the transportation of this aerosol to the substrate's surface, and the generation of the deposit as a result of the droplets' impact on the substrates. The latter two phases are critical in the creation of the microstructure of the coating because the droplet size must be well controlled in relation to the ESD control factors. This technology has various advantages over the traditional deposition procedures, including a simple setup, inexpensive and nontoxic precursors, high deposition efficiency, direct deposition under ambient conditions, and easy control of the formed layers' surface shape.

The fabrication process for graphene/polyvinylphosphonic acid/cotton nanocomposites prepared using spray layer-by-layer assembly 10 coating cycles leading to 20 mg mL^{-1} graphene content resulting in enhanced electrical, thermal, and EMI shielding properties have been exploited.[75]

The utilization of spray deposition of rGO-based styrene-ethylene/butylene-styrene (SEBS) block copolymer coating leading to improvement in the SE to ∼19.3 dB has been established.[2] They postulated that the rGO-based coating resulted in attenuation of the EMWs, thereby enhancing the EMI SE to a large extent.

Another group of researchers examined the EMI shielding properties of hydrophobic, lightweight, and flexible graphene-coated fabric MXene−graphene−PVDF nanocomposite.[76] These authors obtained a non-3D electrospun porous architecture of MXene/graphene foam exhibiting outstanding EMI SE of ∼53.8 dB (99.999%) having a SE_R of ∼13.10 dB as well as SE_A of ∼43.38 dB within the X band with the solo glazed carbon cloth, which demonstrated a very good absolute SE of 35,369.82 dB·cm^2·g^{-1}, and we hereby hypothetically postulate that an electrospun nanocomposite having this same formulation composition will give better properties even at lower overall material density.

This technique is highly attractive for adoption in the fabrication of electrospun graphene-based nanocomposites for EM wave sheltering applications. It has been used for non-electrospun systems in this regard with outstanding performance.[76,77]

4.4. Vacuum Deposition. Other techniques such as vacuum deposition involve the use of a vacuum filtration approach to get the deposition targeted, where the force between the support material and the focus material sediment is employed to achieve the blend. Through vacuum deposition, a conductive layer is repetitively overlaid on the substrate surface to form an efficient shielding stage, while redundant EM waves can be dispelled within the substrate. We focus on the vacuum-filtration-based deposition method and its utilization within the discussed niche with its principle, as depicted in Figure 11. The depiction clearly shows that we can regulate the number as well as propagation of the operating media further into microporous material by tweaking the vacuum pressure or operation duration. In reality, of course, the only drawback is the concentration of the combination as well as the permeability of the material. The operating medium is more closely bonded with the material when it is filtered into

Table 1. Comparison of Deposition Techniques

deposition techniques	advantages	disadvantages	ref
dip deposition technique	method is inexpensive	difficult to control coating thickness	79,80
	multiple dip deposition could be used to achieve thicker films	the annealing temperature, precursor concentration, as well as solvents or additives, used all participate directly in film surface quality	
	parallel preparation of multilayers of different materials is possible		
electroless deposition	there are no issues with the current network	as the solution ages, byproducts may react, affecting the fabrication	79,81
	nanosheets of metals, alloys, and compounds could be coated on insulators such as glass, ceramics, and polymers	nontargeted nucleation as well as failure to nucleate or grow films on selected regions due to contamination or other challenges only with the catalyst surface	
	it is a simple deposition tool	the procedure takes a long time	
	has a low temperature when compared to other vacuum deposition procedures	the deposition rate is relatively low	
	less expensive method	obtaining a film thickness greater than 1 m in a single-dip deposition is difficult	
	there is no need for a vacuum environment		
	produces uniform films of high quality		
	less expensive than the traditional vacuum deposition, spray pyrolysis method		
	chemically deposited materials, such as CdS film deposition, have the lowest energy consumption per unit area of film produced		
spray deposition	a fairly large area film can be easily obtained	generally designed for the inclusion of fibers as well as whiskers, and also the matrix alloy options are limited	82
	less expensive than the traditional vacuum deposition, spray pyrolysis method	the recovery of solvents is a problem	
	there is extensive material utilization		
	scalable		
	low-temperature process, meaning less energy is required		
	other factors affecting solution-based deposition processes are avoided herewith		
vacuum deposition	increased deposition frequency	requires more understanding and research; for example, the relationships between electron density, pulse length, encasing density, as well as microstructural parameters for high-power pulsed plasma deposition microstructure as well as properties in the thickness range of 10−10,000 nm, especially for low-dimensional structures, sub-microelectronics, nanocomposites, tribological coatings, corrosion-resistant materials, reflective surfaces, and thermoelectrics	83
	diverse material properties could only be created using vacuum deposition techniques, such as coatings with properties independent of thermodynamic configurational restrictions due to the extreme versatility of the range and variety of deposited materials applied ability to vary defect concentration across a broad range, tends to result in characteristics equivalent to or far excluded from traditional bulk counterparts	ion energy impact as well as ion flux on coated material characteristics is not fully understood	
	amorphous materials can be deposited at a high quench rate	depositing species energy effect on interfacial interaction, nucleation, as well as development of the deposit is not fully understood	
	controllable generation of microstructures that differ from conventionally materials, such as ultrafine (superlattice, nanostructures) to single-crystalline films		
	enables the manufacturing of fine free-standing shapes, from brittle materials, with environmental benefits	there is also a lack of understanding on the mechanical characteristics of the deposit as well as their impact by control factors on residual stress sculpted thin films as well as highly porous membranes	
		technique costlier than atmospheric deposition technique	

the pores rather than accumulated upon the exterior. Ultimately, upgrading the layout in the figure is indeed not challenging.

This technique has been adopted for the fabrication of graphene-based EMI shields, though not electrospun nanocomposite materials with the display of interesting performance,[78] and can be adopted for fabricating electrospun graphene-based systems as EMI shields. The advantages and disadvantages of various deposition techniques are presented in Table 1.

At present, the presence of articles solely based on electrospun graphene-based EMI shield prepared with the use of the post-treatment approaches is limited. Therefore, we have also considered articles that have also used deposition-based post-treatment processes aimed at enhanced EMI shielding performance of diverse electrospun-fabricated shields. We have observed that the order is thus: Electroless deposition (EMI SE: 53−90 db)[72,84,85] > vacuum/filtration deposition (EMI SE: 42−84 db)[84,86] > spray deposition (EMI SE: 21−42 db)[73,74] > dip deposition (EMI SE: 30−40 dB).[63,87,47] This review aims to also to ignite the interest of researchers within the niche of EMI shielding materials toward utilization of these innovative approaches.

5. FACTORS AFFECTING EMI SHELTERING PERFORMANCE OF GRAPHENE-BASED ELECTROSPUN MATERIALS

Graphene-based electrospun nanocomposite fibers possess the benefits of flexibility, foldability, and/or low density and are deemed as one of the most favorable EM wave shielding materials. Unquestionably, there are additionally many components influencing the EM wave shielding properties of these electrospun fibrous materials, such as the thickness of the membranes consisting of electrospun fibers, its diameter, porosity, conductivity, and magnetism, along with the fiber interactions interfacially. The amalgamation of these aforementioned influences controls the EMI shielding performance of the consequent spun fibrous mat.

5.1. Thickness. Augmenting the width of the graphene-based spun fibrous nanocomposites as EMI shields is an effectual approach to enhancing the reflection as well as absorption operational property of the EMI shield. At a specific frequency, EM waves would be dissipated as they reaches a particular depth inside the fibrous architectural structure. An electrospun fibrous structure having a thicker layer is more capable of shielding EM waves to a higher extent. To this end, Yuan et al. comparatively studied the EMI SE of Fe_3O_4@SiO_2@polypyrrole@GO composite fibers with regard to its diverse width by assembling various pieces together and revealed that EM waves transit via thicker sandwiched structures, resulting in more loss of EM waves.[47] These authors established in their study that the material thickness was directly proportional to the EMI SE and vice versa.

Based on the above discussion, we do therefore postulate that proper management of the graphene-based electrospun materials thickness up to the threshold as per the material in question will go a long way in helping researchers and industrialists to fabricate innovative EMI shields.

5.2. Porosity Coupled with Fiber Diameter. It has been established that the major sheltering phenomenon of graphene-based electrospun nanocomposites in the EMI shielding mechanism is absorption, even though reflection also plays a part in EM wave shielding. EM waves are reflected on the fiber sheath surface; the residual EM waves would transit through the sheaths and are consequently consumed via interaction with the fibers.[47,73] Narrower diameters of these fibers can result in improved interfacial interaction between the pores/voids and spun fibers, in the interim, as the porosity (voids) gets reduced. This factor "porosity" of the electrospun fibers also influences the EM waves' dissipation within its 3D structure.[88]

Studies involving the correlation between pore sizes of electrospun graphene-based nanocomposites/hybrids have been rare/limited at present. However, Nasouri et al.[89] explored the relationships between the diameter of spun "MWCNTs/PVP" (we know that MWCNTs are composed of material properties similar to those of graphene) fibers regarding their ability to shield EMWs. Electrospun fibers having distinct diameters were prepared by altering the ES voltage, the aloofness between the drum collector and tip of the needle, and electrospun solution concentration. They established that the thicker fibers could be electrospun at higher solution concentration, hence boosting the porosity of the spun membranes, but with the EC revealing a declining trend.[89] They attributed this effect to the differences in the surface area and aperture sizes, which resulted in variations in the passage of the flow of electricity. Moreover, higher total EMI SE was achieved with increased surface area. They concluded that smaller diameter fibers imply higher surface area, leading to higher heat losses and vice versa. Chu et al.[1] in their study showed that the dielectric constant of their studied samples increased with decreased fiber diameters (i.e., by adjusting the fiber diameter, the porosity also changed with other properties), indicating that a thinner width will result in more dielectric loss, in order to trigger microwave loss.[88]

With regard to porosity, it is worth noting that the nanofibrous nonwoven perviousness, thought as the volume liberated from polymeric matrix over the all-out volume, can run somewhere in the range of 80 and 90% in a normal nanofibrous mat. This same phenomenon applies to graphene-based electrospun systems aimed at application in EMI shielding.

In graphene-based electrospun systems, there are few reports considering the influence of porosity on the EMI SE and/or the other parameters: higher porosity is directly proportional to the EM wave absorption as reported the literature.[89,90]

5.3. Magnetism and Conductivity. The use of conductive/magnetic fillers can improve the electrospun strands' overall conductivity and magnetic characteristics, as well as the EMI SE of the filaments. A higher conductivity indicates that the strands contain all of the freer electrons. EM waves will be reflected when they arrive at the outer layer of the material; in the interim, the higher porousness owing to the highly porous architecture of electrospun fibers containing active fillers having good conductivity and magnetic properties results in an advanced magnetization, enhancing the magnetic loss along with the absorption of the EM waves. The amalgamation of two or more kinds of fillers has multiple advantages for the performance of EMI shields.

Yuan et al.[47] revealed in their study of electrospun Fe_3O_4@SiO_2@polypyrrole@GO nanocomposite fibers their superparamagnetic nature toward shielding of EM waves owing to the presence of Fe_2O_4. The hybrids have a 32 dB average EMI shielding effectiveness (specific EMI shielding effectiveness can be as high as 12,608.4 dB cm^2 g^{-1}). Several other reports have established beyond doubt the effectiveness of reinforcing fillers

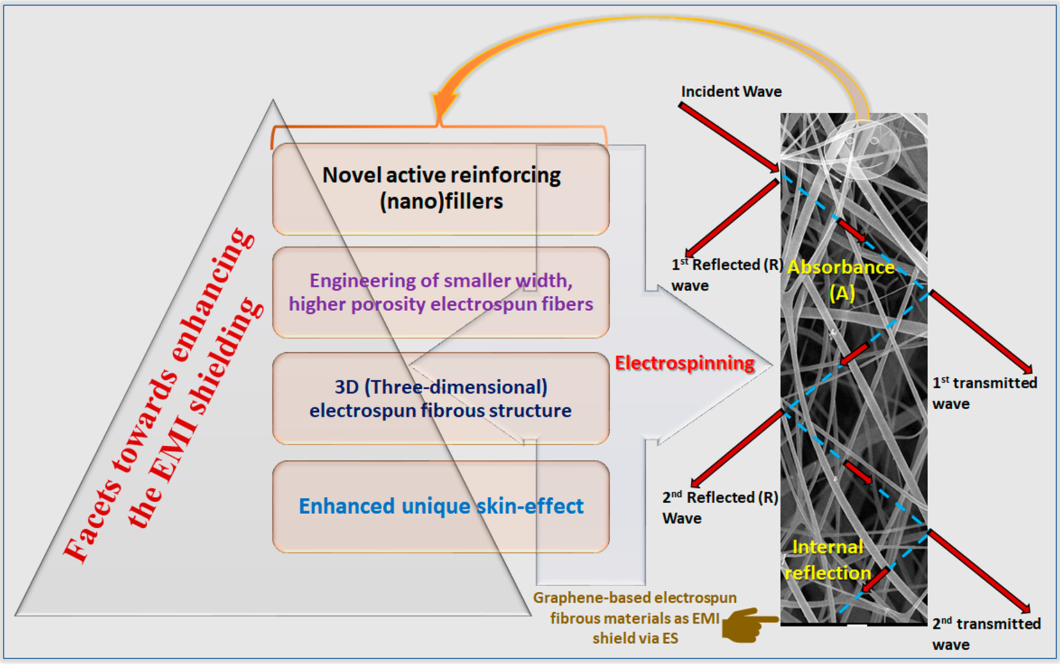


Figure 12. Selected facets toward enhancing the EM wave sheltering function of graphene-based electrospun fibrous materials.

having magnetic and electrical properties suitable to impart efficient EMI shielding properties to any graphene-based predetermined designed nanocomposite materials.[29,63,74]

With the effective and efficient amalgamation of these features with the other ones, researchers and industrialists will be able to design and engineer innovative EMI shields for even state-of-the-art applications.

5.4. Interfacial Interaction. Interfacial interaction between the filler−matrix, filler−filler, filler−other active additives within an electrospun graphene-based EMI shielding material is of great importance, as it directly influences the overall material properties leading to the attainable EMI SE.[45,88]

Extreme interfaces can result in interactions between EMWs and shields, in this manner understanding the specific scattering of EM waves. Electrospun fibers present recompenses such as high porosity, high surface area, etc. However, its high surface construction will, in general, deliver various interfaces among strands. It has been shown that the outer layer of conductive strands brimming with moving carriers can cause partial heat losses to realize the loss of EM waves.[63] Nonetheless, before EMWs are completely absorbed, a portion of them may reflect back and forth within the fiber due to the action of the fiber interface.[63] Because the porous graphene-based electrospun fiber has a low impedance, EM waves can strike the fibers, generating reflection and dissipating between the strands' edges, improving interfacial interactions. Furthermore, the stage and fiber in question exhibit interfacial contacts, which improve attack wave polarization loss.

6. CHALLENGES

With the interfacial interaction between EM waves and graphene-based electrospun fibrous materials, more attention should be given to the correlation between SE and the composite/hybrid materials compact structure. Moreover, the reinforcing active filler selection for the novel nanocomposite consisting of electrospun fibers also requires further explorational studies. Selected facets could be considered to resolve the aforementioned challenges in the forthcoming research on graphene-based electrospun fibrous materials intended to be utilized as EMI shields (Figure 12).

6.1. Enhanced Unique Interfacial Interactions (Skin-Effect) Engineering/Fabrication. Graphene-based electrospun fibrous materials having multicomponent structures have a propensity to present numerous interfaces, thereby causing more loss of EM waves. The fabrication of multicomponent structural architecture of these composites, such as coaxial, side-by-side, and/or triaxial electrospun fibers as well as multicomponent fiber-based material architecture, will promote the shielding of EM waves by these fibrous nanocomposites. Also, the design of diverse modules within the structure is also commendable of consideration. For example, concurrent inclusion of a conductive/magnetic phase into the different structural parts could give rise to the diversity of fiber functions, hence boosting EM wave attenuation. Consequently, researching the correlation between diverse structural facets and EMI shielding performance of graphene-based EMI shields is also crucial for extra research. After these missions are investigated, ES will turn out to be a more probable technique for designing and engineering EMI shields.

6.2. Engineering of Smaller Width, Higher Porosity Electrospun Fibers. Smaller width/diameter electrospun fibers having higher specific surface area possess the ability to cause higher EM waves loss, whereas dense pore structure could result in more internal interaction between EM waves and the fibers. Conversely, the width/diameter of the fibers and their porosity are intimately correlated to/with the electrospun fibrous nanocomposite thickness. Densification of the electrospun fibrous membrane pores successfully results in a direct decrease in the infiltration of EMWs into the EMI shield. Dense structured electrospun fibrous nanocomposites in this way (based on pore structure) can be engineered by post-treatment approaches such as hot pressing technology, vacuum suction filtration, and the like.

Table 2. Performances of Graphene-Based Electrospun Nanofibrous EMI Shields per the Literature

graphene/graphite-based EMI shield	cross-sectional thickness (mm)	electrical conductivity (S/cm)	EMI shielding (dB)	ref
GN/CNF composite networks	0.22−0.27	8	25−28	57
TiO_2/SiO_2/PPy/rGo film	0.26		30	48
GO (r-GO)/WPU composites	1	0.168	34	63
$Fe_3O_4@SiO_2$@PPy@Go film	0.27	0.71	32	47
PANI@PP/GPNF film	0.26	6.84	42	87
GNP/PS composite fibers	3	1	33	45
PP/GPNF-EG film	0.26	9.13	40	87
PP/GPNF/Ag film	0.33	59.199	55	87
PP/GPNF film	0.26	1.7	30	87
graphite/SiC hybrid nanowires	1.7		22	52
Ni@graphene-PVDF	0.5−0.7	76.8	51.4	61
TiO_2/SiO_2@PPy@rGO	0.26	0.39	30−38	48
SiC/rGO	1.9		56.3	50
3D PAN NFs reinforced GO nanocomposite films	0.012	172000	55−57	94
PAN-CNF reinforced bismaleimide (BMI)	2.2		23.9	95
rGO/PEDOT@PSS/PVA films	0.15	1.7	predicted to be high	96
PAN/PVP/Co, core−shell	4		RL < −10	59
PAN/PVP/ZIF-67	4		RL < −10	59
NC@Co/NC-800	4		$RL_{min} = -43.58$	59
NC@Co/NC-900	4		$RL_{min} = -55.82$	59
NC@Co/NC-1000	4		$RL_{min} = -41.12$	59
$PVDF/TiO_2-Fe_3O_4$−GO NFs	0.3		$RL_{min} = -3.5$ to -7.5	58

6.3. Novel Active Reinforcing (Nano)filler Utilization. The utilization of novel reinforcing (nano)fillers, such as nitrides, transition metal carbides, or carbonitride(s) having excellent EC in graphene-based electrospun fibrous nanocomposites toward application in EM wave shielding, is highly encouraged. For instance, 2D layered materials such as MXenes (low density, high surface area, outstanding EC, and straightforward processing) have drawn worldwide attention from researchers and industrialists for the design/engineering of efficient EMI shields.[91] Nevertheless, associated research investigations on MXenes as active reinforcing fillers in electrospun graphene-based EMI shields are rare or not reported. Hence, it will be a potential approach to integrate unique active reinforcing fillers in graphene-based spun NFs for EMI shielding materials. The challenge of reinforcing filler agglomeration in prepared solution for electrospinning that can significantly impact the ES technique along with further limiting of the large-scale fabrication of these composites requires attention. The challenge arises from poor filler dispersion/sedimentation due to a lack of proper interaction in situ between the included fillers and solution(s) and lack of sufficient electrostatic repulsion to enable the fillers to disperse more uniformly within the prepared solution. The inclusion of dispersing agent(s) or modification of the fillers through suitable addition of active surface functional groups is an effective technique toward solving the problems.

6.4. 3D Electrospun Graphene-Based Fibrous Structure. Three-dimensional electrospun graphene-based fibrous structured aerogels or sponges having highly porous architecture accelerate the entrance of EM waves into the structural network for quick dissipation, hence unveiling exceptional EM wave sheltering.[92] Though for a long time it was challenging to directly produce 3D aerogels or sponges having mechanical stable structure(s) via ES, recently, successful formation of aerogels or sponges through a mechanical cutting in solvents followed by freeze-drying of electrospun fibers has been explored.[93] These fibrous aerogels/sponges reportedly exhibited a lot of advantages, such as high explicit surface region, high porosity, abundant interface, low density, controllable functionality, and so on, favoring exceptional EMI shielding, along with anisotropic porous architecture vital for the EMI shielding achieved by steering freeze-drying of short electrospun fibers, signifying smart shielding which can also be realized in graphene-based spun nanocomposite shields through innovative formulations in the near future.

Up until now, ES technology has accomplished great advancement in diverse niches, though research in ES of graphene-based fibrous materials as EMI shields is rare and still in the development phase, which calls for extra efforts for additional practical applications. For the moment, there still exist some problems in the way of commercialization of these materials as EMI shields. Certain challenges and resolutions are offered.

One chief factor limiting the commercialization of electrospun graphene-based electrospun fibrous materials as EMI shields is their technical properties, notably their springiness. Graphene-based electrospun fibrous materials for EMI shielding made of rigid carbon NFs exhibit great restrain in applications in real-life. To resolve this challenge, these electrospun fibrous materials containing CNFs should be processed with softer matrices to enhance the stiffness of the EMI shields. Stretchable soft elastomers such as PDMS (polydimethylsiloxane) or others can be utilized as the matrix/comatrix in the preparation of these kinds of EMI shields.

Graphene-based electrospun EMI shields surface coating with organic/inorganic materials having exceptional resistance to the harsh environment (strong acid, high/low temperature, and/or strong base, etc.) will be an attractive viable solution to the challenge of using graphene-based electrospun EMI shields in harsh environments which is a challenge at the moment.

With a focus on commercialization, the cost of production is a worthy factor. The selection of appropriate materials and technological process optimization can greatly lower the production cost of these (nano)composite materials. For instance, modification on graphene-based electrospun composite NFs with the inclusion of bio-based materials is a good option.

Further considerations should be given on searching innovative graphene-based electrospun fibrous EMI shields, with regard to the modern progress in radar technology which requires innovative EMI shields for broad-band frequency of 8−26.5 GHz in the upcoming state.

The inclusion and/or total use of functional electrospun nanofibrous, especially graphene-based materials as EMI shields in microsize electronic devices, is among the foremost target recently (as presented in Table 2). High-throughput electrospinning units are also expected to open additional opportunities for graphene-based electrospun nonwoven matierals. For these purposes, the latest innovations, such as core−shelled ES, mixing and compound ES, coaxial ES, blow-assisted ES, etc., have been postulated.

7. CONCLUSIONS AND PERSPECTIVES

7.1. Conclusions. As discussed throughout the paper, nowadays the call for EM wave sheltering fabrics having the desired SE along with acceptable architectural makeup has become necessary: the design and production of innovative flexible, lightweight, and robust materials having outstanding EMI sheltering characteristics is now an imperative mission. A novel material such as 1D graphene-based electrospun fibers presents acceptable abilities in shielding EM waves, and the consolidation of nanofillers creating magnetic or dielectric dipoles could enhance microwave shielding. Posttreatment of electrospun fibrous materials to composites opens up a slew of new possibilities for creating high-performance EMI shielding materials with additional functions that are useful in a variety of applications. The compensation of materials having high specific surface area along with high aspect ratio of graphene-based spun NFs is that it makes EMI shields achieve exceptional flexibility and good porosity, along with the promotion of EM wave interactions between individual strands, accordingly, improving the ingestion of EMWs and viably acknowledging EMI protection. The presentation of the ceramic phase could further develop the impedance mismatch between the NFs. Although unadulterated carbon materials such as graphene electrospun NFs display high EMI SE, these materials' poor adaptability, as well as somewhat low dielectric character, restrains their additional utilization. The inclusion of conductive as well as magnetic nanofillers would be a viable procedure to enhance the property performance of these electrospun shields against EMI. The inclusion of conductive reinforcing (nano)fillers both enhances the dielectric performance and SE_T owing to the dielectric loss, along with enhancement of the mechanical characteristics of the electrospun nanocomposite. Moreover, the amalgamation of magnetic phases could create a magnetic loss in the case of electrospun fibers, thereby enhancing the SE_T while post-treatment of graphene-based electrospun fibrous materials further augment the EMI shield performance. Conventional post-treatment techniques such as spray deposition, electroless deposition, dip deposition, as well as vacuum deposition can be helpful in enhancing the effective influence on graphene-based electrospun fibrous materials as EMI shields. The EC, thickness, and magnetic properties of graphene-based EM wave shields are the vital factors influencing their EMI shielding characteristic as novel (nano)composites.

7.2. Perspectives. It is important to note that properties such as excellent EC, TC, in conjunction with thermal insulation are very vital and should be harnessed effectively to achieve effective EM waves shielding.

Processing techniques such as layer-by-layer have been utilized for the fabrication of graphene oxide-based nanocomposites having EC, TC, as well as thermal insulation:[97] this is then an assurance that with synergistic preparation of graphene-based hybrid materials via amalgamation of ordered electrospun layers as well as cast layers, researchers will be able to fabricate better state-of-the-art effective and efficient EMI shields for even emerging 5G signal.

With the aim of fabricating EMI shields having both excellent EMI SE as well as superior flame self-extinguishing character, carbon-based materials were endlessly dispersed in particular layered space by a group of researchers recently to construct interchanging multilayered nanocomposite at a low nanoparticle concentration, where PBS-CNT and TPU-IFR-CNT layers conversely aligned along the extrusion path.[98] The EMI shielding as well as flame retarding synergistic system could well be built in 8- and 16-layer nanocomposite by adjusting the number of layers. As a result of the impedance mismatch between adjacent layers, the average EMI SE result measured in the X band (8.2−12.4 GHz) exceeded 30 dB, far exceeding the commercial criterion of 20 dB.[98] The approach has been seconded/supported by other researchers work even with the use of different polymeric matrices.[99]

We foresee that with the amalgamation of the innovative approaches used/postulated/proven by diverse researchers with electrospinning for the fabrication of graphene-based EMI shields, robust and effective EMI shields will be fabricated by researchers which will meet the current demand of the day.

AUTHOR INFORMATION

Corresponding Author

Suprakas Sinha Ray − *Department of Chemical Sciences, University of Johannesburg, Doorfontein 2028 Johannesburg, South Africa; Centre for Nanostructures and Advanced Materials, DSI-CSIR Nanotechnology Innovation Centre, Council for Scientific and Industrial Research, Pretoria 0001, South Africa;* orcid.org/0000-0002-0007-2595;
Email: rsuprakas@csir.co.za, ssinharay@uj.ac.za

Author

Jonathan Tersur Orasugh − *Department of Chemical Sciences, University of Johannesburg, Doorfontein 2028 Johannesburg, South Africa; Centre for Nanostructures and Advanced Materials, DSI-CSIR Nanotechnology Innovation Centre, Council for Scientific and Industrial Research, Pretoria 0001, South Africa*

Complete contact information is available at:
https://pubs.acs.org/10.1021/acsomega.2c03579

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors would like to acknowledge the financial support from the Department of Science and Innovation (C6ACH20), Council for Scientific and Industrial Research, Pretoria

(086ADMIN), and the University of Johannesburg (086310), South Africa.

■ REFERENCES

(1) Chu, Z.; Cheng, H.; Xie, W.; Sun, L. Effects of diameter and hollow structure on the microwave absorption properties of short carbon fibers. *Ceram. Int.* **2012**, *38*, 4867−4873.

(2) Godoy, A. P.; Amurim, L. G.; Mendes, A.; Goncalves, E. S.; Ferreira, A.; de Andrade, C. S.; Kotsilkova, R.; Ivanov, E.; Lavorgna, M.; Saito, L. A.M.; Ribeiro, H.; Andrade, R. J.E. Enhancing the electromagnetic interference shielding of flexible films with reduced graphene oxide-based coatings. *Prog. Org. Coat.* **2021**, *158*, 106341.

(3) Orasugh, J. T.; Pal, C.; Ali, M. S.; Chattopadhyay, D. Electromagnetic interference shielding property of polymer-graphene composites. In *Polymer Nanocomposites Containing Graphene*; Rahaman, M., Nayak, L., Hussein, I., Das, N., Eds.; Woodhead Publishing, 2022; pp 211−243.

(4) Chen, Y.; Pang, L.; Li, Y.; Luo, H.; Duan, G.; Mei, C.; Xu, W.; Zhou, W.; Liu, K.; Jiang, S. Ultra-thin and highly flexible cellulose nanofiber/silver nanowire conductive paper for effective electromagnetic interference shielding. *Compos. Part A: Appl. Sci. Manuf.* **2020**, *135*, 105960.

(5) Raghavan, P.; Lim, D.-H.; Ahn, J.-H.; Nah, C.; Sherrington, D. C.; Ryu, H.-S.; Ahn, H.-J. Electrospun polymer nanofibers: The booming cutting edge technology. *React. Funct. Polym.* **2012**, *72*, 915−930.

(6) Maccaferri, E.; Mazzocchetti, L.; Benelli, T.; Zucchelli, A.; Giorgini, L. Morphology, thermal, mechanical properties and ageing of nylon 6, 6/graphene nanofibers as Nano2 materials. *Compos. B. Eng.* **2019**, *166*, 120−129.

(7) Liu, J.; Yang, K.; Mo, Y.; Wang, S.; Han, D.; Xiao, M.; Meng, Y. Highly safe lithium-ion batteries: High strength separator from polyformaldehyde/cellulose nanofibers blend. *J. Power Sources* **2018**, *400*, 502−510.

(8) Li, K.; Zhang, Y.; Xu, L.; Zeng, F.; Hou, D.; Wang, J. Optimizing stretching conditions in fabrication of PTFE hollow fiber membrane for performance improvement in membrane distillation. *J. Membr. Sci.* **2018**, *550*, 126−135.

(9) Zhang, W.; Yu, X.; Li, Y.; Su, Z.; Jandt, K. D.; Wei, G. Protein-mimetic peptide nanofibers: Motif design, self-assembly synthesis, and sequence-specific biomedical applications. *Prog. Polym. Sci.* **2018**, *80*, 94−124.

(10) Balea, A.; Fuente, E.; Tarrés, Q.; Pèlach, M. À.; Mutjé, P.; Delgado-Aguilar, M.; Blanco, A.; Negro, C. Influence of pretreatment and mechanical nanofibrillation energy on properties of nanofibers from Aspen cellulose. *Cellulose* **2021**, *28*, 9187−9206.

(11) Li, X.; Yang, L.; Lei, Y.; Gu, L.; Xiao, D. Microwave-assisted chemical-vapor-induced in situ polymerization of polyaniline nanofibers on graphite electrode for high-performance supercapacitor. *ACS Appl. Mater. Interface* **2014**, *6*, 19978−19989.

(12) Orasugh, J. T.; Ghosh, S. K.; Chattopadhyay, D. Nanofiber-reinforced biocomposites. *Fiber-Reinforced Nanocomposites: Fundamentals and Applications*; Elsevier, 2020; pp 199−233.

(13) Dey, A.; Mandal, S.; Bhandari, S.; Pal, C.; Tersur Orasugh, J.; Chattopadhyay, D. Characterization methods. *Fiber-Reinforced Nanocomposites: Fundamentals and Applications*; Elsevier, 2020; pp 7−67.

(14) Li, R.; Cheng, Z.; Yu, X.; Wang, S.; Han, Z.; Kang, L. Preparation of antibacterial PCL/PVP-AgNP Janus nanofibers by uniaxial electrospinning. *Mater. Lett.* **2019**, *254*, 206−209.

(15) Santoro, M.; Shah, S. R.; Walker, J. L.; Mikos, A. G. Poly(lactic acid) nanofibrous scaffolds for tissue engineering. *Adv. Drug Delivery Rev.* **2016**, *107*, 206−212.

(16) Xu, W.; Ding, Y.; Jiang, S.; Ye, W.; Liao, X.; Hou, H. High permittivity nanocomposites fabricated from electrospun polyimide/BaTiO3 hybrid nanofibers. *Polym. Compos.* **2016**, *37*, 794−801.

(17) Cai, N.; Chen, M.; Liu, M.; Wang, J.; Shen, L.; Wang, J.; Feng, X.; Yu, F. Meso-microporous carbon nanofibers with in-situ embedded Co nanoparticles for catalytic oxidization of azo dyes. *J. Mol. Liq.* **2019**, *289*, 111060.

(18) Gao, J.; Li, B.; Huang, X.; Wang, L.; Lin, L.; Wang, H.; Xue, H. Electrically conductive and fluorine free superhydrophobic strain sensors based on SiO_2/graphene-decorated electrospun nanofibers for human motion monitoring. *Chem. Eng. J.* **2019**, *373*, 298−306.

(19) Akshat, T.; Misra, S.; Gudiyawar, M.; Salacova, J.; Petru, M. Effect of electrospun nanofiber deposition on thermo-physiology of functional clothing. *Fibers Polym.* **2019**, *20*, 991−1002.

(20) Udangawa, R. N.; Mikael, P. E.; Mancinelli, C.; Chapman, C.; Willard, C. F.; Simmons, T. J.; Linhardt, R. J. Novel cellulose−halloysite hemostatic nanocomposite fibers with a dramatic reduction in human plasma coagulation time. *ACS Appl. Mater. Interface* **2019**, *11*, 15447−15456.

(21) Chen, Y.; Zhang, L.; Mei, C.; Li, Y.; Duan, G.; Agarwal, S.; Greiner, A.; Ma, C.; Jiang, S. Wood-inspired anisotropic cellulose nanofibril composite sponges for multifunctional applications. *ACS Appl. Mater. Interface* **2020**, *12*, 35513−35522.

(22) (a) Pan, C.-Y.; Xu, G.-R.; Xu, K.; Zhao, H.-L.; Wu, Y.-Q.; Su, H.-C.; Xu, J.-M.; Das, R. Electrospun nanofibrous membranes in membrane distillation: Recent developments and future perspectives. *Sep. Purif. Technol.* **2019**, *221*, 44−63. (b) Mishra, R. K.; Mishra, P.; Verma, K.; Mondal, A.; Chaudhary, R. G.; Abolhasani, M. M.; Loganathan, S. Electrospinning production of nanofibrous membranes. *Environ. Chem. Lett.* **2019**, *17*, 767−800. (c) Yadav, T. C.; Srivastava, A. K.; Mishra, P.; Singh, D.; Raghuwanshi, N.; Singh, N. K.; Singh, A. K.; Tiwari, S. K.; Prasad, R.; Pruthi, V. Electrospinning: an efficient biopolymer-based micro-and nanofibers fabrication technique. *Next Generation Biomanufacturing Technologies*; ACS Publications, 2019; pp 209−241. (d) Ghosh, A.; Orasugh, J. T.; Chattopadhyay, D.; Ghosh, S. Electrospun nanofibres: A new vista for detection and degradation of harmful endocrine-disrupting chemicals. *Groundw. Sustain. Dev.* **2022**, *16*, 100716.

(23) (a) Gilbert, W. *De magnete, magneticisque corporibus, et de magno magnete tellure*; Petrvs Short, 1956. Angammana, C. J.; Jayaram, S. H. Analysis of the effects of solution conductivity on electrospinning process and fiber morphology. *IEEE Trans. Ind. Appl.* **2011**, *47*, 1109−1117. (b) He, P.; Cao, M.-S.; Cai, Y.-Z.; Shu, J.-C.; Cao, W.-Q.; Yuan, J. Self-assembling flexible 2D carbide MXene film with tunable integrated electron migration and group relaxation toward energy storage and green EMI shielding. *Carbon* **2020**, *157*, 80−89.

(24) Huang, W.; Tong, Z.; Wang, R.; Liao, Z.; Bi, Y.; Chen, Y.; Ma, M.; Lyu, P.; Ma, Y. A review on electrospinning nanofibers in the field of microwave absorption. *Ceram. Int.* **2020**, *46*, 26441−26453.

(25) Huang, C.; Thomas, N. L. Fabrication of porous fibers via electrospinning: strategies and applications. *Polym. Rev.* **2020**, *60*, 595−647.

(26) Kim, J.; Lee, S.; Kim, C.; Park, Y.; Kim, M.-H.; Seol, J. H. Electromagnetic interference shield of highly thermal-conducting, light-weight, and flexible electrospun nylon 66 nanofiber-silver multi-layer film. *Polymers* **2020**, *12*, 1805.

(27) Wang, D.; Yu, J.; Duan, G.; Liu, K.; Hou, H. Electrospun polyimide nonwovens with enhanced mechanical and thermal properties by addition of trace plasticizer. *J. Mater. Sci.* **2020**, *55*, 5667−5679.

(28) Duan, G.; Fang, H.; Huang, C.; Jiang, S.; Hou, H. Microstructures and mechanical properties of aligned electrospun carbon nanofibers from binary composites of polyacrylonitrile and polyamic acid. *J. Mater. Sci.* **2018**, *53*, 15096−15106.

(29) Chaudhary, A.; Kumar, R.; Teotia, S.; Dhawan, S.; Dhakate, S. R.; Kumari, S. Integration of MCMBs/MWCNTs with Fe 3 O 4 in a flexible and light weight composite paper for promising EMI shielding applications. *J. Mater. Chem. C* **2017**, *5*, 322−332.

(30) Orasugh, J. T.; Pal, C.; Ali, M. S.; Chattopadhyay, D. 8-Electromagnetic interference shielding property of polymer-graphene composites. In *Polymer Nanocomposites Containing Graphene*, Rahaman, M.; Nayak, L.; Hussein, I. A., Das, N. C., Eds.; Woodhead Publishing, 2022; pp 211−243.

(31) Slonczewski, J.; Weiss, P. Band structure of graphite. *Phys. Rev.* **1958**, *109*, 272.

(32) Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.; Katsnelson, M. I.; Grigorieva, I.; Dubonos, S.; Firsov, a. Two-dimensional gas of massless Dirac fermions in graphene. *Nature* **2005**, *438*, 197−200.

(33) (a) Orasugh, J. T.; Ray, S. S. Prospect of DFT utilization in polymer-graphene composites for electromagnetic interference shielding application: A review. *Polymers* **2022**, *14* (4), 704. (b) Lee, C.; Wei, X.; Kysar, J. W.; Hone, J. Measurement of the elastic properties and intrinsic strength of monolayer graphene. *Science* **2008**, *321*, 385−388.

(34) Balandin, A. A.; Ghosh, S.; Bao, W.; Calizo, I.; Teweldebrhan, D.; Miao, F.; Lau, C. N. Superior thermal conductivity of single-layer graphene. *Nano Lett.* **2008**, *8*, 902−907.

(35) Blake, P.; Brimicombe, P. D.; Nair, R. R.; Booth, T. J.; Jiang, D.; Schedin, F.; Ponomarenko, L. A.; Morozov, S. V.; Gleeson, H. F.; Hill, E. W.; Geim, A. K.; Novoselov, K. S. Graphene-based liquid crystal device. *Nano Lett.* **2008**, *8*, 1704−1708.

(36) Okada, A.; Kawasumi, M.; Usuki, A.; Kojima, Y.; Kurauchi, T.; Kamigaito, O.; Schaefer, D.; Mark, J. Polymer based molecular composites. *MRS Symposium Proceedings* **1990**, *171*, 45.

(37) Spitalsky, Z.; Tasis, D.; Papagelis, K.; Galiotis, C. Carbon nanotube−polymer composites: chemistry, processing, mechanical and electrical properties. *Prog. Polym. Sci.* **2010**, *35*, 357−401.

(38) Yu, M.-F.; Lourie, O.; Dyer, M. J.; Moloni, K.; Kelly, T. F.; Ruoff, R. S. Strength and breaking mechanism of multiwalled carbon nanotubes under tensile load. *Science* **2000**, *287*, 637−640.

(39) Bhattacharyya, A.; Ghorai, S.; Rana, D.; Roy, I.; Sarkar, G.; Saha, N. R.; Orasugh, J. T.; De, S.; Sadhukhan, S.; Chattopadhyay, D. Design of an efficient and selective adsorbent of cationic dye through activated carbon - graphene oxide nanocomposite: Study on mechanism and synergy. *Mater. Chem. Phys.* **2021**, *260*, 124090.

(40) Sadhukhan, S.; Bhattacharyya, A.; Rana, D.; Ghosh, T. K.; Orasugh, J. T.; Khatua, S.; Acharya, K.; Chattopadhyay, D. Synthesis of RGO/NiO nanocomposites adopting a green approach and its photocatalytic and antibacterial properties. *Mater. Chem. Phys.* **2020**, *247*, 122906.

(41) (a) Eizenberg, M.; Blakely, J. Carbon monolayer phase condensation on Ni (111). *Surf. Sci.* **1979**, *82* (1), 228−236. (b) Somani, P. R.; Somani, S. P.; Umeno, M. Planer nano-graphenes from camphor by CVD. *Chem. Phys. Lett.* **2006**, *430*, 56−59.

(42) Wang, J.; Zhu, M.; Outlaw, R.; Zhao, X.; Manos, D.; Holloway, B.; Mammana, V. Free-standing subnanometer graphite sheets. *Appl. Phys. Lett.* **2004**, *85*, 1265−1267.

(43) Schniepp, H. C.; Li, J.-L.; McAllister, M. J.; Sai, H.; Herrera-Alonso, M.; Adamson, D. H.; Prud'homme, R. K.; Car, R.; Saville, D. A.; Aksay, I. A. Functionalized single graphene sheets derived from splitting graphite oxide. *J. Phys. Chem. B* **2006**, *110*, 8535−8539.

(44) Loh, K. P.; Bao, Q.; Ang, P. K.; Yang, J. The chemistry of graphene. *J. Mater. Chem.* **2010**, *20*, 2277−2289.

(45) Guo, Y.; Pan, L.; Yang, X.; Ruan, K.; Han, Y.; Kong, J.; Gu, J. Simultaneous improvement of thermal conductivities and electromagnetic interference shielding performances in polystyrene composites via constructing interconnection oriented networks based on electrospinning technology. *Compos. Part A: Appl. Sci. Manuf.* **2019**, *124*, 105484.

(46) (a) Salehiyan, R.; Nofar, M.; Ray, S. S.; Ojijo, V. Kinetically controlled localization of carbon nanotubes in polylactide/poly (vinylidene fluoride) blend nanocomposites and their influence on electromagnetic interference shielding, electrical conductivity, and rheological properties. *J. Phys. Chem. C* **2019**, *123*, 19195−19207. (b) Badia, J.; Teruel-Juanes, R.; Echegoyen, Y.; Torres-Giner, S.; Lagarón, J. M.; Ribes-Greus, A. Effect of graphene nanoplatelets on the dielectric permittivity and segmental motions of electrospun poly (ethylene-co-vinyl alcohol) nanofibers. *Polym. Degrad. Stab.* **2021**, *183*, 109404. (c) Darzi, M. E.; Golestaneh, S. I.; Kamali, M.; Karimi, G. Thermal and electrical performance analysis of co-electrospun-electrosprayed PCM nanofiber composites in the presence of graphene and carbon fiber powder. *Renew. Energy* **2019**, *135*, 719−728.

(47) Yuan, Y.; Yin, W.; Yang, M.; Xu, F.; Zhao, X.; Li, J.; Peng, Q.; He, X.; Du, S.; Li, Y. Lightweight, flexible and strong core-shell non-woven fabrics covered by reduced graphene oxide for high-performance electromagnetic interference shielding. *Carbon* **2018**, *130*, 59−68.

(48) Huang, L.; Li, J.; Li, Y.; He, X.; Yuan, Y. Lightweight and flexible hybrid film based on delicate design of electrospun nanofibers for high-performance electromagnetic interference shielding. *Nanoscale* **2019**, *11*, 8616−8625.

(49) Mo, R.; Yin, X.; Li, M.; Ye, F.; Fan, X.; Cheng, L. Relationship between microstructure and electromagnetic properties of SiC fibers. *J. Am. Ceram. Soc.* **2020**, *103*, 4352−4362.

(50) Wang, P.; Li, Z.; Cheng, L.; Ye, F.; Zhang, L. SiC/rGO Core−Shell Nanowire as a Lightweight, Highly Efficient Gigahertz Electromagnetic Wave Absorber. *ACS Appl. Electron. Mater.* **2020**, *2*, 473−482.

(51) Li, Y.; Meng, F.; Mei, Y.; Wang, H.; Guo, Y.; Wang, Y.; Peng, F.; Huang, F.; Zhou, Z. Electrospun generation of Ti3C2Tx MXene@graphene oxide hybrid aerogel microspheres for tunable high-performance microwave absorption. *Chem. Eng. J.* **2020**, *391*, 123512.

(52) Wang, P.; Cheng, L.; Zhang, Y.; Yuan, W.; Pan, H.; Wu, H. Electrospinning of graphite/SiC hybrid nanowires with tunable dielectric and microwave absorption characteristics. *Compos. Part A: Appl. Sci. Manuf.* **2018**, *104*, 68−80.

(53) Verma, S.; Padha, B.; Singh, A.; Khajuria, S.; Sharma, A.; Mahajan, P.; Singh, B.; Arya, S. Sol-gel synthesized carbon nanoparticles as supercapacitor electrodes with ultralong cycling stability. *Fuller. Nanotub. Carbon Nanostructures* **2021**, *29*, 1045−1052.

(54) Kour, R.; Arya, S.; Young, S.-J.; Gupta, V.; Bandhoria, P.; Khosla, A. Recent advances in carbon nanomaterials as electrochemical biosensors. *J. Electrochem. Soc.* **2020**, *167*, 037555.

(55) Shen, B.; Zhai, W.; Zheng, W. Ultrathin flexible graphene film: an excellent thermal conducting material with efficient EMI shielding. *Adv. Funct. Mater.* **2014**, *24*, 4542−4548.

(56) Cao, M.-S.; Wang, X.-X.; Cao, W.-Q.; Yuan, J. Ultrathin graphene: electrical properties and highly efficient electromagnetic interference shielding. *J. Mater. Chem. C* **2015**, *3*, 6589−6599.

(57) Song, W.-L.; Wang, J.; Fan, L.-Z.; Li, Y.; Wang, C.-Y.; Cao, M.-S. Interfacial engineering of carbon nanofiber−graphene−carbon nanofiber heterojunctions in flexible lightweight electromagnetic shielding networks. *ACS Appl. Mater. Interface* **2014**, *6*, 10516−10523.

(58) Haji Abdolrasouli, M.; Abdollahi, H.; Samadi, A. PVDF nanofibers containing GO-supported TiO2−Fe3O4 nanoparticle-nanosheets: piezoelectric and electromagnetic sensitivity. *J. Mater. Sci.: Mater. Electron.* **2022**, *33*, 5970−5982.

(59) Huang, W.; Wang, S.; Yang, X.; Zhang, X.; Zhang, Y.; Pei, K.; Che, R. Temperature induced transformation of Co@C nanoparticle in 3D hierarchical core-shell nanofiber network for enhanced electromagnetic wave adsorption. *Carbon* **2022**, *195*, 44−56.

(60) Guo, Y.; Yang, X.; Ruan, K.; Kong, J.; Dong, M.; Zhang, J.; Gu, J.; Guo, Z. Reduced graphene oxide heterostructured silver nanoparticles significantly enhanced thermal conductivities in hot-pressed electrospun polyimide nanocomposites. *ACS Appl. Mater. Interface* **2019**, *11*, 25465−25473.

(61) Liang, L.; Xu, P.; Wang, Y.; Shang, Y.; Ma, J.; Su, F.; Feng, Y.; He, C.; Wang, Y.; Liu, C. Flexible polyvinylidene fluoride film with alternating oriented graphene/Ni nanochains for electromagnetic interference shielding and thermal management. *Chem. Eng. J.* **2020**, *395*, 125209.

(62) (a) Im, J. S.; Kim, J. G.; Bae, T.-S.; Lee, Y.-S. Effect of heat treatment on ZrO2-embedded electrospun carbon fibers used for efficient electromagnetic interference shielding. *J. Phys. Chem. Solids* **2011**, *72*, 1175−1179. (b) Fan, X.; Zhan, Y.; Xu, H.; Hou, Z.; Liu, X.; Riedel, R. Highly flexible, light-weight and mechanically enhanced (Mo2C/PyC) f fabrics for efficient electromagnetic interference shielding. *Compos. Part A: Appl. Sci. Manuf.* **2020**, *136*, 105955.

(63) Hsiao, S.-T.; Ma, C.-C. M.; Liao, W.-H.; Wang, Y.-S.; Li, S.-M.; Huang, Y.-C.; Yang, R.-B.; Liang, W.-F. Lightweight and flexible

reduced graphene oxide/water-borne polyurethane composites with high electrical conductivity and excellent electromagnetic interference shielding performance. *ACS Appl. Mater. Interfaces* **2014**, *6*, 10667−10678.

(64) Wurtz, A. Sur l'hydrure de cuivre. *Ann. Chim. Phys.* **1844**, *11*, 250−251.

(65) Ali, H. O.; Christie, I. R. A review of electroless gold deposition processes. *Gold Bulletin* **1984**, *17*, 118−127.

(66) Zeblisky, R. J.; McCormack, J. F.; Williamson, J. D.; Shneble, F. W. Electroless copper plating. U.S. Patent Appl. 3095309A, 1963.

(67) Shipley, C. Historical highlights of electroless plating. *Plat. Surf. Finish.* **1984**, *71*, 92−99.

(68) Brenner, A.; Riddell, A. Electroless plating comes of age. *Metal Finishing* **1954**, *37*, 61−68.

(69) Sivertz, C. Deposition of silver. U.S. Patent Appl. 3,776,740.21, 1973.

(70) Schultz, P. B.; Yarkosky, E. F. Electroless nickel plating of aluminum. U.S. Patent Appl. 4,840,820, 1989.

(71) Rhoda, R. N.; Vines, R. F. Bath and process for platinum and platinum alloys. U.S. Patent Appl. 3,486,928, 1969.

(72) Zeng, Z.; Jiang, F.; Yue, Y.; Han, D.; Lin, L.; Zhao, S.; Zhao, Y.-B.; Pan, Z.; Li, C.; Nystrom, G.; Wang, J. Flexible and Ultrathin Waterproof Cellular Membranes Based on High-Conjunction Metal-Wrapped Polymer Nanofibers for Electromagnetic Interference Shielding. *Adv. Mater.* **2020**, *32*, 1908496.

(73) (a) Kim, H.-R.; Fujimori, K.; Kim, B.-S.; Kim, I.-S. Lightweight nanofibrous EMI shielding nanowebs prepared by electrospinning and metallization. *Compos. Sci. Technol.* **2012**, *72*, 1233−1239. (b) Yuan, W.; Yang, J.; Yin, F.; Li, Y.; Yuan, Y. Flexible and stretchable MXene/Polyurethane fabrics with delicate wrinkle structure design for effective electromagnetic interference shielding at a dynamic stretching process. *Compos. Commun.* **2020**, *19*, 90−98.

(74) Kim, H.-R.; Kim, B.-S.; Kim, I.-S. Fabrication and EMI shielding effectiveness of Ag-decorated highly porous poly (vinyl alcohol)/Fe_2O_3 nanofibrous composites. *Mater. Chem. Phys.* **2012**, *135*, 1024−1029.

(75) Nooralian, Z.; Parvinzadeh Gashti, M.; Ebrahimi, I. Fabrication of a multifunctional graphene/polyvinylphosphonic acid/cotton nanocomposite via facile spray layer-by-layer assembly. *Rsc Adv.* **2016**, *6*, 23288−23299.

(76) Raagulan, K.; Braveenth, R.; Jang, H. J.; Seon Lee, Y.; Yang, C.-M.; Mi Kim, B.; Moon, J. J.; Chai, K. Y. Electromagnetic shielding by MXene-graphene-PVDF composite with hydrophobic, lightweight and flexible graphene coated fabric. *Materials* **2018**, *11*, 1803.

(77) (a) Valles, C.; Zhang, X.; Cao, J.; Lin, F.; Young, R. J.; Lombardo, A.; Ferrari, A. C.; Burk, L.; Mülhaupt, R.; Kinloch, I. A. Graphene/polyelectrolyte layer-by-layer coatings for electromagnetic interference shielding. *ACS Applied Nano Materials* **2019**, *2*, 5272−5281. (b) Acquarelli, C.; Rinaldi, A.; Tamburrano, A.; De Bellis, G.; D'aloia, A.; Sarto, M. Graphene-based EMI shield obtained via spray deposition technique. In *2014 International Symposium on Electromagnetic Compatibility*; IEEE, 2014; pp 488−493.

(78) (a) Jyoti, J.; Arya, A. K. EMI shielding and dynamic mechanical analysis of graphene oxide-carbon nanotube-acrylonitrile butadiene styrene hybrid composites. *Polym. Test.* **2020**, *91*, 106839. (b) Chen, Y.; Zhang, H.-B.; Wang, M.; Qian, X.; Dasari, A.; Yu, Z.-Z. Phenolic resin-enhanced three-dimensional graphene aerogels and their epoxy nanocomposites with high mechanical and electromagnetic interference shielding performances. *Compos. Sci. Technol.* **2017**, *152*, 254−262. (c) Nguyen, V.-T.; Min, B. K.; Yi, Y.; Kim, S. J.; Choi, C.-G. MXene ($Ti_3C_2T_x$)/graphene/PDMS composites for multifunctional broadband electromagnetic interference shielding skins. *Chem. Eng. J.* **2020**, *393*, 124608.

(79) Eze, F. C. Electroless deposition of CoO thin films. *J. Phys. D: Appl. Phys.* **1999**, *32*, 533−540.

(80) Ahmadian-Yazdi, M. R.; Zabihi, F.; Habibi, M.; Eslamian, M. Effects of process parameters on the characteristics of mixed-halide perovskite solar cells fabricated by one-step and two-step sequential coating. *Nanoscale Res. Lett.* **2016**, *11*, 408.

(81) Grancharova, K.; Bistrev, J.; Bedikjan, L.; Spasova, G. Some properties of thin films of chemically deposited cadmium sulphide. *J. Mater. Sci. Lett.* **1993**, *12*, 852−853.

(82) (a) Abdelhalim, A.; Abdellah, A.; Scarpa, G.; Lugli, P. Fabrication of carbon nanotube thin films on flexible substrates by spray deposition and transfer printing. *Carbon* **2013**, *61*, 72−79. (b) White, J.; Willis, T. The production of metal matrix composites by spray deposition. *Mater. Des.* **1989**, *10*, 121−127.

(83) Pederson, L. R.; Singh, P.; Zhou, X.-D. Application of vacuum deposition methods to solid oxide fuel cells. *Vacuum* **2006**, *80*, 1066−1083.

(84) Ji, H.; Zhao, R.; Zhang, N.; Jin, C.; Lu, X.; Wang, C. Lightweight and flexible electrospun polymer nanofiber/metal nanoparticle hybrid membrane for high-performance electromagnetic interference shielding. *NPG Asia Mater.* **2018**, *10*, 749−760.

(85) Zhang, N.; Zhao, R.; He, D.; Ma, Y.; Qiu, J.; Jin, C.; Wang, C. Lightweight and flexible Ni-Co alloy nanoparticle-coated electrospun polymer nanofiber hybrid membranes for high-performance electromagnetic interference shielding. *J. Alloys Compd.* **2019**, *784*, 244−255.

(86) (a) Xia, Q.; Zhang, Z.; Chu, H.; Liu, Y.; Leng, J. Research on high electromagnetic interference shielding effectiveness of a foldable buckypaper/polyacrylonitrile composite film via interface reinforcing. *Composites Part A: Applied Science and Manufacturing* **2018**, *113*, 132−140. (b) Cui, C.; Xiang, C.; Geng, L.; Lai, X.; Guo, R.; Zhang, Y.; Xiao, H.; Lan, J.; Lin, S.; Jiang, S. Flexible and ultrathin electrospun regenerate cellulose nanofibers and d-$Ti_3C_2T_x$ (MXene) composite film for electromagnetic interference shielding. *J. Alloys Compd.* **2019**, *788*, 1246−1255.

(87) Lai, H.; Li, W.; Xu, L.; Wang, X.; Jiao, H.; Fan, Z.; Lei, Z.; Yuan, Y. Scalable fabrication of highly crosslinked conductive nanofibrous films and their applications in energy storage and electromagnetic interference shielding. *Chem. Eng. J.* **2020**, *400*, 125322.

(88) Huang, C.-L.; Wu, H.-H.; Jeng, Y.-C.; Liang, W.-Z. Electrospun graphene nanosheet-filled poly (trimethylene terephthalate) composite fibers: Effects of the graphene nanosheet content on morphologies, electrical conductivity, crystallization behavior, and mechanical properties. *Polymers* **2019**, *11*, 164.

(89) Nasouri, K.; Shoushtari, A. M. Effects of diameter and surface area of electrospun nanocomposite fibers on electromagnetic interference shielding. *Polym. Sci. Series A* **2017**, *59*, 718−725.

(90) Santos, J. P. F.; Arjmand, M.; Melo, G. H. F.; Chizari, K.; Bretas, R. E. S.; Sundararaj, U. Electrical conductivity of electrospun nanofiber mats of polyamide 6/polyaniline coated with nitrogen-doped carbon nanotubes. *Mater. Des.* **2018**, *141*, 333−341.

(91) Cao, M.-S.; Cai, Y.-Z.; He, P.; Shu, J.-C.; Cao, W.-Q.; Yuan, J. 2D MXenes: Electromagnetic property for microwave absorption and electromagnetic interference shielding. *Chem. Eng. J.* **2019**, *359*, 1265−1302.

(92) Liang, C.; Qiu, H.; Han, Y.; Gu, H.; Song, P.; Wang, L.; Kong, J.; Cao, D.; Gu, J. Superior electromagnetic interference shielding 3D graphene nanoplatelets/reduced graphene oxide foam/epoxy nanocomposites with high thermal conductivity. *J. Mater. Chem. C* **2019**, *7*, 2725−2733.

(93) Jiang, S.; Agarwal, S.; Greiner, A. Low-density open cellular sponges as functional materials. *Angew. Chem., Int. Ed.* **2017**, *56*, 15520−15538.

(94) Zhang, L.; Chen, Y.; Liu, Q.; Deng, W.; Yue, Y.; Meng, F. Ultrathin flexible electrospun carbon nanofibers reinforced graphene microgasbags films with three-dimensional conductive network toward synergetic enhanced electromagnetic interference shielding. *J. Mater. Sci. Technol.* **2022**, *111*, 57−65.

(95) Ma, Y.; Zhuang, Y.; Li, C.; Shen, X.; Zhang, L. Improving Electromagnetic Interference Shielding While Retaining Mechanical Properties of Carbon Fiber-Based Composites by Introducing Carbon Nanofiber Sheet into Laminate Structure. *Polymers* **2022**, *14*, 1658.

(96) Yin, J.; Bai, Y.; Lu, J.; Ma, J.; Zhang, Q.; Hong, W.; Jiao, T. Enhanced mechanical performances and high-conductivity of rGO/

PEDOT:PSS/PVA composite fiber films via electrospinning strategy. *Colloids Surf. A: Physicochem. Eng. Asp.* **2022**, *643*, 128791.

(97) Zhang, X.; Zhang, X.; Yang, M.; Yang, S.; Wu, H.; Guo, S.; Wang, Y. Ordered multilayer film of (graphene oxide/polymer and boron nitride/polymer) nanocomposites: An ideal EMI shielding material with excellent electrical insulation and high thermal conductivity. *Compos. Sci. Technol.* **2016**, *136*, 104−110.

(98) He, L.; Shi, Y.; Wang, Q.; Chen, D.; Shen, J.; Guo, S. Strategy for constructing electromagnetic interference shielding and flame retarding synergistic network in poly (butylene succinate) and thermoplastic polyurethane multilayered composites. *Compos. Sci. Technol.* **2020**, *199*, 108324.

(99) Ji, X.; Chen, D.; Shen, J.; Guo, S. Flexible and flame-retarding thermoplastic polyurethane-based electromagnetic interference shielding composites. *Chem. Eng. J.* **2019**, *370*, 1341−1349.