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Construction strategies and recent advances of flexible EMI phase change composites

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Abstract

With the continuous development of small and medium-sized electronic devices, which bring convenience to people's lives, electromagnetic wave (EMW) pollution has emerged as a significant issue. The development of materials with electromagnetic interference (EMI) shielding capabilities for protection against harmful radiation plays a vital role. Currently, a wide range of multifunctional, lightweight EMI shielding materials have been created to address various environmental requirements. However, a single EMI shielding material has been difficult to meet the requirements of high-speed transmission of electromagnetic radiation of electronic equipment because when such devices operate at high speeds, they typically generate elevated temperatures, and excessive electromagnetic radiation further exacerbates heat accumulation, reducing both efficiency and lifespan. Therefore, thermal management is essential to lower operating temperatures and ensure optimal performance. Phase change materials (PCMs) are known for storing a large amount of energy, and have significant potential in thermal management, so flexible EMI phase change composites (PCCs) have emerged. This review provides a detailed examination of flexible EMI shielding materials based on various fillers, the potential of flexible PCMs in thermal management, and the latest advancements in developing new lightweight EMI PCCs. Finally, we suggest some potential research directions for flexible EMI shielding PCCs, hoping to contribute to the rapid advancement of next-generation flexible electronics, human thermal management, and artificial intelligence.

Keywords: Electromagnetic interference shielding, phase change composites, polymer matrix, flexible

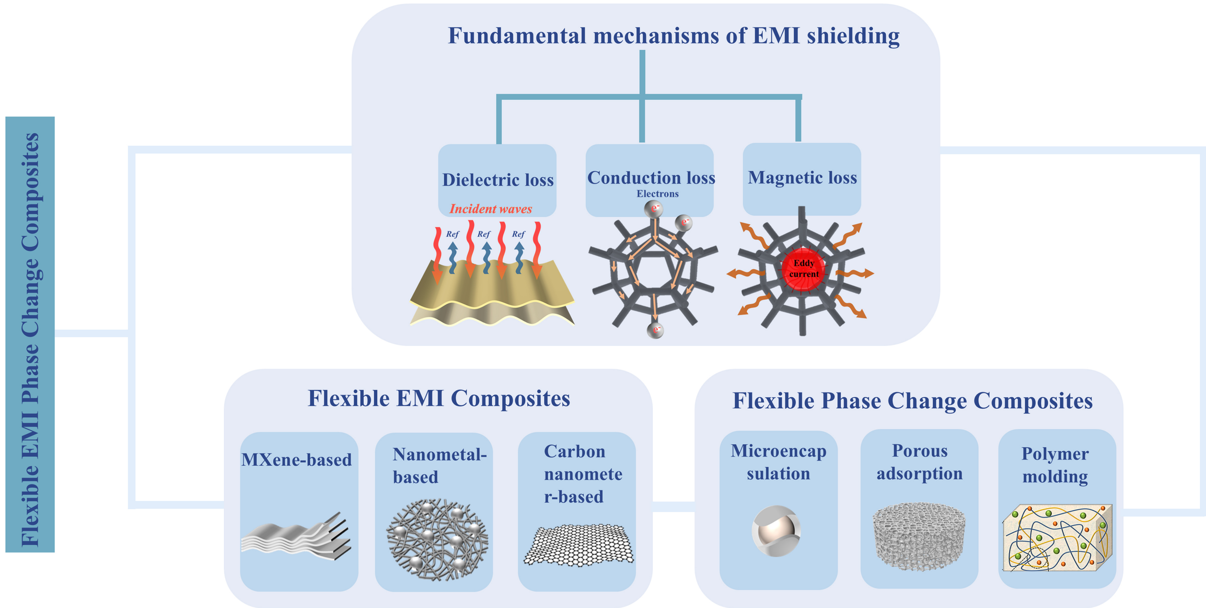
INTRODUCTION

As electronic devices and communication technology advance rapidly, the transmission density of electromagnetic waves (EMWs) has increased, and electromagnetic radiation has gradually become an urgent issue in using new energy[1-7]. On the one hand, unnecessary EMWs can interfere with the normal operation of electronic devices, thereby reducing their lifespan. On the other hand, excessive exposure of the human body to EMWs can lead to various health issues, such as a higher risk of nausea, dizziness, palpitations, insomnia, and even reduced immunity[8-10]. Therefore, electromagnetic interference (EMI) shielding has become a significant focus of research, and the design of materials with efficient EMI shielding has great prospects for development[11-14].

Traditional EMI shielding materials such as metals (Cu, Al, Ni, *etc*.) are widely used because of their outstanding electrical conductivity and fast electron transfer rate. However, their high density, susceptibility to corrosion, processing challenges, and limited flexibility constrain their application in small and medium-sized electronic devices. Therefore, designing lightweight, flexible EMI shielding materials is urgent and essential[15-17]. Polymers offer unique advantages, including low density, corrosion resistance, high mechanical strength, and adjustable elasticity[18,19]. They are considered potential candidates for flexible EMI shielding material substrates and are favored by numerous researchers[20-22]. Intrinsically conductive polymers such as polyacetylene, polypyrrole (PPy), polyaniline, and polythiophene can significantly improve conductivity through doping, thereby exhibiting excellent EMI shielding behavior and effectively mitigating secondary EMI pollution by absorbing losses[23,24]. However, their molecular chains are generally rigid, which complicates processing. In contrast, the addition of conductive fillers to traditional polymers not only enables flexible design and easier processing but also enhances conductivity, thereby improving EMI shielding performance[25-30]. Various fillers with excellent conductivity, including MXene[31-33] (transition metal carbides and nitrides), carbon-based materials[34-38] [graphene, carbon nanotubes (CNTs), carbon black], and metal nanoparticles[39-43] [silver nanowires (AgNWs), Ag, liquid metals], have been used in conjunction with polymer matrices to prepare lightweight and effective EMI shielding materials.

EMW overload is often accompanied by heat accumulation, which can lead to overheating electronic devices, thereby reducing their precision and potentially damaging them. As a result, a single EMI shielding material cannot fully meet the demands of modern electronic devices. The development of flexible materials that provide both electromagnetic shielding and thermal management performance (TMF-EMI) has thus become a major research focus[44-48]. Phase change materials (PCMs) exhibit excellent properties, such as excellent thermal storage, stable thermochemical characteristics, and adjustable phase change temperatures, making them promising for thermal management and waste heat recovery in electronic devices[49-51]. However, issues such as leakage and poor thermal conductivity of PCMs pose significant challenges to their practical application. These problems can be addressed by encapsulating the materials and incorporating thermally conductive fillers[52-55]. Consequently, combining PCMs with lightweight EMI materials to construct flexible EMI shielding thermal management composites is no longer difficult.

In response to this, the article offers a systematic summary of recent advances in flexible EMI shielding materials and the potential applications of PCMs for thermal management and provides a comprehensive introduction to lightweight EMI shielding composites with thermal management capabilities (The structure



Scheme 1. Schematic diagram of the article structure.

of the article is shown schematically in Scheme 1). Additionally, the review also deeply researches and analyzes the fundamental methods and mechanisms of EMW energy conversion and dissipation in various situations, and finally gives an outlook on their development trends and challenges, which provides certain guidance for the design, development, and application of novel EMI shielding composites with thermal management.

FUNDAMENTAL MECHANISMS OF EMI SHIELDING

EMI shielding refers to the prevention of EMWs from propagating through the transmission path. The degree of attenuation of EMWs is represented by shielding effectiveness (SE)[56,57]. According to Schelkunoff's theory of plane-wave electromagnetic transmission, when an EMW reaches the surface of a shielding material, part of the wave is directly reflected (SE_R) due to an impedance mismatch, while the remainder enters the material. Within the material, attenuation occurs due to absorption (SE_A) and multiple reflections (SE_M) [typically, SE_M is negligible when the SE (SE_T) exceeds 15.00 dB][58-60]. Finally, the residual EMW passes through the shielding layer, during which the attenuation process of the shielding material for the EMW includes reflection, absorption, and internal multiple reflection loss, and the electromagnetic shielding capability is expressed in terms of the SE (SE_T), which is given by

$$SE_T = SE_A + SE_R \tag{1}$$

The shielding behavior of a material is usually obtained from the S-parameters (S_{11}, S_{21}, S_{12}, S_{22}), according to which the corresponding reflection loss (R), absorption loss (A), and emission loss (T) can be calculated by

$$R = |S_{11}|^2 = |S_{22}|^2 \tag{2}$$

$$T = |S_{12}|^2 = |S_{21}|^2 \tag{3}$$

$$T + A + R = 1 \tag{4}$$

$$SE_A - 10\log\left(\frac{T}{1-R}\right) \tag{5}$$

$$SE_R - 10\log(1-R) \tag{6}$$

According to these formulas, when A > R, the material is an absorption-dominated shielding material, and vice versa for reflection-dominated shielding materials[61-63]. The loss mechanisms are generally categorized into dielectric loss, conduction loss, and magnetic loss, all of which influence EMW attenuation to varying degrees.

Dielectric loss

Dielectricity refers to a material's ability to dissipate electrostatic energy when exposed to an electromagnetic field. It is typically expressed through the dielectric constant and dielectric loss. Dielectric materials display electronic polarization, atomic polarization, and interface polarization in the presence of electromagnetic fields. When exposed to external electromagnetic fields, the dipoles in dielectric materials align, causing molecular spin and frictional collisions between adjacent molecules. This process converts EMW energy into heat, thereby dissipating EMW effectively[64-66].

Conduction loss

EMI shielding materials are required to have good electrical conductivity which demonstrates high reflectivity of EMWs. When the EMW enters the material, it undergoes multiple reflections, causing the energy of the wave to decay. Meanwhile, the material's internal eddy currents are conducted and transformed into thermal energy through the material's resistance, thereby dissipating electromagnetic energy[67,68].

Magnetic loss

Magnetic materials exhibit strong magnetization when subjected to an external magnetic field, greatly enhancing the strength of the internal magnetic field[69]. In an alternating magnetic field, the change in magnetic induction lags behind the phase of the magnetic field, a phenomenon known as hysteresis and the energy dissipated due to hysteresis is termed magnetic loss[70,71]. In the dynamic magnetization process, energy losses occur not only due to hysteresis but also from eddy currents and natural resonance.

Eddy current loss is defined as the energy dissipation that takes place when a conductive magnetic material is exposed to an alternating magnetic field[72]. An induced current is generated during this process, effectively converting magnetic field energy into electric field energy. The eddy current effect influences the absorption of EMWs in two ways: first, energy is dissipated during the conversion from magnetic energy to electrical energy. Second, eddy currents change the dielectric characteristics of the material, leading to a greater impedance mismatch, causing more EMWs to be reflected at the material surface, effectively resisting the transmission of EMWs[73].

FLEXIBLE EMI PHASE CHANGE COMPOSITES

Flexible EMI composites

Flexible electronic devices offer significant advantages in the information age due to their lightweight nature. They must demonstrate excellent flexibility, stretchability, and ease of processing[74]. Currently, a series of flexible materials have been developed using various components. Among these, polymers with low density, corrosion resistance, high mechanical properties, adjustable elasticity and other unique advantages

Table 1. Properties of different fillers

EMI fillers	Properties	Disadvantages
MXene	High electrical conductivity flexible size adjustment abundant surface functional groups	Prone to oxidation
Nanometal particles	High electrical	Easy to reunite
AgNWs	High electrical High aspect ratio	Low adhesion Atmospheric corrosion
Carbon nanomaterials	High specific surface area High electrical High thermal conductivity Increase roughness Durability	Easy to reunite

EMI: Electromagnetic interference; AgNWs: silver nanowires.

are considered potential candidates for flexible substrates[75,76]. These polymers are then supplemented with a range of excellent conductive fillers [such as MXene, carbon materials (graphene, CNTs), metal nanoparticles] to prepare composite materials with extremely efficient EMI shielding effect (The different filler properties are detailed in Table 1).

MXene-based flexible EMI materials

MXene is a new family of two-dimensional nanomaterials known for their excellent electrical conductivity, flexible size adjustability, strong surface plasmon resonance effects, and abundant surface functional groups, which have unique properties in EMI shielding[77,78]. Additionally, it has been reported that loading MXene nanomaterials onto polymeric porous materials can effectively form a conductive network, resulting in high-performance, lightweight EMI shielding materials[79-81]. Zeng *et al*. prepared a lightweight, flexible, and robust MXene-coated polyimide (PI) (C-MXene@PI) porous composite by chemical crosslinking techniques and dip coating [Figure 1A][82]. The material is hydrophobic, oxidation-resistant, and high-temperature stable, and achieves exceptional EMI shielding performance in the X-band thanks to the combined effects of the micrometer-sized pores, MXene-based conductive network, and interfacial polarization between MXene and PI, with an impressive SE of up to 62.50 dB. Moreover, the material also exhibits excellent electro-thermal properties with an outstandingly fast electro-thermal effect and more uniform thermal distribution at low voltage and can be securely fastened to the skin to evaluate its electromechanical sensing capabilities, proving the sensitivity and dependability of the wearable sensor in detecting human movements, the composites are anticipated to play a significant role in smart gadgets and flexible electronics of the future. Liu *et al*. prepared flexible and hydrophobic porous MXene foams from thin films assembled with lamellar MXene by a hydrazine-induced foaming process [Figure 1B][83]. The material not only exhibits excellent water resistance and durability, but also will play a great role in the fields of defense, aerospace, and smart wearable electronics due to its highly porous structure that exhibits higher EMI shielding (70.00 dB) compared with unframed. Zhang *et al*. prepared polyvinyl alcohol (PVA)-based EMI shielding composite films with electrostatic spinning, lay-up, and hot-pressing techniques [Figure 1C][84]. The top and bottom layers contain electrostatically spun nanofibers of the magnetic substance Fe_3O_4/PVA composite, and the conductive filler MXene/PVA composite electrostatically spun nanofibers are in the intermediate layer, which constructs the process of "absorption-reflection-re-absorption" of EMWs in the sandwich-structured composite film and realizes the unique interface polarization between the highly conductive layer and the anti-match layer. This interfacial polarization enhances EMI shielding performance, achieving a SE of 40.00 dB. Meanwhile, the sandwich structure EMI shielding composite film exhibits excellent thermal conductivity and mechanical properties and can cope with diverse changing scenarios, so it has broad application potential in the field of EMI shielding and human body protection of high-power, lightweight, wearable and flexible electronic devices.

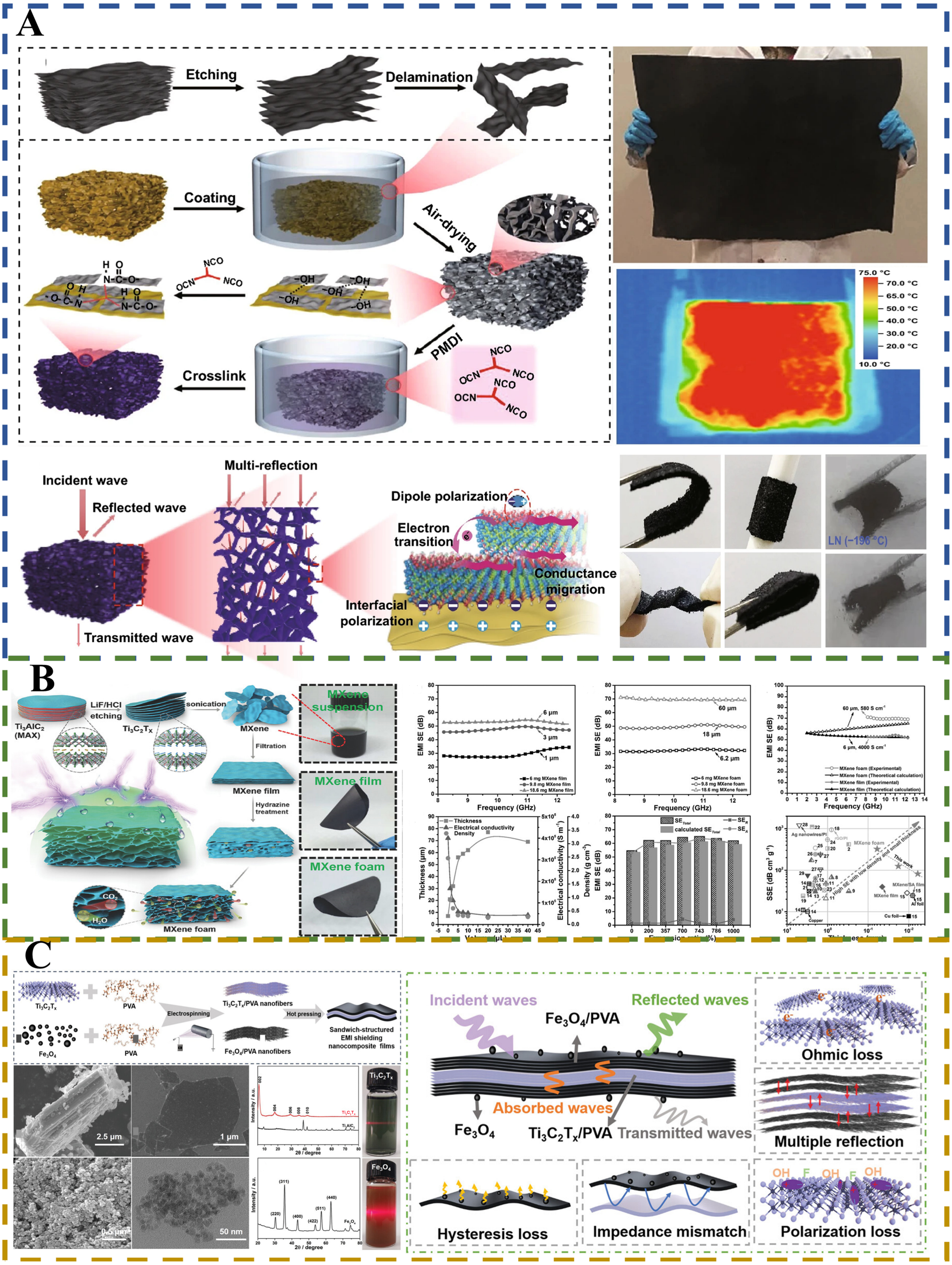


Figure 1. Overview of MXene-based flexible electromagnetic shielding materials. (A) MXene@PI porous composites preparation process, digital photos, and EMI shielding mechanism diagrams. Reproduced with permission from ref[82].Copyright 2022, Springer Nature Link; (B) MXene foam preparation, digital images, and EMI data. Reproduced with permission from ref[83].Copyright 2017,

Wiley-VCH; (C) Preparation of sandwich-structured EMI shielding nanocomposite films and EMI Shielding Mechanisms. Reproduced with permission from ref[84]. Copyright 2021, Wiley-VCH. PI: Polyimide; EMI: electromagnetic interference.

Nanometal-based flexible EMI materials

Recent studies have reported that metal particles, such as silver (Ag), are effective in enhancing EMI shielding performance through chemical deposition without significantly increasing weight[85]. Ag typically grows as particles when it is deposited on porous materials, and the formation of conductive pathways is influenced by the distance between two adjacent Ag particles, which in turn affects the overall electrical conductivity of the material[86]. Xing *et al*. combined chemical silver plating with an ultrathin and flexible carbon fabric/Ag/waterborne polyurethane (CEF-NF/Ag/WPU) film was prepared by chemical silver plating combined with an enhanced pressing process [Figure 2A][87]. The film exhibited a conductivity of 11,986.8 S/cm at 0.183 mm, attributed to the deposition of Ag particles, which form extended pathways for faster electron transport. Furthermore, the material's capacity to attenuate EMWs through internal multiple reflections and absorptions was greatly improved by the rough surface created by Ag deposition and the porous three-dimensional (3D) structure of the Ag-coated fibers, resulting in an EMI SE of 102.90 dB. Furthermore, the film demonstrated excellent reliability following ultrasonic treatment, flexural testing, and thermal reliability assessments, suggesting a great deal of promise for use in flexible materials and wearable technology.

Meanwhile, it is also possible to create high-performance EMI shielding materials using AgNWs with a high aspect ratio, tiny diameter, and high conductivity[88]. In addition, its incorporation into a flexible polymer matrix in combination with MXene may offer a cost-effective approach for EMI shielding applications. Liang *et al*. got highly conductive AgNWs using a polyol method and prepared multifunctional flexible nanowires based on vacuum-assisted filtration and hot pressing[89]. A multifunctional flexible EMI shielding AgNWs/cellulose film was prepared by hot pressing [Figure 2B]. AgNWs were uniformly embedded within the cellulose matrix, forming a 3D network. Due to their ultrahigh electrical conductivity (5,571 S/cm), achieving an EMI SE of 101 dB at a thickness of just 44.5 μm, the highest reported for materials of comparable thickness. Additionally, the AgNWs/cellulose film has a high tensile strength of 60.7 MPa and a tensile modulus of 3.35 GPa due to the many hydrogen bonds that exist between the AgNWs and the cellulose matrix, rendering it ultra-flexible for applications in various extreme environments. The film also exhibits excellent thermal conductivity and outstanding Joule heating properties, effectively dissipating heat accumulation. With its sensitive temperature responsiveness and outstanding stability at safe operating voltages for the human body, it holds considerable potential for use in wearable electronics, artificial intelligence, and the communications sector. Cheng *et al*. combined MXene with AgNWs and constructed a sandwich composite film by coating AgNWs@MXene on the core material of transparent wood (TW) [Figure 2C][90]. Among them, the ordered microchannel array in TW induces multiple reflections of EW to improve EMI shielding. Furthermore, MXene and AgNWs synergistically enhance the conductivity and stability of the material, thereby inducing interfacial polarization that increases absorptive shielding, which results in an average EMI shielding effect of up to 44.00 dB in the X-band (8-12.4 GHz). Cheng *et al*. prepared ultrathin flexible polyvinylidene difluoride (PVDF) films of MXene composite AgNWs by solid solution casting [Figure 2D][91]. The synergistic effect induced by the MXene/AgNWs network resulted in higher SE (41.26 dB) at an ultrathin thickness (600 μm), which was mainly attributed to the fact that the MXene nanosheets and AgNWs could increase the number of free electrons interacting with the electromagnetic radiation and thus improve the substantial absorption of incident radiant energy. Additionally, residual EMWs entering the interior are dissipated through continuous reflection within the 3D interconnected network structure between the MXene nanosheets and AgNWs, and only a small amount of EMWs are eventually transmitted. Furthermore, the 3D interconnected network composed of

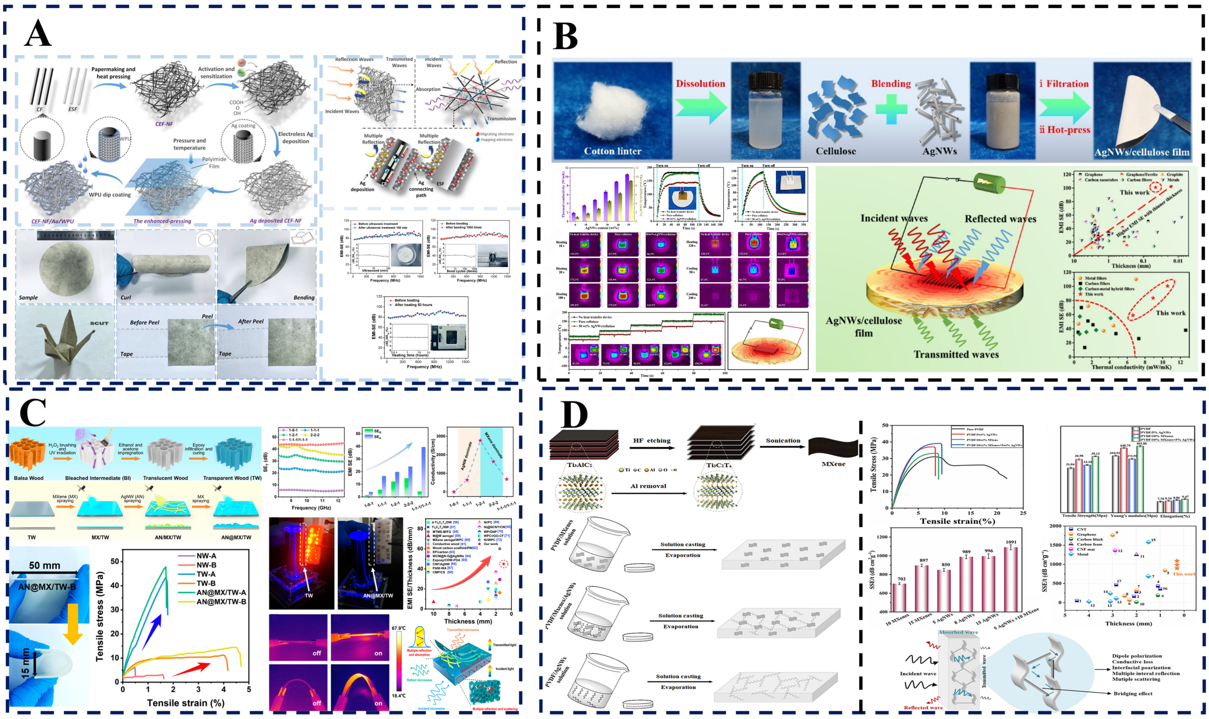


Figure 2. Flexible EMI materials based on Ag nanoparticles. (A) Preparation of CEF-NF/Ag/WPU thin films, digital photographs, and EMI mechanisms. Reproduced with permission from ref[87]. Copyright 2020, Elsevier; (B) Preparation, electrical and thermal properties, and EMI shielding mechanism of AgNWs/cellulose thin films. Reproduced with permission from ref[89]. Copyright 2020, American Chemical Society; (C) Preparation, mechanical properties, EMI shielding effect, and mechanism of AN@MX/TW. Reproduced with permission from ref[90]. Copyright 2022, American Chemical Society; (D) Preparation, mechanical properties, and EMI shielding mechanism of PVDF/MXene/AgNWs films. Reproduced with permission from ref[91]. Copyright 2021, Springer Nature Link. EMI: Electromagnetic interference; CEF-NF/Ag/WPU: flexible carbon fabric/Ag/waterborne polyurethane; AgNWs: silver nanowires; TW: transparent wood; PVDF: polyvinylidene difluoride.

AgNWs and MXenes enhances the material's mechanical characteristics and thermal conductivity, which is favorable for coping with a variety of complex mechanical environments and improving thermal dissipation, and it has a great application prospect in ultra-thin flexible wearable electronics.

Nanocarbon-based flexible EMI materials

Nanocarbons such as graphene and CNTs, are promising EMI fillers due to their excellent electrical conductivity, which possess a high specific surface area, superior thermal and electrical conductivity[92-94], and have been extensively researched and developed as lightweight and efficient EMI shielding materials. Among them, graphene is widely used as an EMI shielding material due to its excellent thermal and electrical conductivity, which enables it to effectively absorb and reflect EMWs[95-97]. Song *et al.* obtained cellulose graphene carbon aerogel (CCA@rGO) by vacuum impregnating graphene oxide (GO) solution with cellulose aerogel (CA), continued freeze-drying and thermal annealing and finally backfilled with polydimethylsiloxane (PDMS) to obtain ultra-highly elastic, lightweight EMI shielding composites [Figure 3A][98]. The epidermal core structure of CCA@rGO allows the CCA (first conductive network) wrapped with reduced GO (rGO) to gradually form a flawless second conductive network. A perfect 3D double-layer conductive network is then produced by combining the two conductive networks. Increasing the interfaces between rGO and CCA, rGO and rGO, and CCA@rGO and PDMS matrix simultaneously improves the conductive loss, impedance mismatch, and interfacial polarization loss between the CCA@rGO/PDMS EMI shielding composite material and the incident EMW. Moreover, the material

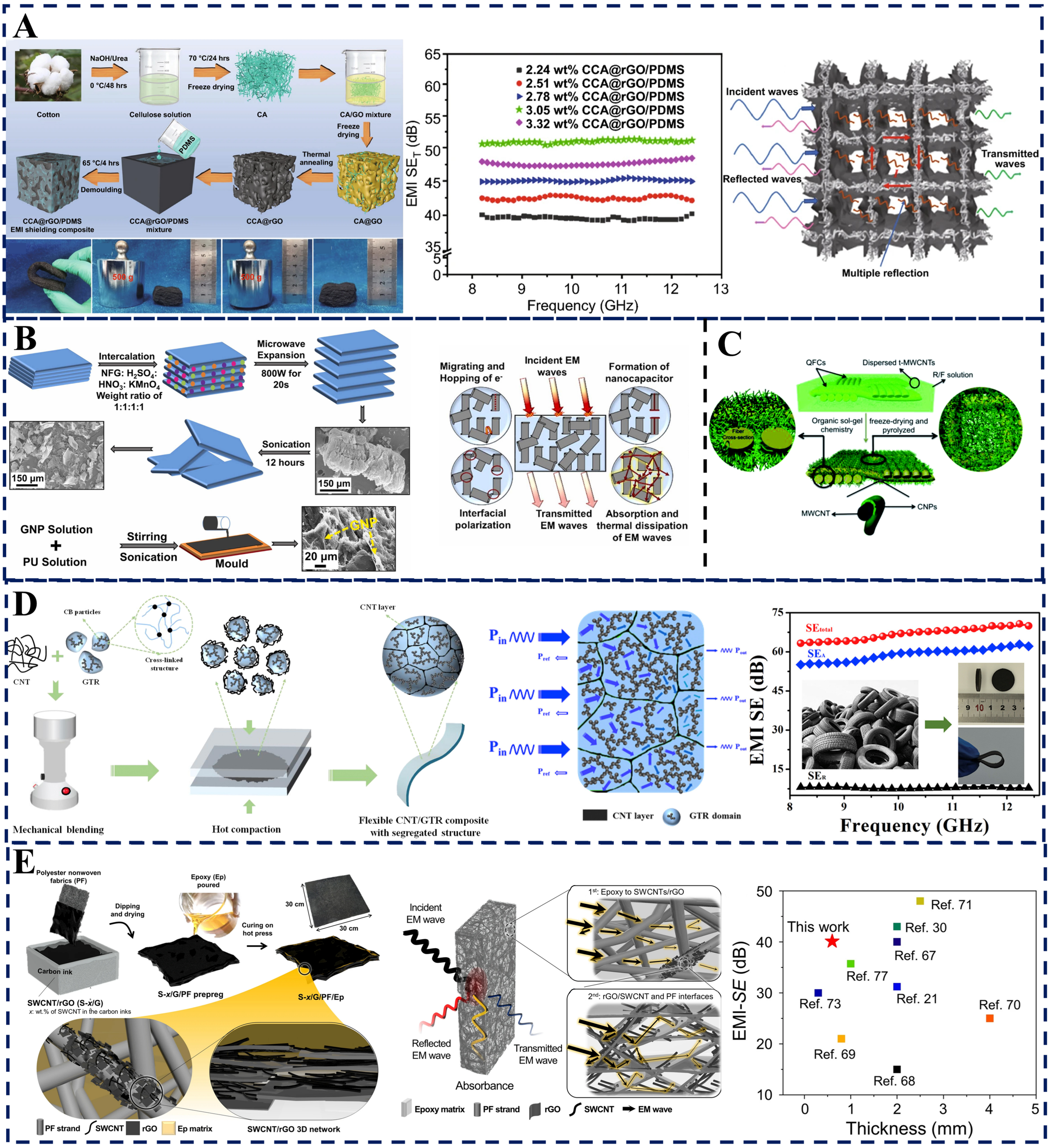


Figure 3. Flexible EMI materials based on carbon nanomaterials. (A) Preparation, EMI performance, and mechanism of CCA@rGO/PDMS. Reproduced with permission from ref[98]. Copyright 2021, Springer Nature Link; (B) Preparation of GNP/PU nanocomposites and EMI mechanism. Reproduced with permission from ref[99]. Copyright 2022, Elsevier; (C) Preparation of QMCA-PDMS composites. Reproduced with permission from ref[103]. Copyright 2014, Royal Society of Chemistry; (D) Preparation, EMI shielding performance, and mechanism of CNTs/GTR composites. Reproduced with permission from ref[104]. Copyright 2017, Elsevier; (E) Preparation, EMI performance, and mechanism of SWCNTs/rGO-loaded polyester fabrics. Reproduced with permission from ref[109]. Copyright 2022, Springer Nature Link. EMI: Electromagnetic interference; CCA: cellulose graphene carbon aerogel; rGO: reduced graphene oxide; PDMS: polydimethylsiloxane; GNP: graphene nanoparticle; PU: polyurethane; QMCA: quantum-modified carbon aerogel; CNTs: carbon nanotubes; GTR: waste tire rubber; SWCNTs: single-walled CNTs.

exhibits excellent mechanical characteristics and thermal conductivity, which are promising for portable wearable electronics applications. Tahalyani *et al.* synthesized graphene nanoparticles (GNPs) by microwave intercalation and encapsulated them in polyurethane (PU) to fabricate highly efficient and durable EMI shielding composites [Figure 3B][99]. Due to the EMW impedance mismatch caused by the high conductivity of GNPs and the multiple reflection shielding mechanism inside PU, the composite material has a shielding effect of up to 70.50 dB at a thickness of 2 mm. It is noteworthy that even after 500 cycles of mechanical bending and two hours of immersion in pure water and ultrasonic treatment, the composite material's maximum EMI shielding effectiveness (SE) essentially stays the same, indicating the material's EMI shielding stability in harsh conditions. Furthermore, the composites have superior mechanical strength (1.4 times that of PU) and tensile mechanical characteristics (> 400%), opening up new possibilities for flexible electronic gadgets of the future and aeronautical applications.

Similarly, CNTs, which possess excellent electrical properties, are commonly used to construct conductive pathways in elastomeric matrices[100-102]. Chen *et al*. used quartz fiber cloth (QFC) reinforced multi-walled CNTs (MWCNTs) to obtain carbon aerogel [QFC reinforced MWCNTs carbon aerogel (QMCA)], which was then combined with PDMS to prepare a new flexible conductive composite (FCM) [Figure 3C][103]. The high dielectric constant is caused by the polarization of the MWCNTs and the polymer matrix, which helps achieve absorption-based shielding. In addition, the large number of nanotubes will build a denser conductive network and reduce the interstitial distance of MWCNTs in the matrix, which makes the fiber network rougher and improves the EMW conduction loss, thus allowing the composite to have a high EMI shielding effect. Meanwhile, the flexible PDMS endowed the material with strong mechanical properties (tensile strength and modulus of 129.6 MPa and 3.41 GP, respectively) and compressive strength. This method maximized the advantages of both materials, significantly enhancing the mechanical and EMI shielding performance of the composite, while also broadening the potential applications of the new flexible conductive material. Even more praiseworthy is the work of Jia *et al*., who converted waste tire rubber (GTR) combined with CNTs into a high-performance EMI shielding material with high value [Figure 3D][104]. When combined with conductive CNTs, the inherent 3D crosslinking structure of GTR, which is akin to that of high-viscosity gels, prevents CNTs from penetrating the interior of GTR and creates a deviated structure that is selectively distributed at the interfaces of the structural domains of GTR. This creates a high-density conductive network that improves the electrical and EMI shielding capabilities of the polymer composites. Consequently, the composite material containing only 5.0 wt% CNTs has an EMI shielding performance of 66.90 dB, much higher than the general commercial standard (20.00 dB). Moreover, CNTs/GTR composites demonstrate excellent flexibility and stability, maintaining 93% of the EMI SE after 5,000 cycles of repeated bending. Therefore, low-cost, high flexibility, and high EMI shielding performance composite materials will shine in the next generation of flexible electronic products and wearable flexible product applications.

If the highly conductive graphene and CNTs are combined, not only can the EMI energy be dissipated through the conductive mismatch at the filler-matrix interface, but CNTs can also fill the gaps between graphene sheets and form a conductive network by bridging the neighboring graphene to intertwine with each other as a unique hierarchical structure, which can increase the propagation path of permeable EMI to promote the interfacial polarization loss to attenuate EMI waves[105-108]. Lee *et al*. prepared epoxy-based fabrics [polyester fabrics (PFs)] nanocomposites using single-walled CNTs (SWCNTs) combined with rGO as conductive fillers [Figure 3E][109]. The effective dielectric loss at multiple interfaces in the SWCNTs/rGO network, the enhanced dissipation of EMWs by the extended EM pathway, and the multi-scale porous structure consisting of 3D micropores of the SWCNTs/rGO hybrids and macropores of the PF skeleton lead to the increase in the impedance mismatch, interfacial polarization loss, and multiple scattering inside the composites, which results in a higher shielding effect in the X-band, with higher SE (41.00 dB). Additionally, PFs served as a flexible substrate, supporting the high dispersion of nanofillers in the polymer matrix, resulting in excellent mechanical durability of the composites, which maintained stable electrical conductivity and EMI shielding capability after 1,000 bending tests. These outcomes are ascribed to the hierarchical structure of the nanoscale SWCNTs/rGO network and the macroscale PF skeleton, which produce superior EMI shielding performance, which provides a valuable guideline for next-generation wearable electronic devices in terms of improving EMI SE. In summary, the properties of various Flexible EMI Composites are detailed in Table 2.

Flexible phase change composites

PCMs exhibit excellent thermal storage during phase transitions caused by variations in external temperature, which have garnered significant attention in the areas of thermal management and thermal energy storage[110]. However, most PCMs suffer from issues such as leakage and low thermal conductivity, which limit their practical applications[111,112]. To address these issues, three encapsulation techniques can be employed to effectively prevent leakage of PCMs: microencapsulation[113-116], porous adsorption[117], and polymer molding[118-120]. Additionally, if PCMs become more flexible to tolerate a certain amount of deformation and to make closer contact with the substrate surface, this will apply to a wider range of application scenarios, ultimately providing better thermal management capabilities.

Based on microencapsulation

Microencapsulation is an effective method for preventing PCM leakage by encapsulating pulverized solid or liquid particles in a solid shell, thereby isolating the PCM from the external environment. It also expands the region for heat transfer, prevents the PCM from leaking during repeated thermal cycling, and enables the support material to withstand the frequent volume changes of the core PCM during phase transitions[121]. The resulting microencapsulated PCM (micro-PCM) can achieve shape-stable structures, overcoming the limitations of bulk PCMs and making them suitable for various practical applications. Shen *et al*. prepared phase change microcapsules [microcapsule PCM (MPCM)] with high energy storage density using n-octadecane, melamine formaldehyde resin, and TiO_2 nucleating agent, which were then supplemented with polyurethane acrylate (PUA) and MXene to construct phase change composites (PCCs) with shape-stabilized excellent thermal storage and thermal management as a whole [Figure 4A][122]. The results indicated that the composite had a subcooling of 0 °C and an enthalpy of phase change of 183.7 J/g, demonstrating excellent thermal storage capability. Additionally, the thermal conductivity is increased by 48.1% compared with that of MXene-free, which is conducive to heat transfer and heat dissipation in practical applications. Furthermore, the flexibility imparted by PUA further expands the application scope of the composites, with significant potential in the thermal management of electronic materials. Not only that, the PCM encapsulation technology brought by microcapsule technology can also be used for thermal management of the human body using spinning. Ma *et al*. prepared a kind of fiber membrane based on MPCM and uniaxial electrostatic spinning [Figure 4B][123]. To stop leaks during the phase change process, n-octadecane was successfully encapsulated utilizing microencapsulation technology. The MPCM [silver nanoparticle-modified MCPM (APCM)] was then modified *in situ* using Ag nanoparticles to improve its electrical and thermal conductivity. The PCC fiber membrane was prepared via continuous uniaxial electrostatic spinning with the phase change medium as the matrix of APCM and PVDF as the matrix resin to effectively prevent its displacement or separation during use. Subsequently, the composite phase change film was obtained by modification with polydopamine (PDA) and PPy. The composite film not only has an ideal enthalpy (90.34 J/g) but also demonstrates outstanding photothermal and electro-thermal conversion capabilities as a result of PDA and PPy working in concert, allowing the body to accomplish all-season thermal comfort control in any weather conditions and at any time of day. Moreover, the composite film has excellent flexibility and mechanical strength and can be folded into a “crane” without any cracks or breaks, ensuring the comfort and long-term durability and stability of the composite phase change film as a wearable product, making it an excellent candidate for personal thermal management.

Table 2. Characteristics of different flexible EMI composites

Type of filler		Method	Properties	EMI shielding effect	Refs.
C-MXene@PI	MXene	Dip coating Chemical crosslinking	Hydrophobicity Antioxidant properties High-temperature stability	62.50 dB	[82]
MXene foams		Hydrazine-induced foaming	Water resistance Durability	70.00 dB	[83]
The sandwich-structured nanocomposite		Electrospinning Lay-up Hot-pressing techniques	Thermal conductivity High mechanical property	40.00 dB	[84]
CEF-NF/Ag/WPU	Ag	Chemical silver plating Enhanced pressing process	Thermal reliability High electrical High mechanical property	102.90 dB	[87]
AgNWs/cellulose	AgNWs	Vacuum-assisted filtration Hot pressing	High electrical High mechanical property High thermal conductivity Joule heating	101.00 dB	[89]
AN@MX/TW	MXene AgNWs	Spray layer by layer	High electrical Stability	44.00 dB	[90]
PVDF/MXene/AgNW		Solid solution casting	High mechanical property High electrical High thermal conductivity Ultrathin	41.26 dB	[91]
CCA@rGO/PDMS	GO	Freeze drying Vacuum impregnation Thermal annealing	High thermal conductivity High mechanical property Thermal stability	51.00 dB	[98]
GNP/PU	GNPs	Microwave intercalation Solution casting	High mechanical property Extreme shielding stability	70.50 dB	[99]
FCM	MWCNTs	Organic sol–gel chemistry method	High mechanical property High compressive strength	20.00 dB	[103]
CNT/GTR	CNTs	Mechanical blending Compression molding	High electrical Flexibility Stability	66.90 dB	[104]
PFs	SWCNTs GO	Vacuum impregnation Mechanical mixing	High electrical Durability High mechanical property Stability	41.00 dB	[109]

EMI: Electromagnetic interference; PI: polyimide; CEF-NF/Ag/WPU: flexible carbon fabric/Ag/waterborne polyurethane; AgNWs: silver nanowires; TW: transparent wood; PVDF: polyvinylidene difluoride; CCA: cellulose graphene carbon aerogel; rGO: reduced graphene oxide; PDMS: polydimethylsiloxane; GNP: graphene nanoparticle; PU: polyurethane; FCM: flexible conductive composite; MWCNTs: multi-walled carbon nanotubes; CNT: carbon nanotube; GTR: waste tire rubber; PFs: polyester fabrics; SWCNTs: single-walled carbon nanotubes.

Based on porous scaffolds

The selection of flexible porous scaffolds to adsorb the PCM not only successfully prevents the leakage of PCM at the phase change temperature, but also enhances the thermal conductivity and photothermal conversion performance of PCM, in addition to the flexible scaffolds can endow the composites with stronger mechanical properties to adapt to the application in extreme environments[124,125]. Liu *et al*. prepared PDMS/boron nitride (BN)/PPy porous foams with high flexibility, thermal conductivity, and light-absorbing ability by the sugar stencil method, and subsequently prepared flexible PDMS/BN/PPy/paraffin wax (PW) PCCs by the vacuum impregnation method [Figure 5A][126]. The porous structure of the PDMS/BN/PPy foam gives it a very low density and exceptional mechanical flexibility, and the BN nanosheets are on the boundaries between the neighboring sugar particles to form a 3D thermally conductive network to improve the thermal conductivity of the composites. For PCCs, the PDMS/BN/PPy foam serves as a flexible scaffold, preventing PW leakage and allowing flexible deformation. Meanwhile, the photothermal

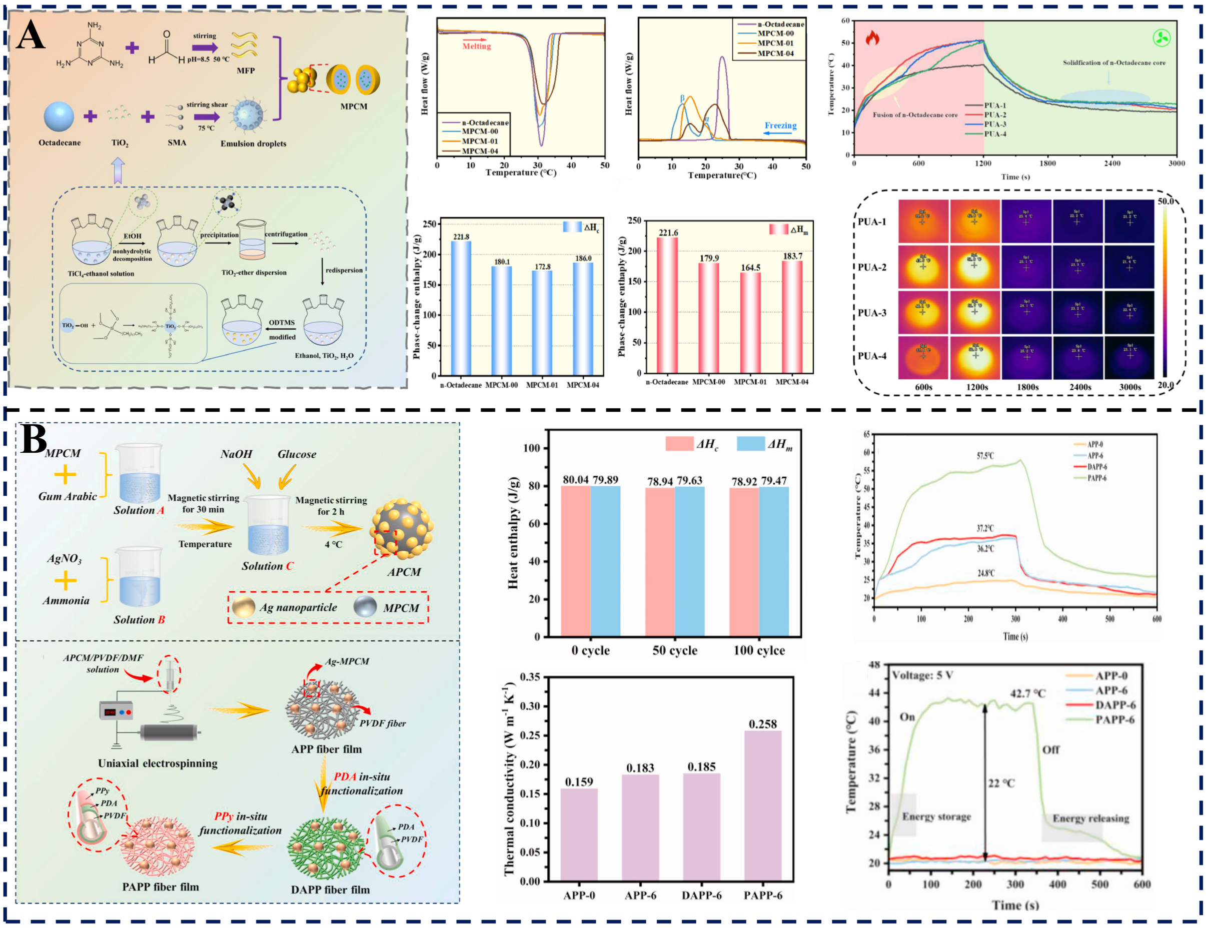


Figure 4. Flexible PCCs based on microcapsule encapsulation. (A) Preparation of MXene/MPCM/PUA composites, energy storage density, and photothermal conversion properties. Reproduced with permission from ref[122]. Copyright 2024, Elsevier; (B) Preparation of PAPP composite membranes, thermal storage capacity, and photothermal and electro-thermal conversion. Reproduced with permission from ref[123]. Copyright 2024, Elsevier. PCCs: Phase change composites; MPCM: microcapsule phase change material; PUA: polyurethane acrylate; PAPP: polypyrrole and polydopamine modified microencapsulated films.

conversion efficiency is improved by the PW's high storage energy density, pleated PPy, and continuous BN network. This work offers an effective strategy for the development of flexible PCCs with multifunctional capabilities, which have broad application potential. Designing unique network structures to adsorb PCMs is also one of the strategies for obtaining flexible PCCs, Cheng *et al*. prepared a flexible support material [CS/PVA/CNTs (CPC)] with a folded layer-bridge network structure by dispersing CNTs in an acetic acid solution of chitosan (CS) and PVA using a directional freezing method, and then vacuum impregnated polyethylene glycol (PEG) to obtain a CPC@PEG PCC [Figure 5B][127]. The material's exceptional mechanical qualities and flexibility include a high tensile strength of 2.42 MPa and flexural resistance surpassing 100 cycles. In addition, it demonstrates excellent latent heat storage (enthalpy up to 152.7 J/g) because of the many macropores in the multistage pores and the hydrogen bonding contacts between the PEG molecules and the CPC scaffolds. With its exceptional mechanical qualities and high thermal storage capacity, the composite PCM holds promise for flexible temperature control materials that can be worn on the skin in the future. Flexible PCCs not only play a role in solar energy harvesting, electronics thermal management, and human body thermal management but also have unique applications in the face of medical thermotherapy, Chen *et al*. proposed a simple and low-cost organic solvent-free route to construct 3D freestanding flexible cellulose nanofibers (CNF) sponges with layered interconnections, followed by

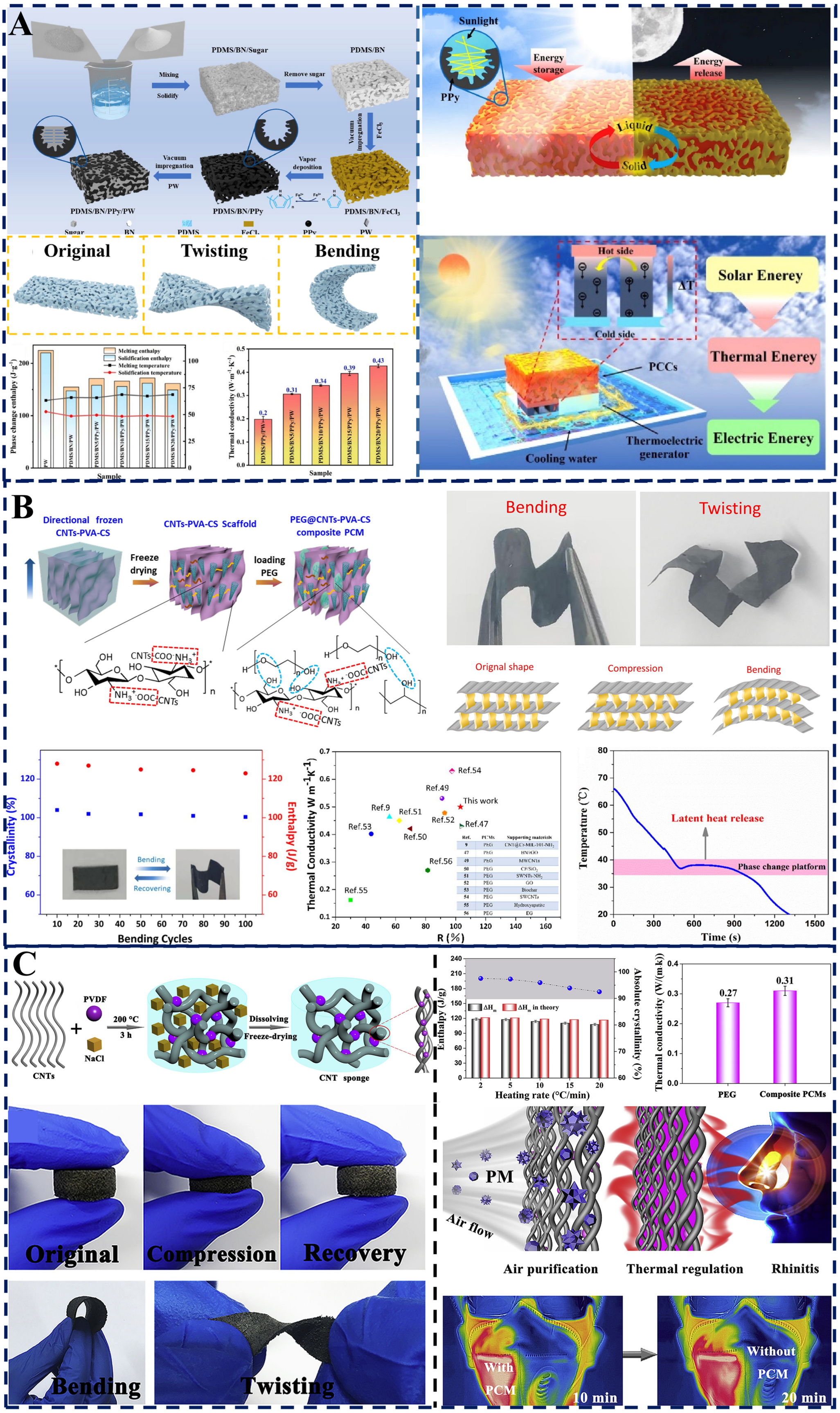

A
PDMS/BN/Sugar
PDMS/BN
Mixing
Solidify
Remove sugar
Vacuum impregnation
PW
Vapor deposition
PDMS/BN/PPy/PW
PDMS/BN/PPy
PDMS/BN/FeCl3
Sunlight
PPy
Energy storage
Energy release
Liquid
Solid
Original
Twisting
Bending
Hot side
Cold side
Solar Enerey
Thermal Enerey
Electric Enerey
PCCs
Cooling water
Thermoelectric generator
B
Directional frozen CNTs-PVA-CS
CNTs-PVA-CS Scaffold
PEG@CNTs-PVA-CS composite PCM
Freeze drying
loading PEG
Bending
Twisting
Orignal shape
Compression
Bending
Bending Cycles
Crystallinity (%)
Enthalpy (J/g)
Recovering
This work
Thermal Conductivity W m-1K-1
R (%)
Latent heat release
Phase change platform
Temperature (℃)
Time (s)
C
PVDF
NaCl
CNTs
200 °C 3 h
Dissolving Freeze-drying
CNT sponge
Heating rate (°C/min)
Absolute crystallinity (%)
Thermal conductivity (W/(m k))
PEG
Composite PCMs
Original
Compression
Recovery
PM
Air flow
Air purification
Thermal regulation
Rhinitis
Bending
Twisting
With PCM
10 min
Without PCM
20 min

Figure 5. Flexible PCCs based on porous scaffolds. (A) Preparation, mechanical properties, thermal storage capacity, photothermal conversion, and photothermoelectric conversion properties of PDMS/BN/PPy/PW PCCs. Reproduced with permission from ref[126]. Copyright 2024, Elsevier; (B) Preparation, mechanical properties, energy storage density, thermal conductivity, and photothermal conversion of CPC@PEG PCCs. Reproduced with permission from ref[127]. Copyright 2020, Elsevier; (C) Preparation of composite PCMs, thermal storage capacity, mechanical properties, and thermal therapy mechanisms. Reproduced with permission from ref[128]. Copyright 2020, Elsevier. PCCs: Phase change composites; PDMS: polydimethylsiloxane; BN: boron nitride; PPy: polypyrrole; PW: paraffin wax; CPC: CS/PVA/CNTs; PEG: polyethylene glycol; PCMs: phase change materials.

impregnation of PEG into the sponges to prepare PCCs with flexible layered frameworks [Figure 5C][128]. The developed CNT sponge-based composite PCM exhibited high thermal energy storage capacity and excellent thermal stability even after 100 melt-freeze cycles, and the functional mask designed with it comprises an inner thermal conditioning layer (CNT sponge-based PCM layer) and an outer air-purifying layer (pristine CNT sponge layer) for thermal therapy of allergic rhinitis. By continually releasing heat, the inner CNT sponge-based PCMs dominate thermal therapy, whereas the outer air purification layer only serves as a supplementary function by trapping particulates and purifying the air that is inhaled. The cleansed air amplifies the thermotherapy effect, and the original layered CNT sponge layer works well as a PM trap. The PEG-based mask can supply sufficient thermal energy to warm the airflow into the nasal cavity and sustain the temperature plateau at 43.5 °C for 33 min, according to the heat release performance. The associated medical results also demonstrate that the thermotherapy mask can greatly reduce the nasal mucosa's inflammatory damage, which certainly broadens the way for flexible PCCs in the medical field.

Based on polymer encapsulation

The use of polymer encapsulation is also a major means of solving the solid-liquid PCM leakage problem by dispersing the micromolecular PCM into a macromolecular polymer matrix to form a shape-stable mixture, with the crosslinked polymer matrix acting as a support material to prevent leakage of the molten PCM[129,130]. Commonly used polymers include polyethylene, PU, block copolymers olefin block copolymers (OBC), *etc.*[131]; the polymer matrix also helps improve the flexibility of the composite material, supplemented with various thermally conductive fillers to prepare phase change thermal management materials with ultra flexibility has broad application prospects. Wu *et al*. used a dual network of polymers and graphite nanosheets as a functional matrix for PCMs to prepare composite PCMs with high thermal conductivity, flexibility, and shape stabilization, in which PW was used as the PCM, and the macromolecular olefinic OBC formed a crosslinked polymer network to encapsulate the molten PCM and give the composite film was flexible, and expanded graphite (EG) with a long chain structure and high thermal conductivity formed a permeable network of aligned interconnected graphite nanosheets to confer high thermal conductivity properties to the PCC [Figure 6A][132]. The prepared flexible phase change film exhibits high energy storage density (169.5 J/g) and excellent thermal conductivity (radial thermal conductivity of up to 4.2-32.80 $W \cdot m^{-1} \cdot K^{-1}$), and also good flexibility and mechanical strength under bending and stretching, which expands its scope of application. Furthermore, the flexible film was used for thermal management of lithium batteries, reducing their operating temperature by more than 12 °C and demonstrating efficient and reliable performance. Also using a combination of polymers and conductive nanosheets to immobilize PCMs, Wu *et al.* synthesized hybrid semi-interpenetrating composites (PEG@PU) by infiltrating PEG into the crosslinking network of PU; they then assembled them by pressure-induced assembly with reticulated graphite nanoparticles (RGNP) doping to prepare a highly conductive and thermally flexible PCCs (PEG@PU-RGNP) [Figure 6B][133]. The hybrid network allowed the PCCs to demonstrate excellent properties, including superior mechanical strength, shape stability, and thermal storage. In addition, the PCCs exhibit a thermal conductivity of up to 27.00 $W \cdot m^{-1} \cdot K^{-1}$ and an electrical conductivity of 51.0 S/cm, which is superior to that of the latest PEG-based PCCs, and the flexible PCC can be applied for efficient battery thermal management to meet the main requirements for cold environment

Figure 6. Flexible PCCs based on polymer encapsulation. (A) Preparation, thermal storage capacity, thermal conductivity, mechanical properties, and battery thermal management applications of PW-OBC/EG composites. Reproduced with permission from ref[132]. Copyright 2020, Royal Society of Chemistry; (B) Preparation of PCCs flexible PCCs, thermal storage capacity, thermal conductivity, electrical conductivity, and cooling cell schematics. Reproduced with permission from ref[133]. Copyright 2021, Wiley-VCH; (C) Preparation of TIPCM composites, self-healing and shape memory capabilities, and schematic diagram of chip thermal management capabilities. Reproduced with permission from ref[134]. Copyright 2024, Elsevier. PCCs: Phase change composites; PW: paraffin wax; OBC: olefin block copolymer; EG: expanded graphite; TIPCM: thermal interfacial phase change material.

preheating in cold environments and passive cooling in hot environments. Polymer encapsulation technology can not only meet the basic shape stabilization and thermal management but also realize PCCs with specific functions by designing structures. Huang *et al*., by introducing a boron ester crosslinking skeleton into PW and ethylene-octene copolymer (EOC), designed a thermal interfacial PCM (TIPCM) [Figure 6C] to realize a 3D support skeleton with a dynamic crosslinking network thereby maintaining the mechanical stability of the TIPCM[134]. The TIPCM exhibits a high energy storage density (178.4 J/g) and an excellent thermal conductivity (3.23 $W \cdot m^{-1} \cdot K^{-1}$). Notably, TIPCM surface is rich in dynamic and non-conjugated boron ester bonds that can be thermally induced to rearrange networks, reconstruct the topology of boron ester bonding, and exhibit self-healing properties, meaning that when subjected to mechanical damage brought about by extreme environments, the unique self-healing ability of TIPCM can ensure its normal use and prolong its service life, and if it is used in the thermal management of the chip, in actual tests, TIPCM was found to reduce the operating temperature of a CPU by 67 °C at 5 V, demonstrating its potential for efficient thermal management, which is an important advantage for the design of PCMs with excellent thermal storage and special physical properties. In summary, polymer encapsulation technology combined with various conductive and thermally conductive fillers offers a promising path for the preparation of highly conductive, thermally conductive, and shape-stable PCCs for waste heat harvesting and thermal management of electronic products. Summary of properties of different flexible PCCs is detailed in Table 3.

Flexible EMI PCCs

Researchers have recently investigated polymer-based flexible EMI shielding composites based on fillers such as MXene, carbon materials, and metal nanoparticles. These composites effectively address the drawbacks of metal materials in real-world applications, such as their high density, poor weather resistance, and susceptibility to corrosion[135]. However, a single EMI composite material can no longer satisfy the use in many fields, particularly as 5G or even higher communication technology advances; the powerful EMWs that are converged by electronic micro-devices will cause heat to accumulate. This will not only lower the equipment's operational efficiency but also significantly shorten its service life and possibly endanger people's health[136-139]. The design of flexible PCCs with both thermal management and electromagnetic shielding is a matter of urgency.

By incorporating MXene nanomaterials into polymer matrices to form a conductive network, it is possible to create lightweight, high-performance EMI shielding materials. When PCMs are adsorbed onto this network, flexible EMI PCCs with excellent thermal management properties are formed, offering significant potential for various applications. Li *et al.* obtained a new type of Janus multifunctional film by encapsulating PCMs in an ultra-flexible EMI film with continuous electrostatic spinning and spraying technology[140]. The phase change microcapsules are connected with PVA to form a stable "candied fruit structure", and a layer of polylactic acid (PLA) is superimposed on its surface, and finally an ultra-flexible EMI phase change film is obtained by spraying a conductive layer of MXene on the PLA surface [Figure 7A]. A large number of spherical PCCs and flexible PVA fibers are integrated to impart excellent bendability to the film. The strong conductivity of the MXene layer is the main cause of the EMI shielding effect. When EMW strikes the MXene sheet, a large number of free electrons are attenuated by impedance matching, which absorbs the energy of the EMW incident on the film surface. This transforms the EMW energy into thermal energy, which is then released through hysteretic losses. Meanwhile, due to the foldability of the film, the remaining EMW, after passing through the first MXene layer, encounters a barrier in the next layer; i.e., the second layer acts as an emissive layer generating multiple internal reflections leading to a continuous attenuation of the EMW until it is completely absorbed by the structure, which can be attributed to the reflection coefficient of the folded film being significantly higher than that of a single film, and thus, through the ingenious origami process, the EMI shielding effect of the composite film has been improved. Moreover, the incorporation of PCCs with heat storage capacity imparts the film with excellent phase change temperature regulation and radiation cooling, contributing to effective human

Table 3. Summary of properties of different flexible PCCs

Encapsulation technology		PCM	Properties	Enthalpy	Potential direction	Refs.
MPCM	Microencapsulation	n-Octadecane	Excellent thermal storage High thermal conductivity	183.70 J/g	Solar heating	[122]
PAPP			Photo-thermal effect Electro-thermal response High thermal conductivity High electrical High mechanical property	90.34 J/g	Personal thermal management	[123]
PDMS/BN/PPy/PW	Porous adsorption	PW	Mechanically flexible Excellent thermal storage Photo-thermal effect	172.00 J/g	Thermoelectric conversion	[126]
CPC@PEG		PEG	Excellent thermal storage Mechanically flexible	152.70 J/g	Wearable skin-friendly temperature-controlled	[127]
CPCM		PEG	Excellent thermal storage High mechanical property Thermal stability	120.00 J/g	Hyperthermia	[128]
PCC	Polymer molding	PW	Excellent thermal storage High thermal conductivity High mechanical property Flexibility	169.50 J/g	Lithium battery thermal management	[132]
PEG@PU-RGNP		PEG	High thermal conductivity High electrical High mechanical property Excellent thermal storage	163.50 J/g	Battery thermal management	[133]
TIPCM		PW	High mechanical property Excellent thermal storage Shape memory ability Self-healing ability	178.40 J/g	Chip thermal management	[134]

PCCs: Phase change composites; PCM: phase change material; MPCM: microcapsule phase change material; PAPP: polypyrrole and polydopamine modified microencapsulated films; PDMS: polydimethylsiloxane; BN: boron nitride; PPy: polypyrrole; PW: paraffin wax; CPC: CS/PVA/CNTs; PEG: polyethylene glycol; CPCM: composite phase change material; PU: polyurethane; RGNP: reticulated graphite nanoparticles; TIPCM: thermal interfacial phase change material.

body heat management. The high mid-infrared emissivity of the composite film (90.15%) allows it to drastically lower the surface temperature (10.13 °C) in hot conditions, while only 1.5 V is needed to keep the human body at a long-term thermal comfort temperature at night or in cold conditions. Combined with the photothermal properties, excellent thermal storage (141.4 J/g), and thermoregulation, the human body can achieve thermal comfort in all seasons, day or night, cold or hot. This unique Janus-type structural composite film not only brings convenience in the field of EMI but also combines excellent thermal management capabilities, providing a new strategy for manufacturing wearable products for the human body. Inspired by the "brick-mortar" sandwich structure, Gong *et al.* used n-octadecane phase change capsules as rigid "bricks", MXene as "sand", and PVA as the "cement", and constructed flexible PCCs/MXene/PVA (PMP) sandwich with a "brick-wall" structure by vacuum filtration[141]. MXene/PVA

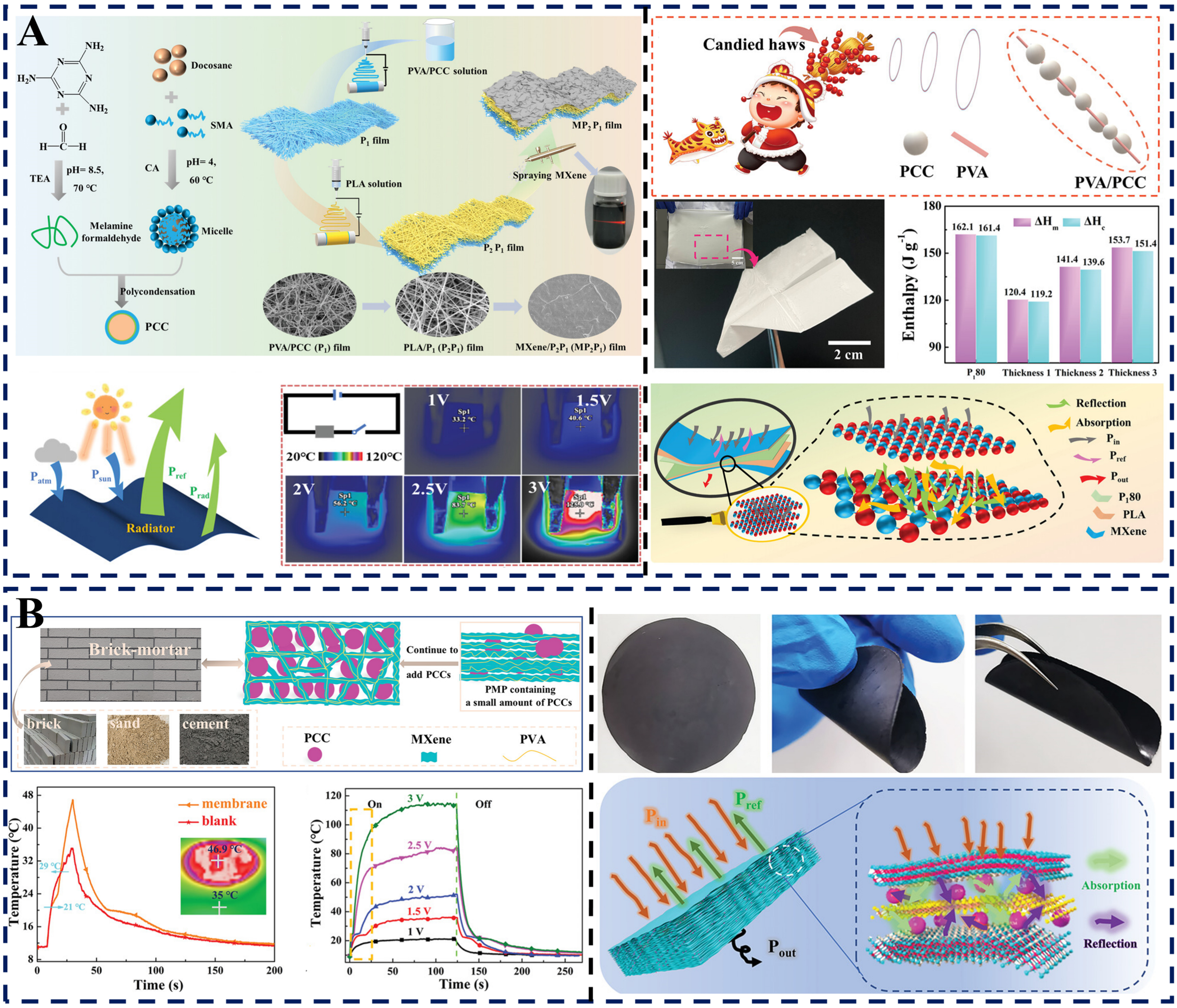


Figure 7. Loaded MXene builds flexible EMI PCCs. (A) Preparation of composite materials, flexible digital images, thermal storage capacity, photothermal and electro-thermal conversion, and electromagnetic shielding mechanisms. Reproduced with permission from ref[140]. Copyright 2023, Wiley-VCH; (B) Preparation of PMP multifunctional composite films, demonstration of flexibility, demonstration of photothermal and electro-thermal conversion capabilities, and EMI shielding mechanisms. Reproduced with permission from ref[141]. Copyright 2022, Wiley-VCH. EMI: Electromagnetic interference; PCCs: phase change composites; PMP: PCCs/MXene/PVA.

multifunctional composite film with a "brick-wall" structure was constructed by vacuum filtration technology [Figure 7B]. Due to the close bonding of hard and soft layers, the films exhibit excellent mechanical strength, and the spacing between the layers is significantly reduced with the addition of PVA, and the flexibility and strength of the PMP films are also increased. Furthermore, PMP offers excellent EMI shielding due to its excellent conductivity and unique "brick-mortar" layered structure. This can be attributed to two aspects: on the one hand, the MXene dipole interface polarization and impedance mismatch loss; on the other hand, the interaction of EMW with the high electron density in the MXene lattice, which results in ohmic loss and internal reflection and depletion until it is completely absorbed by the structure. Moreover, the film also exhibits superior thermal management performance due to the incorporation of PCCs. With its higher energy storage density (136.5 J/g) and thermal conductivity (0.69 $W \cdot m^{-1} \cdot K^{-1}$), PMP can continuously supply heat to the body through passive radiation, absorb solar energy, and convert it into thermal energy. Additionally, the excellent electrical conductivity (5,000 S/m) conferred by the Joule heat enables PMP to achieve thermal management even at very low voltages at night, greatly broadening its potential applications. The methodology presented in this work provides a promising new approach for the development and preparation of integrated multifunctional PMP films that can be used in the field of flexible wearable electronics with effective thermal management and EMI shielding.

Carbon nanomaterials, such as CNTs, graphene, *etc.*, can also be combined with polymer matrix and PCMs to achieve dual-functionality in both EMI shielding and thermal management. Yang *et al*. fabricated flexible, thin, and robust phase-change nonwoven fabric (PCNF) in a scalable manner using SWCNTs-embedded PEG-grafted TPU (TPEG) prepolymers as phase change filaments (PCWs) by a simple, accurate, and controllable 3D printing strategy [Figure 8A][142]. Because of their exceptional mechanical resilience and remarkable flexibility, PCNF films can tolerate a wide range of extreme deformations, including folding, coiling, and bending. They can even be readily kneaded into flat surfaces without suffering any structural fracture. This makes them ideal for use as wearable fabrics. Additionally, SWCNTs uniformly embedded within the PCNF network absorb and reflect EMWs multiple times, resulting in excellent electromagnetic shielding performance. Moreover, the film exhibits excellent thermal properties, and the test of placing it into the jacket liner reveals that the temperature at this location is significantly lower compared with other parts, showing the superior thermal regulation performance of the PCNF. Also using SWCNTs as a conductive filler to prepare flexible EMI PCMs is Ge *et al*, who proposed a phase change composite SWCNT/PEG/PDMS (SPP) consisting of two fillers, SWCNTs and PEG, in a PDMS matrix [Figure 8B][143]. Among the two fillers, SWCNTs contribute to the dielectric properties necessary for microwave attenuation. Compared to MWCNTs, SWCNTs have a smaller diameter and larger aspect ratio, resulting in a lower percolation threshold. This minimizes the filler loading, thereby leaving more space for the PCM to enhance energy storage density. The results show that the crosslinked network structure of the PDMS matrix plays a role in the excellent mechanical properties of the composites, making it easy to bend and twist to maintain its flexibility. Additionally, the fast chain dynamics and crosslinking properties enable the PDMS molecules to move and recover rapidly. Furthermore, the EMI shielding capability of the SPP is synergistically enhanced by conduction losses in the SWCNT network, interfacial polarization between the SWCNTs and the polymer matrix, and dipolar polarization of the polymer. Since SPP has good phase transition properties and high latent heat, it can be used as a heat sink or buffer layer to stabilize the temperature fluctuation of electronic devices, so the material may become a multifunctional material for EMI shielding and thermal management, especially in the miniaturization of electronic devices. In addition, Li *et al*. prepared PP/CNTs/Fe_3O_4/PW shape-stabilized PCCs (SSPCCs) by scalable melt mixing, salt templating, and vacuum impregnation methods [Figure 8C][144]. The integration of CNTs, which form an enhanced thermal conduction path, with the regular porous structure of the SSPCCs led to remarkable thermal conductivity (0.59 $W \cdot m^{-1} \cdot K^{-1}$), good thermal stability, and high enthalpy (133.8 J/g), enabling a reduction of the smartphone's operating temperature by 10 °C due to its superior temperature control effect. Furthermore, due to the increase in the Fe_3O_4 content, the EMI shielding effect of the SSPCCs was gradually increased with the SE_T as high as 41.20 dB, which was attributed to the high electrical conductivity and Fe_3O_4 magnetism of SSPCCs. Meanwhile, the significant mechanical strength and flexibility offer the possibility of multifunctional applications for flexible electronic devices.

Because of its high electrical conductivity and huge surface area, graphene filler has been shown to improve the thermal conductivity of composites while also being a perfect material for creating high-performance EMI shielding materials. Its excellent properties have led many researchers to utilize it in constructing a diverse range of composites with varied functionalities[96,97]. Liang *et al*. added polystyrene microspheres as hard stencils into GO, and then impregnated PW to obtain multifunctional integrated films [Figure 9A][145]. The addition of PS microspheres caused a large number of bubble-like micropores to develop inside the graphene-based structure. These bubble-like micropores increase the capillary force between PW and the

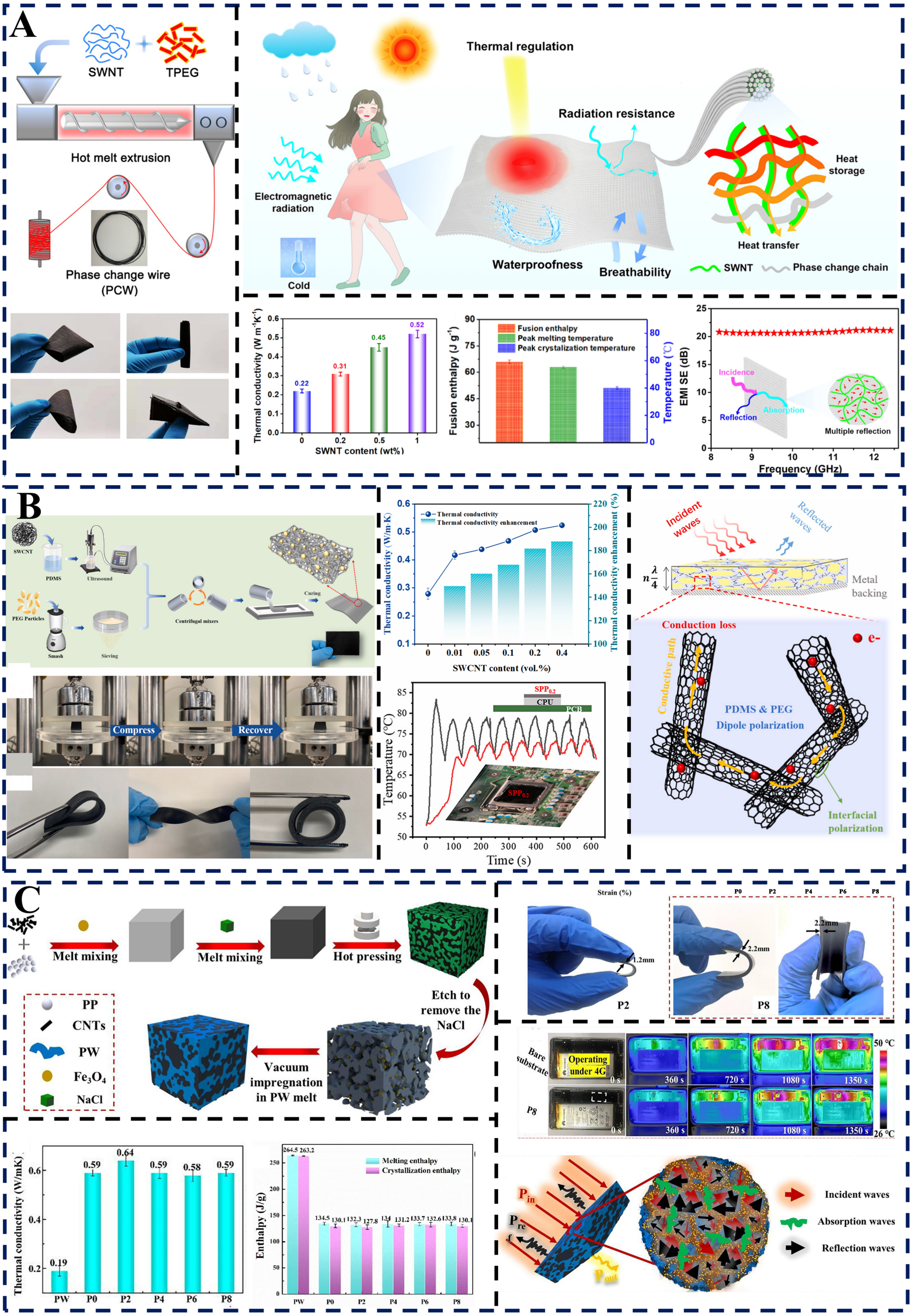

A
SWNT
TPEG
Hot melt extrusion
Phase change wire (PCW)
Thermal regulation
Radiation resistance
Heat storage
Electromagnetic radiation
Heat transfer
Waterproofness
Breathability
SWNT
Phase change chain
Cold
Thermal conductivity (W m^{-1}K^{-1})
SWNT content (wt%)
Fusion enthalpy
Peak melting temperature
Peak crystalization temperature
Fusion enthalpy (J g^{-1})
Temperature (℃)
EMI SE (dB)
Incidence
Reflection
Absorption
Multiple reflection
Frequency (GHz)
B
SWCNT
PDMS
Ultrasound
PEG Particles
Centrifugal mixers
Curing
Smash
Sieving
Compress
Recover
Thermal conductivity
Thermal conductivity enhancement
Thermal conductivity (W/m·K)
Thermal conductivity enhancement (%)
SWCNT content (vol.%)
SPP$_{0.2}$
CPU
PCB
Temperature (℃)
Time (s)
Incident waves
Reflected waves
Metal backing
Conduction loss
e-
Conductive path
PDMS & PEG
Dipole polarization
Interfacial polarization
C
Melt mixing
Melt mixing
Hot pressing
PP
CNTs
PW
Fe$_3$O$_4$
NaCl
Etch to remove the NaCl
Vacuum impregnation in PW melt
Strain (%)
P0 P2 P4 P6 P8
1.2mm
2.2mm
2.2mm
P2
P8
Bare substrate
Operating under 4G
P8
0 s 360 s 720 s 1080 s 1350 s
50 ℃
26 ℃
Thermal conductivity (W/mK)
Melting enthalpy
Crystallization enthalpy
Enthalpy (J/g)
P$_{in}$
P$_{ref}$
P$_{out}$
Incident waves
Absorption waves
Reflection waves

Figure 8. Construction of lightweight EMI PCCs based on carbon material realization. (A) Preparation of PCNF films, multifunctional integration schematic, thermal storage capacity, and EMI shielding mechanism. Reproduced with permission from ref[142]. Copyright 2022, American Chemical Society; (B) Preparation of SPP composites, digital images of mechanical properties, thermal conductivity, chip thermal management performance, and EMI shielding schematics. Reproduced with permission from ref[143]. Copyright 2023, Elsevier; (C) Preparation of PP/CNTs/Fe_3O_4/PW composites, material flexibility display schematic, thermal storage capacity and thermal conductivity, cell phone thermal management performance display, and EMI shielding mechanism. Reproduced with permission from ref[144]. Copyright 2022, Elsevier. EMI: Electromagnetic interference; PCCs: phase change composites; PCNF: phase-change nonwoven fabric; SPP: SWCNT/PEG/PDMS; PP: polypropylene; CNTs: carbon nanotubes; PW: paraffin wax.

graphene-based framework in addition to absorbing and holding significant amounts of PW, thereby ensuring excellent shape stabilization of the prepared composite phase change material (CPCM). Additionally, the synergistic effect of the high porosity and high electrical conductivity of the graphene-based framework ensures that when incident EMWs enter the prepared CPCM, the high conductivity of the graphene skeleton and numerous graphene interfaces cause repeated reflections within the micropores, leading to significant attenuation of the EMWs. Meanwhile, the thermal conductivity path formed by the porous graphene-based framework leads to a dramatic increase in thermal conductivity with the increasing density of graphene. This, combined with the high energy storage density (189.10 J/g) provided by PW, was used to evaluate the temperature-regulating capability of the thermal management components for electronic cooling, and it was found that the temperature of the electronic products simulated to be almost unchanged due to the latent heat of the phase change storage during the simulated operation, and the maximum temperature is 8.9 °C lower than that of the electronic products without CPCM. This indicates that it is a strong candidate for thermal management and efficient EMI shielding in electronic devices due to its exceptional temperature control capability, quick thermal response, and great EMI shielding performance. The combination of graphene and MXene to prepare thermal management materials with EMI shielding performance may result in a synergistic effect, where the combined performance exceeds the sum of their contributions. Hu *et al*. designed a dual-encapsulated multifunctional composite material by constructing a graphene aerogel in a vertical MXene, and then thermal annealing treatment to construct a highly hybridized semi-interpenetrating framework MXene-graphene foam (MGGF) with thermal/electrical behavior [Figure 9B][146]. The framework composed of graphene and MXene demonstrates superior EMI shielding performance due to its higher conductivity and more densely packed continuous network compared to a single graphene framework. On the one hand, due to the large impedance mismatch triggered by free electrons, the incident EMW interacts with the high density of electrons in the continuous MGGF framework, which increases the conduction ohmic loss and leads to energy dissipation of electromagnetic radiation. On the other hand, the hierarchical porous MGGF framework provides a large number of interfaces and channels for multiple interlayer reflections of EMWs, increasing wave interactions at multi-conducting interfaces and their propagation routes. This helps dissipate most of the residual electromagnetic radiation and leads to additional attenuation through absorption, ultimately allowing only a small fraction of EMWs to be transmitted. The dual-encapsulated composite also exhibits high thermal conductivity (11.39 $W \cdot m^{-1} \cdot K^{-1}$) and an optimal energy storage density (160.3 J/g), which is used to cool high-power semiconductor devices. After 960 s of chip operation, the heatsink with MGGF not only reaches a stable state more rapidly but also exhibits the lowest saturation temperature, allowing the chip to operate at a lower temperature (6.8 °C) and extending its service life. This demonstrates the composite material's exceptional ability to reduce temperatures and achieve efficient thermal management. This dual-encapsulation approach has far-reaching significance and guiding strategies for designing multifunctional composites with excellent EMI shielding performance and ultra-efficient thermal management, and for broadening their use in high-power electronics, artificial intelligence, and other fields.

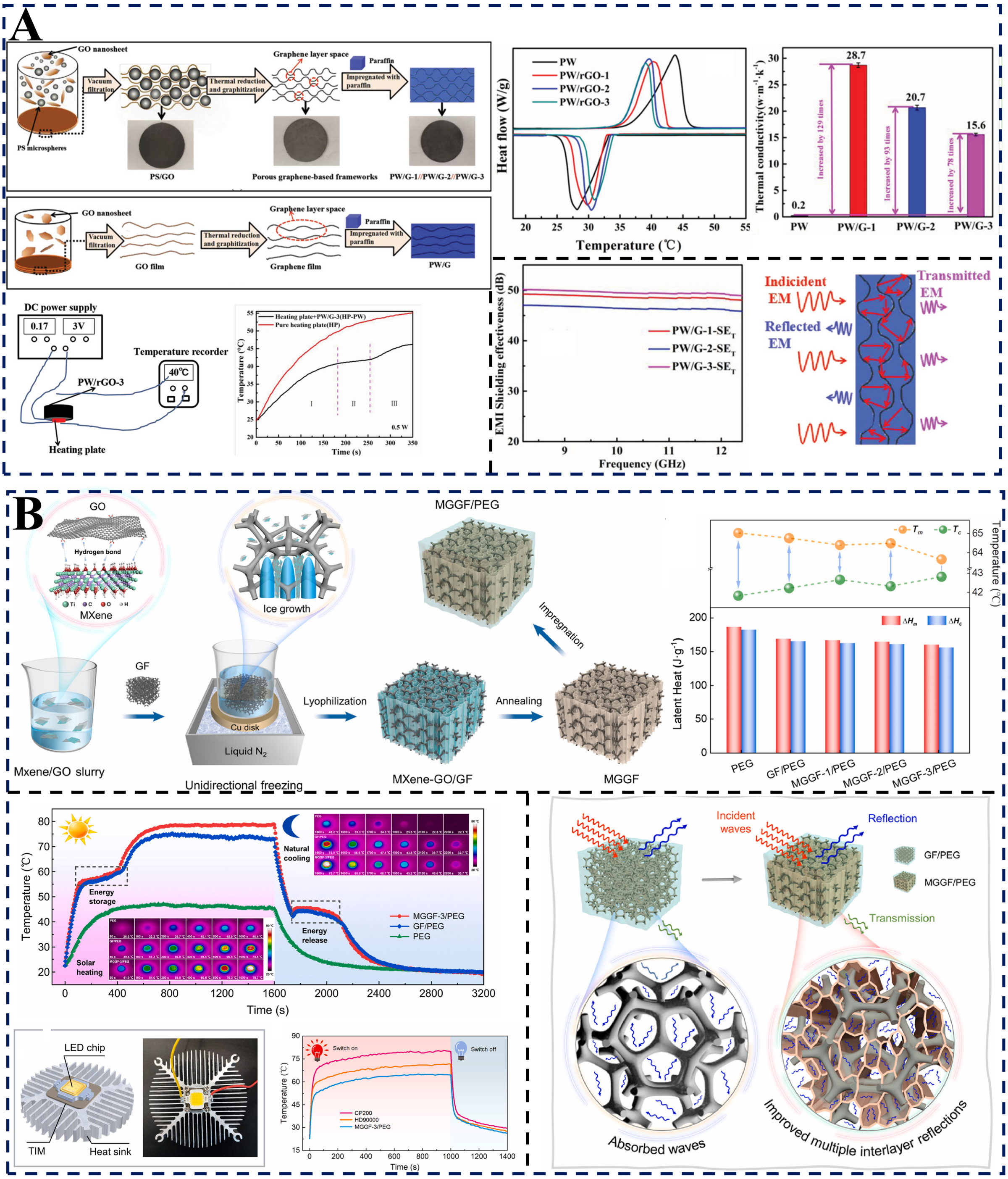


Figure 9. Flexible EMI PCCs constructed based on graphene. (A) Preparation of CPCM composites, thermal storage capacity and thermal conductivity, schematic diagram of thermal management of electronic cooling and EMI shielding mechanism. Reproduced with permission from ref[145]. Copyright 2021, Wiley-VCH; (B) Preparation of MGGF composites, thermal storage capacity, schematic diagram of photothermal conversion, schematic diagram of chip thermal management, and EMI shielding mechanism. Reproduced with permission from ref[146]. Copyright 2023, Elsevier. EMI: Electromagnetic interference; PCCs: phase change composites; CPCM: composite phase change material; MGGF: MXene-graphene foam.

Adding a small number of metal nanoparticles can overcome the drawbacks of traditional metals, such as susceptibility to corrosion, high density, and difficulty in processing while imparting high electrical and thermal conductivity to the composites, combined with the excellent thermal storage of the PCM, accompanied by the excellent EMI shielding performance and excellent thermal management capabilities, which have been widely used by researchers in recent years for the preparation of ultra-flexible EMI PCCs. Guo *et al.* plated silver *in situ* on hard ferromagnetic NdFeB to improve its thermal and electrical conductivity, then used styrene-ethylene/propylene-styrene block copolymer (SEPS) to make a flexible skeleton and mixed with paraffin wax (PA) to obtain PA/SEPS (PSP), and finally, the NdFeB@Ag magnets were immediately mixed with the PSP melt, and then underwent a magnetic induction process to construct a directional interconnection network, resulting in the fabrication of an adjustable flexible and magnetically fastened PCM (FMP) [Figure 10A][147]. The material benefits from the typical block structure and inherent flexibility of SEPS, along with the ability of NdFeB@Ag to dissipate stresses and reduce stress concentrations, which increases the tensile strength and ensures the flexibility of the FMP. Consequently, the FMP can withstand significant mechanical impacts, such as bending, torsion, and rolling, without cracking. FMP also exhibits excellent EMI shielding performance, which can be attributed to the fact that most of the impedance mismatch occurs when EMWs irradiate the EMP surface and are directly reflected, and the interaction of the incoming EMWs with the high-density charge carriers accelerates the interfacial polarization and conduction loss of EMW. In addition, the orientation and porous network offer numerous stacking interfaces for repeated reflections and absorptions, and this structure significantly lengthens the propagation path of the EMWs, which is advantageous for significant energy attenuation of the surviving EMWs through hysteresis loss. Meanwhile, FMP has high thermal conductivity (2.59 $W \cdot m^{-1} \cdot K^{-1}$) and energy storage density of up to 120 J/g. FMP can be used for Joule-heat-driven active thermal management and solar-driven energy storage conversion, and this work provides a new strategy for the design of metal particles for EMI and heat transfer, which makes the composite materials useful for high-power semiconductor devices and energy utilization systems with promising applications. Inspired by mussels, Xiao *et al*. used melamine foam (MF) with a 3D porous structure as a template, constructed a conductive network by chemical silver plating, introduced strong adhesive PDA, and subsequently vacuum impregnated PEG to obtain flexible composites (MPA_{χ}-PEG) [Figure 10B][148]. Based on the excellent deformation and recovery ability brought by MF, the composites still have good deformation ability even after being encapsulated by AgNPs, and can be easily bent, twisted, and stretched. The successful construction of the continuous 3D conductive Ag network gives the MPA_{χ}-PEG PCM the required electrical conductivity. A portion of the EMW is instantly reflected when it hits the material's surface because of the impedance mismatch caused by the high electrical conductivity, and the remaining wave enters the interior to be rapidly captured and dissipated by the high electron density AgNPs through ohmic losses, while the internal multiple reflections eventually dissipate it. Overall, the excellent EMI shielding performance (SE_T up to 82.02 dB) exhibited by the composites is a result of the combination of high conductivity and the porous structure that brings about reflection, absorption, and multiple reflection shielding mechanisms. In addition, MPA_{χ}-PEG also has a high energy storage density (148.9 J/g) and thermal conductivity, which is used for smartphone thermal management. The composite material's flexibility makes it simple to cover the smartphone CPU's surface, which helps accelerate the heat transfer by lowering the thermal resistance, and the operating temperature is significantly slowed down when the CPU is wrapped by MPA_{χ}-PEG, and when the CPU is wrapped with MPA_{χ}-PEG, the operating temperature slows down significantly, and its maximum temperature is much lower than that of the pure CPU, which means that the PCM in the composite material can absorb a large amount of heat generated by the CPU and has excellent thermal management capability. A typical example of also using AgNWs as a conductive filler to provide EWI functionally integrated PCMs is presented by Liang *et al*., who proposed wrapping polyethylene terephthalate (PET) textiles with AgNWs and then encapsulating them in PEG with 3-isocyanatopropyltriethoxysilane (IPTS) crosslinked phase change coating to obtain a composite textile material with flexible thermally responsive EMI shielding performance and excellent heat dissipation function [Figure 10C][149]. The homogeneous distribution of AgNWs along the textile's in-plane direction

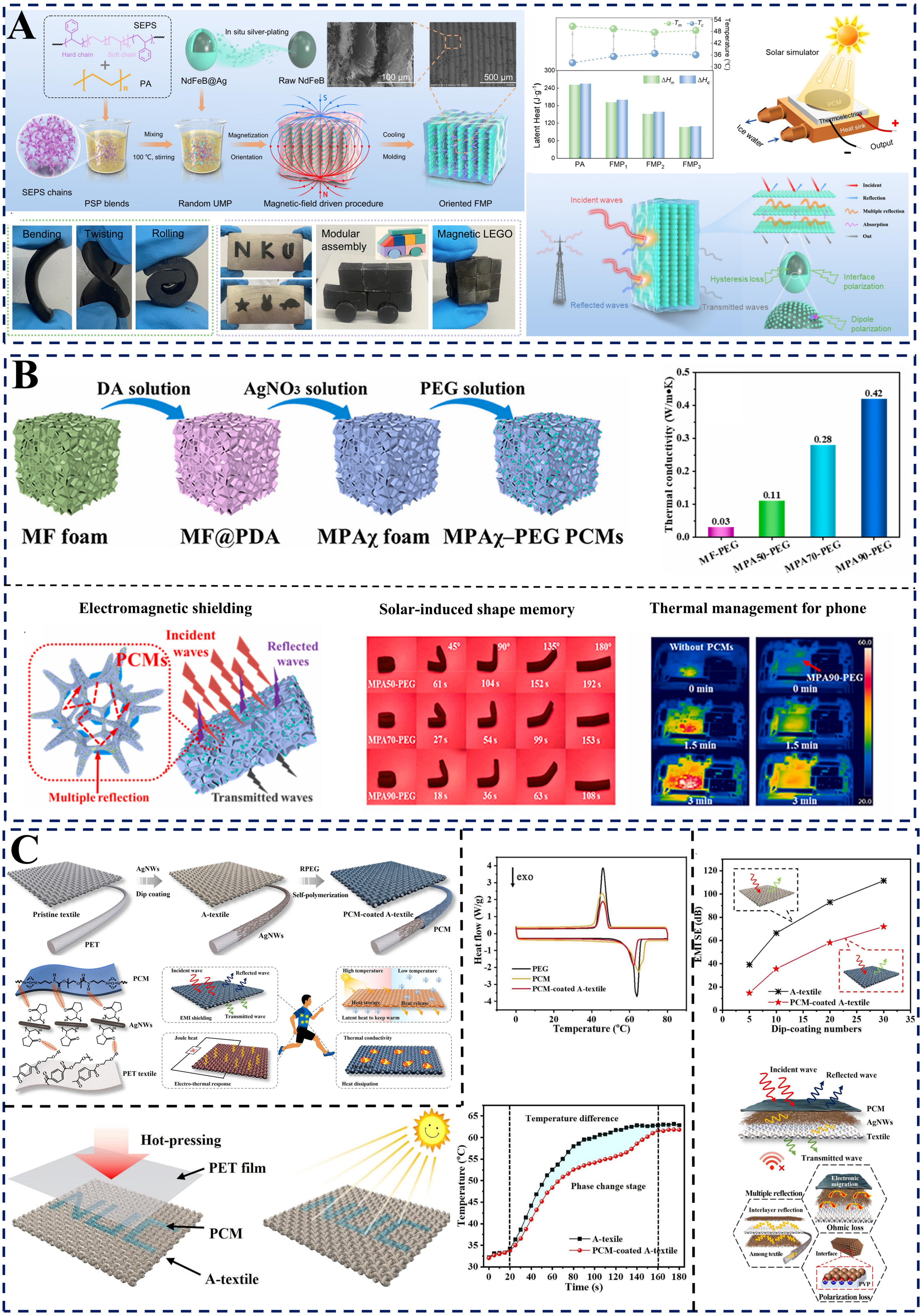

A
SEPS
Hard chain
Soft chain
PA
In situ silver-plating
NdFeB@Ag
Raw NdFeB
100 μm
500 μm
SEPS chains
PSP blends
Mixing
100 ℃, stirring
Random UMP
Magnetization
Orientation
Magnetic-field driven procedure
Cooling
Molding
Oriented FMP
Latent Heat (J·g⁻¹)
Temperature (℃)
PA
FMP1
FMP2
FMP3
Solar simulator
PCM
Thermoelectrics
Heat sink
Ice water
Output
Bending
Twisting
Rolling
N K U
Modular assembly
Magnetic LEGO
Incident waves
Reflected waves
Transmitted waves
Incident
Reflection
Multiple reflection
Absorption
Out
Hysteresis loss
Interface polarization
Dipole polarization
B
DA solution
AgNO3 solution
PEG solution
MF foam
MF@PDA
MPAχ foam
MPAχ–PEG PCMs
Thermal conductivity (W/m•K)
0.03
0.11
0.28
0.42
MF-PEG
MPA50-PEG
MPA70-PEG
MPA90-PEG
Electromagnetic shielding
Solar-induced shape memory
Thermal management for phone
PCMs
Incident waves
Reflected waves
Multiple reflection
Transmitted waves
45°
90°
135°
180°
MPA50-PEG
61 s
104 s
152 s
192 s
MPA70-PEG
27 s
54 s
99 s
153 s
MPA90-PEG
18 s
36 s
63 s
108 s
Without PCMs
MPA90-PEG
0 min
1.5 min
3 min
C
AgNWs
Dip coating
RPEG
Self-polymerization
Pristine textile
A-textile
PCM-coated A-textile
PET
AgNWs
PCM
PET textile
EMI shielding
Joule heat
Electro-thermal response
Thermal conductivity
Heat dissipation
High temperature
Low temperature
Latent heat to keep warm
exo
Heat flow (W/g)
Temperature (℃)
PEG
PCM
PCM-coated A-textile
EMI SE (dB)
Dip-coating numbers
A-textile
PCM-coated A-textile
Hot-pressing
PET film
PCM
A-textile
Temperature difference
Phase change stage
Temperature (℃)
Time (s)
A-textile
PCM-coated A-textile
Incident wave
Reflected wave
PCM
AgNWs
Textile
Transmitted wave
Multiple reflection
Interlayer reflection
Among textile
Electronic migration
Ohmic loss
Interface
PVP
Polarization loss

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Figure 10. Construction of flexible EMI PCCs based on metal particles. (A) Preparation of FMP composites, flexible display of digital images, thermal storage capacity, thermal charging, and schematic of EMI shielding mechanism. Reproduced with permission from ref[147]. Copyright 2024, Elsevier; (B) Preparation of MPA_{χ}-PEG composites, thermal conductivity, shape memory capability, schematic diagram of thermal management, and EMI shielding mechanism for cell phone chips. Reproduced with permission from ref[148]. Copyright 2022, Elsevier; (C) Schematic of composite material preparation, thermal storage capacity, photothermal conversion, thermal management, and EMI shielding mechanism. Reproduced with permission from ref[149]. Copyright 2023, Elsevier. EMI: Electromagnetic interference; PCCs: phase change composites; FMP: flexible and magnetically fastened phase change material; PEG: polyethylene glycol.

results in the formation of a complete conductive network. The impedance mismatch brought on by the buildup of free electrons on the surface of the AgNW conductive network causes a portion of an EMW to be instantly reflected when it strikes the composite textile material. Meanwhile, the addition of the PCM layer significantly improves impedance matching at the air/textile interface, enabling efficient dissipation of EMWs within the textile, while the AgNWs wrap the textile tightly to form a "wave-lock" structure. The EMI shielding performance is enhanced by this structure, which allows multiple EMW reflections and scattering within the textile cavity. When an alternating magnetic field is present, the AgNWs and the surface-wrapped polyvinylpyrrolidone (PVP) insulating layer also cause an asymmetric distribution of charge density and produce localized dipoles, which further aids in the attenuation of EMWs through polarization loss. Composite textile materials with ultra-high thermal storage capacity and thermal conductivity provide a promising solution for waste heat utilization in electronic devices and thermal management. These composite materials, when used as integrated heat sinks for LED chips, can significantly reduce the chip's surface temperature and enable stable operation at lower temperatures, demonstrating excellent heat dissipation capacity and thermal management. Therefore, flexible PCCs with heat storage capacity decorated with AgNWs have great potential in electromagnetic radiation protection, wearable smart clothing, and personal thermal management. Summary of properties of different flexible EMI PCCs is detailed in Table 4.

In summary, PCCs incorporating MXene, carbon nanomaterials, and metal nanoparticles as fillers provide a highly conductive network and thermal conductivity, while the flexible polymer matrix ensures flexibility and EMI shielding performance. These composites not only offer effective EMI shielding to reduce radiation damage to electronic equipment and the human body but also leverage the latent heat storage properties of PCMs for thermal management. This ability can lower the operating temperature of small and medium-sized electronic devices, prevent overheating, and extend their lifespan. Furthermore, these composites can also be used in wearable devices to regulate the comfortable temperature of the human body, reducing the external transmission of EMW radiation and ensuring human health. Additionally, PCMs can utilize electromagnetic radiation to prepare environmentally friendly and reliable cooling materials, i.e., radiate excess heat into outer space to cool objects spontaneously without causing any energy emissions, greatly expanding their thermal management performance and showing great potential. It is worth noting that the proportion of PCMs in the whole composite material should not be too large, not at the expense of EMI performance; weighing the proportion of the two is an important issue that deserves our further consideration. In conclusion, flexible EMI PCCs have broad applications in EMI shielding, thermal management of electronic devices, and human body thermal regulation.

CONCLUSION AND OUTLOOK

In recent years, flexible electromagnetic shielding materials have been continuously developed, with the EMI shielding effect improving due to the emergence of new materials and innovative structural designs, but a single EMI material cannot meet the needs of the information age of high-speed development, especially when the electronic equipment operation of the overloaded heat must be conducted to ensure the normal operation of the heat dissipation to prevent damage caused by thermal shock. To ensure normal

Table 4. Summary of properties of different flexible EMI PCCs

Type of filler		Phase change packaging technology	Properties	EMI shielding and enthalpy	Potential direction	Refs.
MP_2P_1	MXene	Microencapsulation	Skin affinity High mechanical property Thermoregulation Excellent thermal storage	87.80 dB 141.4 J/g	Personal thermal management EMI protection	[140]
PCC/MXene/PVA			High mechanical property High thermal conductivity High electrical	43.13 dB 136.80 J/g		[141]
PCNF	SWCNTs	Polymer molding	Breathability Water resistance Thermal regulation Radiation resistance Tensile properties	21.50 dB 65.00 J/g	Wearable phase change fabrics and EMI protection	[142]
SPP			High mechanical property Excellent thermal storage Thermal stability	51.70 dB 119.20 J/g	Heat dissipation and EMI shielding of small electronic devices	[143]
SSPCCs	CNTs	Porous adsorption	High thermal conductivity Thermal stability High mechanical property	41.20 dB 133.80 J/g	Thermal management of mobile phones	[144]
CPCM	GO		High thermal conductivity Radiation resistance Shape stability	45.00 dB 175.80 J/g	Thermal management and EMI shielding of electronic devices	[145]
MGGF	GO MXene		Thermal/electrical behavior Thermal reliability Shape stability	55.60 dB 160.30 J/g	Cooling and anti-EMI of electronic equipment	[146]
FMP	Ag	Polymer molding	High mechanical property High thermal conductivity High electrical Durability	35.42 dB 120.00 J/g	EMI shielding and thermal management of high-power devices	[147]
MPA_χ-PEG	AgNPs	Porous adsorption	Shape memory performance Excellent thermal storage High mechanical property	82.02 dB 148.90 J/g	CPU thermal management	[148]
PCM-coated A-textile	AgNWs	Polymer molding	Joule heating Excellent thermal storage Infrared anti-counterfeiting High thermal conductivity	72.00 dB 138.90 J/g	Wearable EMI protection and personal thermal management	[149]

EMI: Electromagnetic interference; PCCs: phase change composites; PVA: polyvinyl alcohol; PCNF: phase-change nonwoven fabric; SWCNTs: single-walled carbon nanotubes; SPP: SWCNT/PEG/PDMS; SSPCCs: shape-stabilized PCCs; CNTs: carbon nanotubes; CPCM: composite phase change material; GO: graphene oxide; MGGF: MXene-graphene foam; FMP: flexible and magnetically fastened phase change material; PEG: polyethylene glycol; AgNPs: silver nanoparticles; PCM: phase change material; AgNWs: silver nanowires.

operation and protect against damage from thermal shock, the PCM, as a new type of energy storage material, can provide thermal management. When combined with flexible electromagnetic shielding materials, the PCM design enhances both EMI shielding effect and thermal management performance. This new composite material accelerates the progress of multifunctional EMI shielding materials and broadens its potential applications. This paper systematically reviews the EMI shielding mechanisms, the preparation and recent advancements in flexible EMI shielding materials, the significant potential of flexible PCMs for thermal management, and typical examples of integrating flexible EMI shielding materials with PCMs. It is important to note that even though EMI shielding is equipped with an additional dual effect of thermal management, there are still challenges to overcome in the face of the rapid development of future electronic devices.

(1) When electronic devices are exposed to high levels of EMI radiation, PCMs may undergo thermal hysteresis. As a result, a large concentration of EMWs can cause a rapid increase in heat, potentially exceeding the material's ignition point. This may lead to irreversible damage; to mitigate this, a small amount of flame retardant can be added to flexible EMI PCCs (a high additive level may affect the material's brittleness and pose a challenge to processing), increasing the upper limit of the material's service life.

(2) To better play the EMI shielding effect in extreme environments, the new EMI composite materials that intelligently respond to external stimuli (strain, temperature, humidity, electric field, magnetic field) are worth further exploration. At the same time, the EMI performance will be greatly affected when the material is damaged due to excessive external action. At this time, EMI composites with special self-healing abilities need to be paid more attention.

(3) As the external environment continues to change, flexible materials with both EMI and thermal management will not be able to keep up with people's needs, so more attention should be paid to designing more special properties such as hydrophobicity, high sensitivity (sensor function), flame retardancy, and high transparent compression. It is worth noting that there may be mutual constraints between the functions, so the structure and performance of the EMI shielding must be designed in an integrated manner and the optimal balance of functions must be realized in the future.

In conclusion, the development of flexible EMI PCCs with multifunctionality is very rapid and necessary, and we believe that the widespread use of small and medium power appliances and wearable electronics will inspire flexible EMI PCCs with more novel functionalities to improve human life. We hope that this review will shed light on the future challenges and opportunities for the development of lightweight EMI thermal management materials and open new avenues for the creation of advanced and multifunctional integrated EMI shielding materials.

DECLARATIONS

Authors' contributions

Methodology, investigation, writing - original draft, writing - review and editing: Yan, R.
Investigation, writing - original draft, writing - review and editing: Lin, Q.
Investigation: You, K.
Methodology, investigation: Zhang, L.
Resources, review and editing, funding acquisition: Chen, Y.
Validation, resources: Huang, Z.
Writing - review and editing, supervision, project administration, funding acquisition: Sheng, X.

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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