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Review

From low conductivity to high energy efficiency: The role of conductive polymers in phase change materials

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ABSTRACT

Phase change materials (PCMs) face a significant obstacle in practical applications and energy efficiency due to their inherently low thermal conductivity. One promising solution to this limitation involves integrating conductive polymers (CPs) into PCMs. This approach not only enhances thermal conductivity—critical for efficient energy storage and release—but also introduces electrical conductivity, enabling dual functionalities such as electrothermal conversion and rapid charging and discharging. Although CPs have been extensively utilized for this purpose, there is a noticeable gap in existing reviews that specifically focus on CP-enhanced PCMs. To address this gap, this comprehensive review examines experimental research aimed at improving the electrothermal characteristics of PCMs, with an emphasis on boosting conductivity and storage efficiency through CP incorporation. The review begins by providing an overview of the fundamental principles of electrical and thermal conduction in materials. It then explores commonly used CPs—such as polypyrrole, polyaniline, and poly(3,4-ethylenedioxythiophene) (PEDOT)—and their integration strategies with PCMs. The discussion highlights the unique properties of these polymers and their contributions to enhancing the thermal and electrical conductivity of PCMs. Additionally, it investigates the formation of conductive pathways and their role in amplifying the energy efficiency of nano-enhanced PCMs, comparing the effects of various nano-additives. The study further explores potential applications of CP-enhanced PCMs across diverse fields, including electronics, wearables, energy systems, and advanced thermally regulative materials. To provide a well-rounded perspective, the review outlines recent advancements, identifies current challenges and limitations, and highlights future research opportunities. By fostering a deeper understanding of the interplay between PCMs and CPs, this review contributes to the ongoing efforts to optimize thermal properties and multifunctionality, paving the way for innovative applications and improved energy solutions.

1. Introduction

Phase change materials (PCMs) store heat by undergoing a phase transformation, typically absorbing thermal energy as they melt. This heat can then be released upon cooling, as the PCM crystallizes back into a solid. The use of PCMs opens intriguing prospects for thermal management across various domains [1–3]. Particularly, organic solid–liquid PCMs, including paraffinic waxes [4,5], fatty acids [6–8], sugar alcohols [9,10] and polyethylene glycols (PEG) [11–13], have garnered considerable scientific and industrial interests. This preference stems from their noteworthy attributes, such as high latent heat enthalpies, environmentally relevant melting temperatures, absence of phase segregation, and exceptional stability for prolonged usage [14–17]. To further enhance the properties and applicability of PCMs, considerable efforts have been directed toward shape stabilization and microencapsulation techniques, which improve their structural integrity and

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thermal management efficiency [18,19].

Despite their notable advantages, PCMs face a fundamental limitation in their intrinsic low thermal conductivity, causing low heat transfer rates [1,2,20]. The thermal conduction of materials is influenced by their structural and chemical properties. PCMs' low thermal conductivity stems from the amorphous arrangement and disruptions in the vibration of the molecular chains as well as the presence of weak intermolecular interactions [21,22]. The structural disturbances hinder efficient heat transfer by scattering phonons, while the weak interactions between molecules further impede the propagation of thermal energy [15–17,23]. Phonon transport is limited to the crystalline structure of PCMs, while in their liquid phase the transport mechanism is based on random motion of particles. This inherent constraint significantly hampers the practical applicability and efficiency of PCMs in energy utilization. Therefore, a thorough understanding of thermal conduction is imperative for developing efficient thermal management systems and the optimization of various devices based on PCMs. Consequently, substantial research efforts have been directed towards improving the conductivity and functionality of PCMs, with a specific emphasis on advancing applications in sophisticated energy systems.

PCMs show the capacity to store heat generated from electricity. This makes them useful in interesting ways, including in delay switches, converting off-peak electricity, storing temperature-sensitive items, cooling mobile electronics, and powering hybrid electric vehicles [24,25]. Unfortunately, typical PCMs have low or no low electrical conductivity, [26] a factor that can be addressed by integrating highly (electrically) conductive additives, resulting in hybrid PCMs [27,28]. Conductive additives such as metals, carbon, and polymers have been considered for addition to PCMs through solution or melt blending, aiming to create enhanced electrical conductivity.

Nanoscale design of materials, processes, and systems enable new interactions unfeasible with bulk materials and reduces charge transport pathways [29]. Nanoparticles, e.g., nano-sized carbon [8,30] and mental oxide particles [31–33], are known to elevate the electrical conductivity. Yet, the significant thermal or electrical resistance at the interface between nanoparticles and the matrix material typically offsets any potential advantage of these highly conductive additives. At low volumetric fractions, nanoparticles within the PCM matrix do not form a percolating network, leading to high interfacial resistance at the boundaries of particles with PCM and substantial thermal resistance among the particles. This results in reduced thermal conductivity in the hybrid additive-PCM systems [34,35]. While augmenting the loading of additives can establish a continuous network, an excessively high fraction may compromise other crucial PCM properties, including mechanical characteristics and thermal storage capabilities. Hence, there is a tradeoff between additive addition and electrical and thermal performance.

Going deeper in the possible nano-additives that can be used with PCMs, it is worth considering their dimensionality, often classified as zero-, one-, two-, and three-dimensional structures (0D, 1D, 2D, and 3D) [36,37]. The 0D nanoparticles, like carbon quantum dots and metal nanoparticles, enhance nanoscale micro-convection and thermal conductivity. However, their aggregation and phase separation tendency in the PCM melt hinders phonon transport, negatively affecting PCMs' thermal properties. The 1D nanostructures, like carbon nanofibers and carbon nanotubes (CNT), form linear heat transfer pathways. Meanwhile, 2D nanostructures, such as graphene and graphene nanoplatelets (GNP) [38,39], create planar heat transfer paths but can suffer from phonon scattering and unstable thermal conductivity during phase transitions. The 3D nanostructures, including expanded graphite (EG), graphene aerogel, and graphene foam, form network-type thermal pathways that enhance phonon transport, reduce scattering, and prevent nanoparticle agglomeration. The heat transfer efficiency of nano-additives follows the trend 3D > 2D > 1D > 0D, as higher-dimensional nano-additives form more efficient thermal networks, improving phonon transport, reducing scattering, and lowering contact thermal resistance [36,37]. Fig. 1 illustrates these mechanisms, showing that thermal conductivity increases with both additive dimension and fraction. In Fig. 1 e, the thermal conductivity enhancement multiplier (k_{eff}), calculated as $(k_c–k)/k$ (where k_c is the composite conductivity and k is the PCM conductivity), demonstrates an order-of-magnitude improvement as dimensions increase, driven by the formation of robust thermal networks [36].

Building on nanoscale innovations, conductive polymers (CPs) offer a transformative path for enhancing PCMs, marking a pivotal step forward in the creation of next generation energy materials and systems. The CPs blend the beneficial qualities of traditional polymers with the exceptional electronic characteristics of metals and semiconductors [29]. They are distinguished by their highly π-conjugated polymeric chains. By forming continuous conductive networks through uniform crosslinking, CPs can substantially improve thermal and electrical conductivity. Especially, nanostructured CPs provide several key benefits, including enhanced surface area and flexibility as well as reduced charge transport pathways. Various forms of CPs have been already implemented in energy conversion and storage systems [40]. CPs with 3D polymeric networks can majorly improve heat transfer within PCMs by fostering optimal interfacial interactions between the PCM and polymer, thereby boosting conductivity. For instance, CP hydrogels [41,42] with 3D hierarchical porous frameworks have gained great interest thanks to their intrinsic conductive characteristics. CPs have been also utilized to enhance the efficacy of metal catalysts by boosting their catalytic activity, conductivity, and stability [43]. Additionally, combining other types of conductive nanocomponents—such as metal or carbon nanoparticles—further improves the electrical and thermal conductivity of CP-based hybrid systems, enhancing their suitability for cutting-edge energy applications [44].

CPs offer several advantages over other conductive additives such metallic and ceramic fillers for enhancing the thermal conductivity of PCMs. Their lightweight and flexible nature makes them ideal for applications in electronics and wearables where such features are essential [45]. They minimize the risks of sedimentation or phase separation during thermal cycling, ensuring consistent performance. Additionally, CPs provide corrosion resistance and better thermal expansion compatibility with PCMs, particularly corrosive ones like salt hydrates and fatty acids [46,47]. Their thermal and electrical properties are tunable, and they are often more cost-effective and environmentally friendly, with lower processing demands [48] compared to metals or ceramics. Furthermore, functionalization with materials like graphene can significantly enhance their thermal conductivity [49], by reducing the thermal interface resistance for instance. These advantages make conductive polymers a superior choice for applications requiring durability, stability, and adaptability.

This review formulates an in-depth synthesis of the key factors governing electrical and thermal conduction in polymeric materials and PCMs, with a particular emphasis on improving PCM conductivity through the integration of CPs. It begins by introducing the primary mechanisms underlying thermal and electrical conductivity, alongside the factors that influence PCM performance. The review then delves into the incorporation of various CPs, examining in detail how these materials impact and enhance PCM conductivity. Finally, it addresses the challenges and prospects associated with the integration of CP-enhanced PCMs, exploring both technical challenges and potential advancements. Although CPs offer exceptional potential for energy applications, no comprehensive review currently exists that addresses this specific intersection of CPs and PCMs. Here, we provide valuable insights into designing CP-enhanced PCMs, highlighting recent innovations and emerging applications that promise to elevate energy efficiency in electrothermal conversion and storage technologies. This review thus positions CP-enhanced PCMs as a promising foundation for the next generation of advanced energy systems.

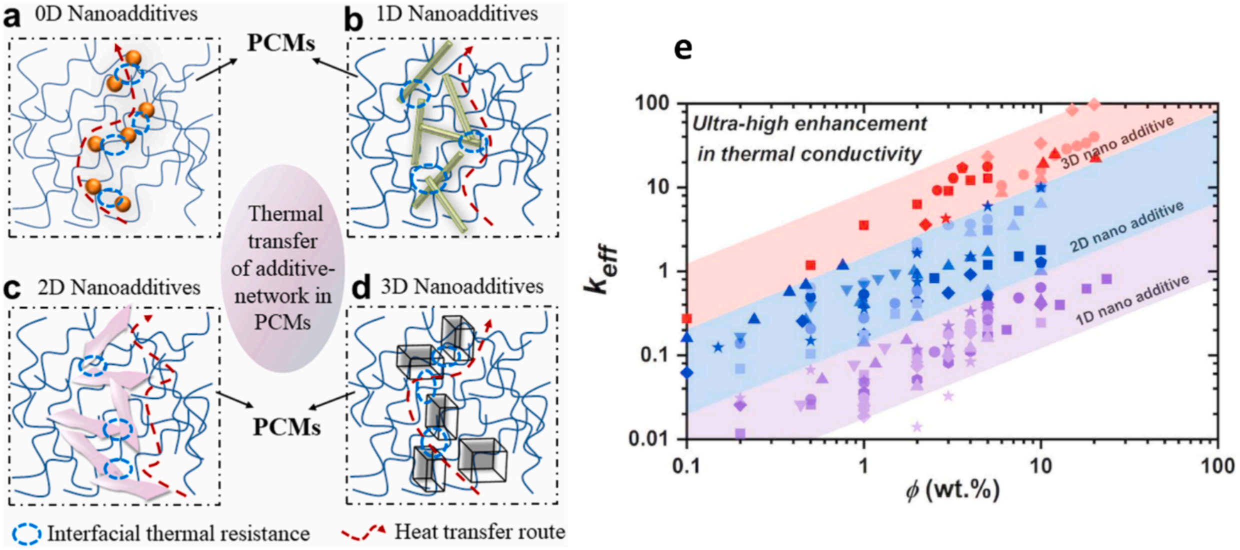

Fig. 1. Heat transfer of nano enhanced PCMs by nano-dimensional additives: a) 0D, b) 1D, c) 2D and d) 3D nano-additives, reprinting permission from [37]. e) Thermal conductivity enhancement multiplier versus nano-additive loading (wt.%), permission from [36].

2. Fundamentals of conductivity

2.1. Electrical conduction

Electrical conduction measures a material's capacity for enabling the passage of electric current. The electrical conductivity of materials is the reciprocal of their resistivity, typically expressed in unit of S/cm. Materials can be considered as insulators (conductivities $< 10^{-8}$ S/cm), semiconductors ($10^{-8} <$ conductivities $< 10^{3}$ S/cm), and conductors (conductivities $> 10^{3}$ S/cm) [50]. Pure PCMs, apart from the less common metallic types, lack the desired electrical conductivity (ranging from 10^{-12} to 10^{-7} S) and require further modifications [51]. Common approaches include integration with conductive metal, carbon, or nanoparticle additives. The electrical performance of these additive-enhanced PCM composites depends on continuous conductive networks formed by the conductive additives. Inter-aggregate conduction, field emission, and electron tunneling play a role in electrical conduction in such nano-enhanced composites [52,53]. The conduction mechanism depends on the distribution of additive nanoparticles, whether they are separated or in contact to form a conductive network. Conduction can occur via direct contact between particles or through electron tunneling across a thin layer of polymer.

Nano-additives create a continuous conductive pathway, or percolated network, which forms above a critical particle fraction called the percolation threshold [54], typically 5 vol% or less [22]. Under an electric field, electrons travel across this network, significantly increasing electrical conduction. Proper nanoparticle dispersion is essential, as agglomeration hinders network formation. Interestingly, non-uniform nanoparticle distribution can also enhance conductivity at lower loadings through creation of segregated structures, where nano-additives are concentrated in specific regions. This local concentration reduces the percolation threshold for creating conductive nanocomposites with lower additive loadings [55].

When the additive fraction (ϕ) reaches the percolation threshold (ϕ_C), the composites shift from insulators to conductors, dramatically increasing conductivity when the initial conducting channels form. As more conductive additive is added, additional pathways are created, gradually increasing conductivity until it saturates. The electrical behavior of composites is explained by the power law [54,55]:

$$\sigma = \sigma_0(\varphi - \varphi_c)^t \tag{1}$$

where σ represents the electrical conductivity, σ_0 is a constant related to the intrinsic conductivity of the additive, and t symbolizes the critical exponent, approximately 2 for 3D and 1.3 for 2D structures. However, experimental values often deviate from these predictions [55]. The relationship between σ and resistivity, ρ, is given by the reciprocal relationship:

$$\sigma = \frac{1}{\rho} \tag{2}$$

The value of ρ is determined using the resistance (R), typically assessed through the 4-point probe method. For this technique, a steady electrical current (I) passes through two electrodes positioned on the surface of the material, while the voltage (V) generated across another pair of electrodes is measured [45,56]. The ρ is defined by the following relationship:

$$\rho = R.\frac{A}{L} \tag{3}$$

where R represents the resistance, Ω, while A and L indicate the cross-sectional area (m^2) and the length of the material (m). By combining these formulas, the electrical conductivity can also be expressed as:

$$\sigma = \frac{L}{R.A} \tag{4}$$

For instance, the electrical conductivity of electrospun PCM fibers was measured through the four-point probe method [57]. As loadings of conductive additives were increased, electrical conductivity of the composites was considerably enhanced from 6.13×10^{-4} to 0.01 S/m. More details on the electrical conduction of CP-enhanced PCMs are provided in section 4. However, the breakdown mechanisms of solid materials, polymers in this case, are complex and not yet fully understood. Specifically, for solid materials, there is a lack of precise information regarding their electrical conductivity and the shift from insulation to conduction [58]. This area is characterized by significant approximations, which can lead to uncertainties and inconsistencies. For instance, as noted in Eq (1), existing approximations may fail to accurately predict behavior under varying conditions.

2.2. Thermal conduction

Thermal conduction measures a material's performance in conducting heat. In solids, the transmission of thermal energy occurs either through charge carriers like holes and electrons or phonons which are energy packets produced by vibrations in the atomic lattice. The thermal conductivity in insulators and semiconductors is predominantly influenced by phonons, while in metals, charge carriers play the major role. Phonons-based energy transmission is the main way of thermal transfer in most polymers [59,60]. The phonon-carried thermal conductivity, k, is measured via Eq (5):

$$k = \frac{VlC_v}{3} \tag{5}$$

where V denotes the velocity of phonon, l signifies its mean free path, and C_v represents the heat capacity. Thermal conduction in materials is majorly influenced by density and temperature. Typically, it rises with temperature until it peaks, then declines sharply as the material approaches its melting point. In general, it is highest in solids and lowest in gases; while in porous or low-density materials, air pockets hinder heat transfer, reducing conductivity. Additional factors such as grain boundaries, crystal orientation, and chemical complexity also impact thermal conductivity, with smaller grain sizes and complex compositions typically reducing its value [61]. Anisotropic materials exhibit directional dependencies, allowing heat to flow more easily along certain orientations, such as fiber directions. While higher density often correlates with greater thermal conductivity, this relationship can vary with temperature. For gases, heat conduction usually increases with temperature, whereas most liquids experience a decrease. In solids, conductivity strongly depends on molecular arrangement, with ordered crystal structures often showing high conductivity that remains stable with temperature changes [62].

Although most polymers, excluding CPs, exhibit a low thermal conductivity, they offer significant benefits including great processability, lightweight nature, corrosion resistance, and cost-effectiveness [59,63]. Consequently, polymer-based conductive materials are frequently sought after in various applications. The conductivity of constituent components influences the overall conductivity of polymeric composites. Nevertheless, polymers' thermal conductivity is frequently overlooked in the development of conductive composites, as it mainly is lower than that of the nano-additives. However, the *k* value of polymers becomes especially crucial at lower additive fractions, where the additives lack forming a continuous network in the matrix. Therefore, the matrix acts as a thermal blockage and constrains the heat transfer pathway [59].

Polymer composites are widely understood to follow a thermal conduction mechanism rooted in percolation theory, akin to the electrical conduction mechanism. The blends of polymers with thermally conductive additives often necessitate a substantial volume fraction of the additives (typically exceeding 30 vol%) to provide a notable improvement in thermal conductivity. The percolation threshold for thermal conduction is considerably higher than the electrical percolation threshold, which is usually 5 vol% or less [22]. At low additive fractions, the particles remain isolated and are unable to form a continuous conductive network. Under these conditions, heat conduction is primarily influenced by the coupling between the additive and the polymeric media, the interfacial and thermal resistance between them, and the geometry and loading of the additive particles [64]. Once the additive fraction reaches a sufficiently high level, the particles make contact, leading to the creation of thermal conduction pathways.

Thermal conductivity measurements are classified as steady-state and transient approaches [62,65]. Steady-state techniques, like the heat flowmeter and guarded hot plate, rely on Fourier's law by estimating the temperature difference across the materials under a steady flow of thermal energy. These methods work well for composites with low conductivity but require larger samples and longer test times [62]. Transient methods, such as the transient plane source (TPS) and laser flash apparatus, use heat pulses or periodic signals, measuring thermal diffusivity (α) by a temporary heat flow. The material's density (ρ) and specific heat capacity (C_p) influence α as described below.

$$\alpha = \frac{k}{\rho C_P} \tag{6}$$

These measurement techniques function within specific temperature limits and are designed to evaluate thermal conductivity across various ranges as thoroughly has been discussed by Palacios et al. elsewhere [62]. The TPS method provides the broadest measurement range, capable of assessing thermal conductivity values from 0.001 to 1800 W/m·K. The laser flash and the transient hot wire/hot strip techniques cover ranges of 0.1–1000 W/m·K and 0.005–500 W/m·K, respectively. In terms of operating temperature ranges, the laser flash, TPS, and guarded hot plate methods are the most versatile, with ranges spanning −120 to 2800 °C, −35 to 1000 °C, and −160 to 700 °C, respectively. For instance, the TPS technique has been widely used to measure the thermal conductivity of organic PCMs [10]. Factors such as sample preparation method, surface morphology, and crystalline structure were found to be affecting the thermal conductivity of measured PCMs. Further details on the thermal conductivity values of CP-enhanced PCMs are discussed in Section 4. However, it should be mentioned that measuring thermal conductivity accurately is generally challenging in thermal energy storage field, as issues like thermal contact resistance and material non-homogeneity can affect measurement reliability.

3. Conductive polymers (CPs)

Initially viewed as electronically insulating due to the presence of covalent bonds within their saturated carbon chain, polymers underwent a transformative shift with the groundbreaking work of Nobel laureates MacDiarmid, Shirakawa, and Heegeron [66–68] on the discovery and subsequent development of CPs. Unlike their traditional counterparts, CPs feature an extended π conjugated system, facilitating the delocalization of a significant number of π electrons. This delocalized electron structure endows these polymers with both electrical and thermal conductivity, placing them within the semiconductor range by creating a distinct gap between the valence and conduction bands [48]. However, a doping process is required for achieving enhanced conductivity. In CPs, electron mobility is facilitated by the widespread delocalization along the entire polymer chain, a phenomenon governed by the symmetry of the chain itself. Thus, the conductivity of these polymers is intricately tied to their molecular structure and arrangement [45].

Polypyrrole (PPy), polyaniline (PAni), and poly(3,4-ethylenedioxythiophene) (PEDOT) stand out as highly promising CPs with diverse applications, including in energy [69], electronics [69,70], sensors [71,72], and biomedical contexts [73]. These CPs demonstrate electrical and optical characteristics resembling those of both metals and semiconductors, while preserving the benefits typically linked with traditional polymers, like their straightforward production process [45]. These properties combined with structural and thermal stability and lightweight make CPs increasingly attractive for replacing metals in electronic devices. For example, flexible electronics based on CPs have gained significant attention for applications in wearable sensors, soft robotics, and human–machine interfaces.

You et al. [74] introduced PPy into porous gelatin-reduced graphene oxide organohydrogel, designed for high electrical conductivity (5.25 S/m), breathability (14 g/(cm^2.h)), and mechanical flexibility. The developed material was used as a flexible electrode to capture electrophysiological signals and control a robotic arm and virtual character. It demonstrated excellent performance as a human–machine interface, with promising applications in health monitoring, sports tracking, and operational control. In another study, nanocomposites were synthesized via chemical oxidative polymerization of aniline with silica-coated iron oxide, yielding nanotubular and granular forms by adjusting acid concentration and mixing parameters of the synthesis method [75]. The PAni-enhanced nanocomposites exhibited magnetic and electrical responsiveness, with granular particles showing higher conductivity (1.1 S/cm) but lower magnetization (21 emu/g) compared to the nanotubular form (4.8×10^{-2} S/cm, 27 emu/g). Cytocompatibility and hemocompatibility varied dose-dependently, highlighting their potential for biomedical applications such as biosensing, drug delivery, bioelectronics, and regenerative medicine. PEDOT was hybridized with multiwall carbon nanotubes (MWCNTs) for solid-state supercapacitors using an anion-doped ionic liquid gel electrolyte [76]. Composite electrodes were fabricated via co-electrodeposition at 4 mA cm^{-2} with 10 ms pulses in an aqueous medium containing MWCNTs, Na-dodecyl sulfate surfactant, and 10–20 % poly(styrene sulfonate) dopant (PSS). The optimized PEDOT:PSS (10 %)-MWCNT electrodes exhibited a high specific capacitance of 138 F/g and exceptional charge transfer properties with minimal resistance. The supercapacitors demonstrated a high-power density of 6.33 kW/kg at 3.43 Wh/kg and retained 80 % capacitance over 500 cycles, stabilizing beyond 2000 cycles with 95 % Coulomb efficiency.

The preparation methods, reaction conditions, and the used dopants can all influence the electrical conductivity of CPs. Consequently, a given CP may show a wide electrical and thermal conductivity range [48]. The conductivity observed in CPs is attributed to the conjugated double bonds on the polymeric chain. The π electrons within the conjugated structure can delocalize into a conduction band. However, this system becomes unstable because of bond alternation, causing an energy gap in the electronic spectrum. To bridge this energy gap and confer conductivity, dopant ions can be incorporated into their structure. These dopant ions hold additional electrons charge, neutralizing the unstable oxidized polymeric chain by either donation or acceptance of electrons. Under applied potential, a charge is transmitted through the material due to a flux of ions. P- and N-type dopants including Cl^{-}, Br^{-}, or NO^{3-} ions as well as hyaluronic acid and polymeric dopants can be used in the doping process of CPs [50]. For instance, the conductivity of PPy can change from approximately 0.1 to 100 S/cm with doping. This reversible doping and de-doping process makes CPs attractive for applications in

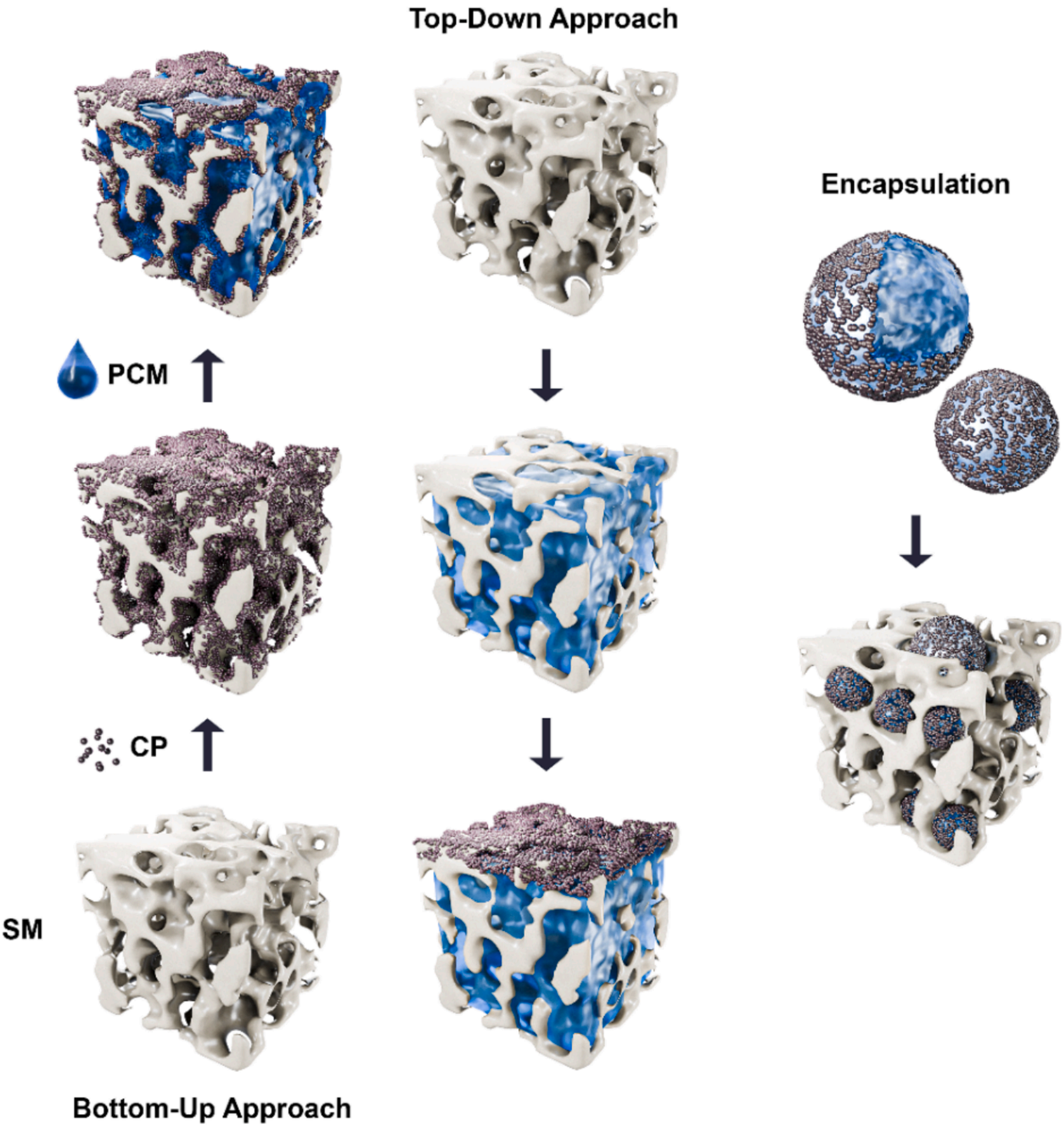

Fig. 2. The incorporation of CPs with PCMs through three distinct methods: bottom-up, top-down, and encapsulation. SM stands for the support matrix.

energy systems, sensors, and actuators [29].

The synthesis of CPs is performed via chemical or electrochemical approaches. Condensation polymerization and addition polymerization are the primary approaches in chemical synthesis. The former approach is grounded in the removal of the small molecules like hydrochloric acid or water, while the latter method encompasses various types such as radical, cation, and anion polymerizations. Chemical synthesis offers numerous pathways to create a diverse array of CPs and enables the scalability of these materials. In the electrochemical polymerization, the electrodes are placed in the monomer solution with a suitable solvent and the dopant electrolyte. The passage of current causes electrodeposition primarily at the positively charged working electrode, where the monomers undergo oxidation for the generation of radical cations. The radical cations then engage in reactions with other substances, leading to the formation of insoluble polymer chains [45].

4. CP-enhanced PCMs

Integrating CPs into PCMs greatly enhances their thermal conductivity and functionality, thanks to their inherent electrical conductivity and capacity to form continuous conductive networks within the PCM. This improvement arises from the creation of a CP conductive network, which promotes efficient heat transfer during phase transitions, boosting the composite's overall thermal conductivity. A thorough review of the literature reveals three distinct approaches for incorporating CPs into PCMs. Here, these approaches are classified as bottom-up, top-down, and encapsulation, as illustrated in Fig. 2. In bottom-up approach, the CP is first coated on the surface of a support matrix (SM). Subsequently, the PCM is impregnated or confined within this CP-coated SM. Top-down approach involves initially shape-stabilizing the PCM with SM. After the PCM is stabilized, the CP is coated onto the surface of the PCM-SM composite. In encapsulation method, the PCM is first encapsulated with the CP. The CP-encapsulated PCM can then be incorporated into a support carrier or be used as it is. Each of these methods offers unique advantages depending on the targeted applications and the desired properties of the composites including mechanical stability, latent heat, and conductivity.

These methods have also their unique set of challenges that need to be addressed to optimize their performance for specific thermal management applications. Balancing the advantages and disadvantages will help the identification of the most appropriate method for a desired application. Table 1 is a tabulation of the advantages and disadvantages associated with these methods, tailored for different thermal management applications. The bottom-up approach enhances thermal conductivity through a uniform CP coating on a support matrix, which can ensure effective heat dissipation and mechanical stability. This method is suitable for applications requiring precise thermal management, such as electronic cooling [77,78] or building materials. However, its complex manufacturing process, challenges in achieving uniform coatings, and material compatibility issues can pose significant hurdles [6]. In contrast, the top-down approach focuses on surface functionalization, enabling post-stabilization modifications that enhance properties like hydrophobicity and durability during thermal cycling [7]. While this approach is flexible and suited for advanced thermal systems and smart wearables, its impact is often limited to surface properties, with concerns about coating durability and potential delamination [79]. Finally, the encapsulation method provides robust leakage protection and improved phase change efficiency by pre-encapsulating the PCM with CP, making it suitable for applications in battery thermal management [80] and aerospace [81]. However, the added volume, weight, and thermal interface resistance introduced by encapsulation, coupled with its complexity, can limit its adoption in space- and weight-sensitive scenarios. Each method balances trade-offs between thermal performance, durability, and manufacturing complexity, making them suitable for different application needs. Table 2 lists various CP-enhanced PCMs developed through the three approaches identified in recent literature.

Table 1

Benefits and drawbacks of three identified methods of CPs integration with PCMs, tailored for different thermal management applications.

Method	Advantages	Disadvantages
Bottom-Up Approach	Enhanced thermal conductivity through uniform CP coating Efficient heat dissipation Ideal for applications requiring efficient heat dissipation, for example to avoid the heat concentration in small areas in miniaturized electronics [77,78] Controlled impregnation leading to more precise thermal management Suitable for electronic cooling where specific thermal performance is critical Mechanical stability and structural integrity provided by the support matrix Beneficial for applications where mechanical stability is essential, such as in building materials for thermal regulation	Complex and time-consuming manufacturing process Uniform coating challenges potentially causing inconsistencies in thermal conductivity and performance Material compatibility limited by the adhesion and stability of CP, support matrix, and PCM
Top-Down Approach	Improved surface functionalization and surface properties (e.g., hydrophobicity) by CP coating Ideal for applications like smart textiles or flexible electronics [82,83] Flexibility to tune thermal and electrical properties through post-stabilization modifications Beneficial for advanced thermal storage systems where performance tuning is needed after initial fabrication Enhanced thermal cycling stability provided by the CP coating	Surface coating limitations Limited enhancements to surface properties Durability concerns CP coating wearing off, especially in applications involving mechanical abrasion Delamination concerns due to thermal expansion mismatches
Encapsulation Method	Superior protection against leakage and degradation due to encapsulation Suitable for high-reliability applications such as battery thermal management [80] Flexibility in choosing support materials The choice of support can be optimized for different applications without compromising PCMs' stability advantageous for thermal management of automotive and aerospace industries [81] Improved phase change efficiency and transitions due to uniform thermal interface	Encapsulation complexity potentially increasing production costs and time. Volume and weight increase due to encapsulation Thermal and interface resistance due to the encapsulation layer and discontinuity in between the capsules The dispersion of high conductive parts does not offer continuous pathways for heat carriers Lack of continuous pathways for heat flow

The long-term usability of these PCM composites largely depends on the thermal cycling capacity of the incorporated PCMs. A common approach for evaluating cycling performance involves assessing the thermal durability of CP-enhanced PCMs, typically based on changes in phase transition temperature and enthalpy values over more than 100 heating–cooling cycles. For instance, DSC curves for PPy-coated PCM textiles [7] and PEDOT-enhanced PCM textiles [49] investigated over 80–100 thermal cycles showed minimal changes in melting and crystallization temperatures, as well as latent heat, demonstrating the

Table 2

Commonly used CPs, their incorporation mechanisms, support matrix, and developed properties in PCMs.

CP	Incorporation approach	PCM	Support matrix	Shape Stabilization mechanism	Melting enthalpy (J/g)	k (W/mK)	σ (S/m)	Ref.
PPy	Top-down	Decanoic acid	Polyamide 11	Electrospinning	113	0.28	28.89	[7]
PPy	Encapsulation	Commercial microcapsules made of MF resin shell & paraffin core	Polyurethane (PU)	Wet Spinning	231	–	–	[98]
PPy	Bottom-up	n-octacosane	CNF	Aerogel	239–258			[99]
PPy	Bottom-up	Paraffin	PPy	Aerogel	72	–	–	[100]
PPy	Bottom-up	Fatty acid	Sodium alginate & flax fiber	Sponge	101 & 154	0.45	1.52	[6]
PPy	Bottom-up	Paraffin	CNT	Aerogel	148	0.64	–	[95]
PPy	Bottom-up	PEG	Wood	Delignified Wood	160	0.76	–	[101]
PPy	Bottom-up	PEG	CNF	Sponge	170	–	178	[26]
PPy	Bottom-up	PEG	CNF	Membrane	155	–	613	[102]
PPy	Bottom-up	Paraffin	Fe_3O_4- functionalized hollow kapok fiber	Aerogel	161	1.06	–	[103]
PPy	Bottom-up	PEG	Expanded Graphite (EG)	Porous graphite	148	–	–	[104]
PPy	Bottom-up	1-Hexadecylamine	EG	Porous graphite	205	0.344	–	[105]
PAni	Top-down	Palmitic acid	Exfoliated graphite	Porous graphite	158	1.08	–	[106]
PAni	Encapsulation	Paraffin	PAni	Blending	183	0.55	–	[107]
PAni	Encapsulation	Paraffin	PAni	Probe sonication	166	0.29	–	[108]
PAni	Encapsulation	Erythritol	PAni + EG	Encapsulation	254.3	0.80	–	[109]
PAni	Encapsulation	Erythritol	PAni + Silver nanowires	Encapsulation	220	0.35	–	[110]
PAni	Encapsulation	Stearic acid	PAni	Encapsulation	113	–	0.70	[111]
PAni	Encapsulation	Paraffin	PAni	Encapsulation	103	0.28	–	[112]
PAni	Bottom-up	n-docosane	Silica	Encapsulation + CNT	143	0.84	–	[113]
PAni	Bottom-up	Erythritol	GO modified Melamine foam	Foam	259	1.59	–	[114]
PAni	Bottom-up	PEG	MXene	Aerogel	151	0.292	–	[115]
PEDOT: PSS	Top-down	Lauric acid	PU	Electrospinning	124		0.29	[116]
PEDOT	Encapsulation	n-eicosane	Silica	Encapsulation	130	–	–	[117]
PEDOT: PSS	Bottom-up	PEG	GO + GNPs	Aerogel	211	1.12	–	[118]
PEDOT: PSS		n-octadecane	PU + CNT+ graphene	Wet spinning	166	–	1.96×10^4	[49]
PEDOT: PSS	Bottom-up	Sodium sulfate decahydrat	Polyacrylamide + MXene	Organohydrogels	130	–	–	[119]
PEDOT: PSS	Bottom-up	PEG	MXene	MXene framework	237	0.36	–	[120]
PEDOT: PSS	Bottom-up	Paraffin	Melamine	Encapsulation	108	0.16	–	[121]

excellent thermal cycle stability of CP-enhanced PCMs. Additionally, some studies have investigated cycling performance over thousands of DSC cycles. For example, a study on a PAni-enhanced paraffin composite demonstrated superior latent heat reliability compared to pure paraffin, even after 3,000 thermal cycles [84].

4.1. Polypyrrole (PPy)-enhanced PCMs

PPy stands out as a promising CP because of its conductivity, chemical stability, corrosion resistance and redox properties. It also benefits from commercially available monomers and straightforward synthesis process for achieving various structures [85], showing electrical conductivity within 2–100 S/cm range [86]. These attributes make PPy highly suitable for various applications, including biosensors [87–89], mechanical actuators [90,91], drug delivery systems[92,93], and supercapacitors [85]. Additionally, PPy is more flexible and has a greater mass density compared to most other CPs, allowing it to deliver greater performance in a smaller size, adaptable to various shapes. This flexibility and lightweight nature make PPy ideal for fabricating high-performance materials for portable and flexible electronics, a rapidly growing area in the energy storage field. The desirable properties of PPy are closely linked to the synthesis methods. To achieve specific characteristics such as high mechanical flexibility and biocompatibility, various PPy composites with different morphologies and structures are produced, ranging from microscale to nanoscale [85]. Therefore, exploiting conductive PPy for the shape stabilization of PCMs is a valuable approach.

When integrated into PCMs, PPy can significantly enhance their thermal conductivity and functionality, primarily due to its intrinsic electrical conductivity and ability to form continuous conductive networks within the PCM matrix. The mechanism behind this improvement involves the establishment of a conductive network by PPy, facilitating efficient heat transfer during the phase transition process and thereby improving the overall thermal conductivity of the composite material. For example, a study demonstrated that incorporating PPy into paraffin wax increased the thermal conductivity from 0.271 W/m·K of the pure paraffin to 0.391 W/m·K of the hybrid material, representing a 44 % enhancement [94]. Additionally, PPy provides good adhesion to the substrate, which can reduce interfacial thermal resistance [95]. Its strong near-infrared absorbance and photothermal conversion efficiency enable high photostability during prolonged and repeated near-infrared irradiation [96]. The PPy ring holds numerous nitrogen atoms, enhancing its stabilization ability of organic PCMs, like PEG, by facilitating hydrogen bonding interactions [97]. As a result, PPy emerges as a strong contender for light/electric-to-thermal energy conversion, as it combines the abilities to absorb light, conduct electricity, and efficiently transfer heat during the phase transition process.

The development of flexible PCM textiles with photothermal conversion and storage properties represents a promising method for solar energy utilization. In this context, photothermal microcapsules were developed through the PPy encapsulation method and integration into a flexible polyurethane (PU) textile seen in Fig. 3 [98]. With 1.91 wt% PPy coating, the microcapsules demonstrated 94 % photothermal storage efficiency under solar irradiation coupled with 231 J/g latent heat

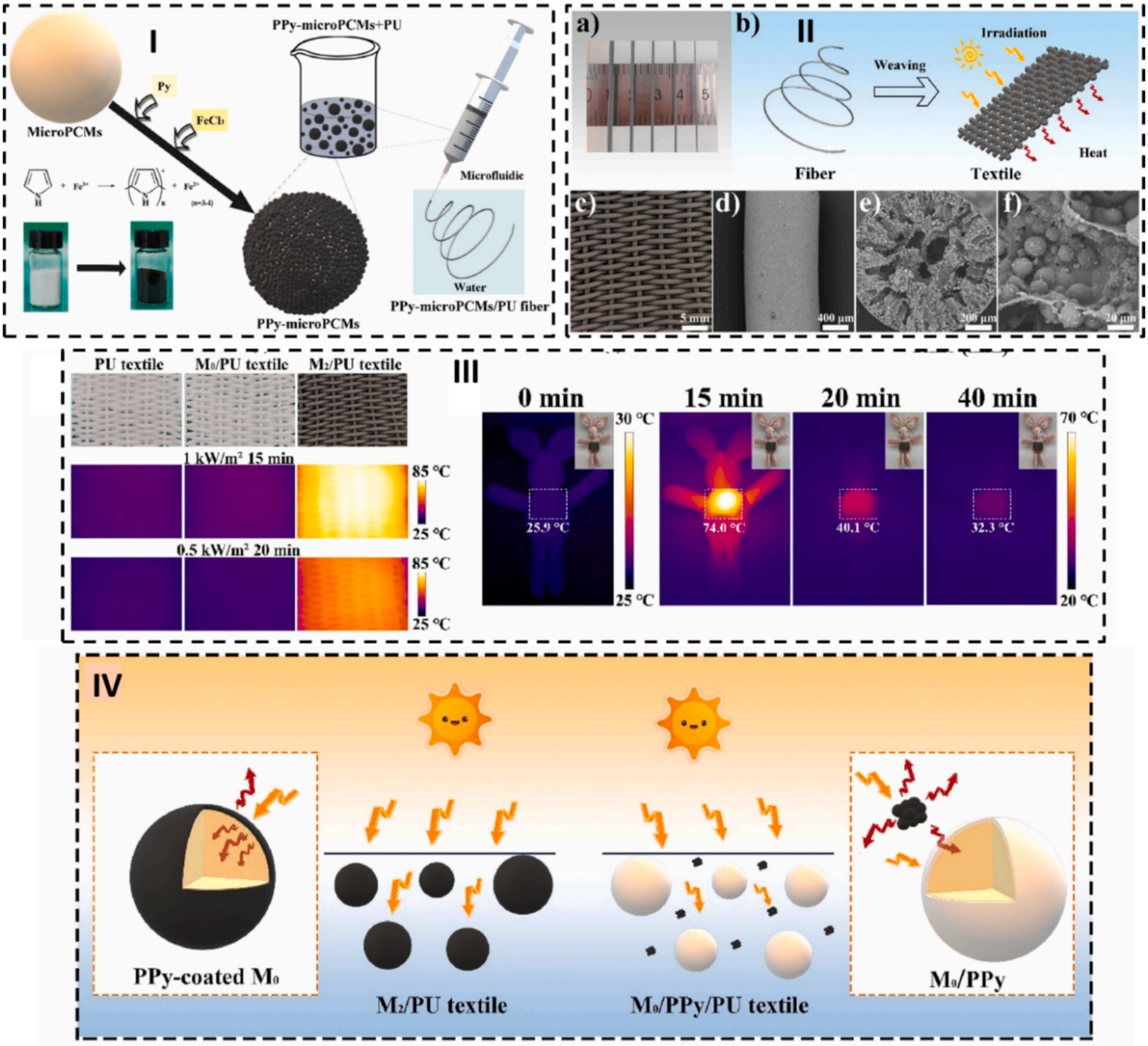

Fig. 3. I) The preparation route of PPy-modified PCM microcapsules-PU fibres. II) a) Image of the fibres with different thickness. b) Schematic representation of photothermal approach. c,e,f) Surface morphology of the material with different magnification. III) e) Optical and infrared images under irradiation and demonstration of thermotherapy enabled by the material. IV) Schematic presentation of photothermal efficiency, permission from [98].

absorbance. The microcapsules-PU composite with 79 wt% PCM loading retained its highly elastic nature, boasting an elongation of 365 %. These advantageous properties make the composite highly promising for applications related to solar energy. In another study, the top-down approach was applied by in-situ polymerization of PPy on electrospun PCM textiles consisting of decanoic acid and polyamide (PA) 11 [7]. The PPy-coated textile showed 28.89 S/m electrical conductivity and 113 J/g melting enthalpy. The textiles demonstrated great electro- and photothermal conversion and storage capabilities, indicating potential usage for wearable and protective systems.

According to the literature tabulated in Table 2, bottom-up approach appears to be the most used method for integrating PPy with PCMs. In this context, we adopt the term “aerogels” loosely to encompass a variety of light-weight materials produced by different drying methods, including cryogels (freeze drying) and xerogels (ambient pressure drying), in addition to traditional aerogels. This generalized use simplifies references across the different types of materials. Utilizing bottom-up approach, PPy aerogels were synthesized by inducing ionic surfactant-oleic acid interactions as support carrier for paraffin. The incorporation of oleic acid significantly enhanced the uptake of paraffin by the PPy aerogel [100], highlighting its potential for energy-saving applications, antistatic functionality, and advancing clean production. Specifically, with 7/1 pyrrole monomer to oleic acid molar ratio, the paraffin loading reached 60 %, exceeding that of PPy aerogel without surfactant by 117 %. The resistivity of the developed material was approximately 40 Ω·cm, meeting the required resistivity of conductive materials. In another study, n-octacosane was loaded in PPy-modified cellulose nanofibril scaffolds [99], resulting in 239–258 J/g energy storage and solar-thermal conversion capabilities suitable for practical utilization and storage of solar energy. Increased PPy content in the composition notably enhanced photothermal conversion efficiency under simulated sunlight. Fatty acids were also impregnated in sponge composed of sodium alginate and flax fiber coated with PPy [6], making it suitable for energy harvesting, storage, and management in electronics, buildings, transportation, and related fields. The PPy coated composite indicated 1.52 S/m electrical conductivity and 0.453 W/mK thermal conductivity. The porosity of PPy-sponge effectively impregnated the PCM, resulting in 154 J/g phase change enthalpy. In another study with a bottom-up approach, PPy was first deposited into CNT porous aerogel as the carrier matrix which was then infiltrated with paraffin [95], showcasing broad potential for thermal management applications. The composite demonstrated an enthalpy of 160.9 J/g and 0.64 W/m·K thermal conductivity, 2.56 folds larger than that of sole paraffin. This improvement is attributed to the PPy coating acting as a binder between the CNTs. Molecular dynamics simulations revealed that additional thermal transfer pathways are formed by the in-situ polymerization of PPy on CNT aerogels, which enhances heat transfer by synchronizing the vibrations of carbon atoms. With this approach, unidirectional heat transfer was achieved in a wood-PCM composite through in-situ PPy polymerization, depicted in Fig. 4. Boron nitride and PPy were integrated into the wood via electrostatic assembly which resulted in 0.76 W/mK thermal conductivity value, greater than that of neat PEG (0.177 W/mK) [101]. These features make the composite PCM suitable for applications in the field of solar-to-thermal energy management.

4.2. Polyaniline (PAni)-enhanced PCMs

As a semi-flexible rod polymer, PAni is a vastly researched CP. Due to its versatility, thermal stability, and unique electrical properties (conductivity 0.01–100 S/cm), it has become a key component in the realm of CPs [43,86]. It is widely employed in thermoelectric devices, photocatalysis, batteries and supercapacitors [122]. Its popularity further stems from its cost-effectiveness, non-toxicity, synthesis simplicity, and water insolubility [108].

When integrated into PCMs, PAni enhances thermal conductivity due to its intrinsic electrical conductivity and ability to form conductive networks within the PCM matrix. It is also compatible with PCMs like paraffin wax [107]. The improvement arises from conductive chains formed by PAni particles, which act as heat bridges, reducing thermal resistance and enabling faster heat distribution. However, the enhancement depends on PAni concentration; for instance, Janumala et al. [112] observed a 40 % conductivity increase at 1 wt%, while higher concentrations showed diminishing effects.

Encapsulation approach appears to be a more popular method of fusing PAni with PCMs in scientific literature over two other methods, as tabulated in Table 2. Hierarchical microcapsules functionalized with PAni and CNTs were developed to address the challenge of deteriorating electrochemical performance in supercapacitors under high temperatures caused by exothermic redox reactions during charge–discharge cycles [113]. As shown in Fig. 5, emulsion-templated interfacial polycondensation was used to create a layer-by-layer shell structure with SiO_2 around the n-docosane core followed by an electrochemically active layer of PAni/CNTs formed through in-situ oxidation polymerization. The resulting microcapsules demonstrated temperature self-regulation with 140 J/g latent heat enthalpy. At high temperatures, the composite exhibited superior supercapacitor performance compared to control samples with no PCM as shown in Fig. 5 b. This improvement stems from the thermal regulation by the PCM core. Furthermore, the synergy between PAni and CNTs enhanced pseudocapacitive behavior as well as charging and discharging cycle stability by improving electrical conductivity in the active surface layer. The study introduced smart self-thermoregulatory electrodes using CP-enhanced PCMs for high-performance supercapacitors across a broader temperature range.

PAni was employed as a support media for shape-stabilization of paraffin, yielding a highly thermally conductive shape stable PCM [107]. While increasing PAni in the composite enhanced thermal conductivity of paraffin, 20 % PAni exhibited suitable leakage-preventive properties with 180 J/g latent heat enthalpy and 65.45 % increase in thermal conductivity (0.55 W/mK) [107]. The paraffin-PAni composite exhibited superior latent heat reliability over that of sole paraffin upon a 3000 thermal cycling tests [84]. This makes it an attractive material for solar collectors, photovoltaic systems, and low-concentrated photovoltaic systems. In another study, erythritol was encapsulated with PAni through encapsulation method for thermal management application [109]. The erythritol-PAni capsules demonstrated leakage-resistant properties and a high enthalpy (up to 254 J/g). Integration of GNPs further improved thermal conductivity and photothermal conversion, with 8 wt% GNPs resulting 0.80 W/m·K thermal conductivity, a dramatic 196 % increase compared to capsules without GNPs. Moreover, PAni and GNPs acted as nucleating agents effectively mitigated the supercooling issue of erythritol, reducing it from approximately 100 °C to about 56 °C with just 2 wt% GNPs. In additional studies, PAni effectively resolved leakage issues and significantly mitigated the supercooling degree thanks to the nucleation sites provided by PAni and the heterogeneous nucleation of erythritol [110,123].

Fatty acids are another type of PCMs requiring leakage preventive and conductivity enhancement steps. In this context, stearic acid was effectively encapsulated within a PAni shell utilizing a self-assembly encapsulation technique shown in Fig. 6 [111]. PAni nuclei formed on the stearic acid surface, preserving its original morphological features, and culminating in a distinctive peony flower-like structure (Fig. 6 b). The maximum mass fraction of stearic acid reached 62 wt% without any leakage from the capsules after melting. Secondary doping with carboxyl groups of the PCM capsules (Fig. 6 a) resulted in a significant enhancement of electrical conductivity, increasing from 0.397 S/cm to 0.704 S/cm. Such PAni-enhanced fatty acids, with their electrical conductivity, high heat storage, and thermal stability, show great potential for thermal energy storage, antistatic textiles, and other thermal applications. In another study, in situ polymerization was used to encapsulate palmitic acid with PAni without the use of emulsifiers or doping agents [124]. The successful formation of spherical PCM capsules occurred by initiating polymerization above PCM`s melting point, using moderate oxidant concentrations, and gradually completing the polymerization at lower temperatures without adding any acid dopant. Fig. 6 c represents a bottom-up approach for the preparation of PCM loaded MXene/PAni aerogel [115]. The PAni/MXene framework exhibited strong solar absorption and conversion. When combined with PEG, the composite demonstrated high solar response and thermal energy storage, with latent heat values up to 151 J/g and over 95.29 % solar-to-thermal conversion efficiency.

4.3. Poly(3,4-ethylenedioxythiophene) (PEDOT)-enhanced PCMs

Poly(3,4-ethylenedioxythiophene) (PEDOT) is a CP frequently doped with poly(styrenesulfonate) (PSS) for its water stability enhancement. This compound is extensively utilized in solar cells, electrostatic coatings, organic electrodes, and wearable devices, thanks to its exceptional conductivity, transparency, flexibility, and ability to be processed in water. For instance, when integrated with textiles, PEDOT:PSS significantly enhances their conductivity [29].

As an effective CP, PEDOT:PSS can enhance the thermal conductivity of PCMs by forming interconnected conductive networks within the matrix, facilitating efficient heat transfer through phonon and electron movement [49]. Conformational changes in PEDOT chains through solvent treatments can further improve the overal conductivity by promoting alignment and connectivity of conductive domains [125]. Additionally, doping PEDOT:PSS with acids increases charge carrier concentrations and optimizes its morphology, further improving thermal transport properties [126]. Although studies on PEDOT-enhanced PCMs are less extensive compared to PPy and PAni, these mechanisms indicate that incorporating PEDOT:PSS into PCMs could enhance their thermal conductivity and overall effectiveness for thermal applications.

The top-down method was used for dip-coating of PEDOT:PSS on an electrospun textile made from lauric acid (LA) PCM and PU modified

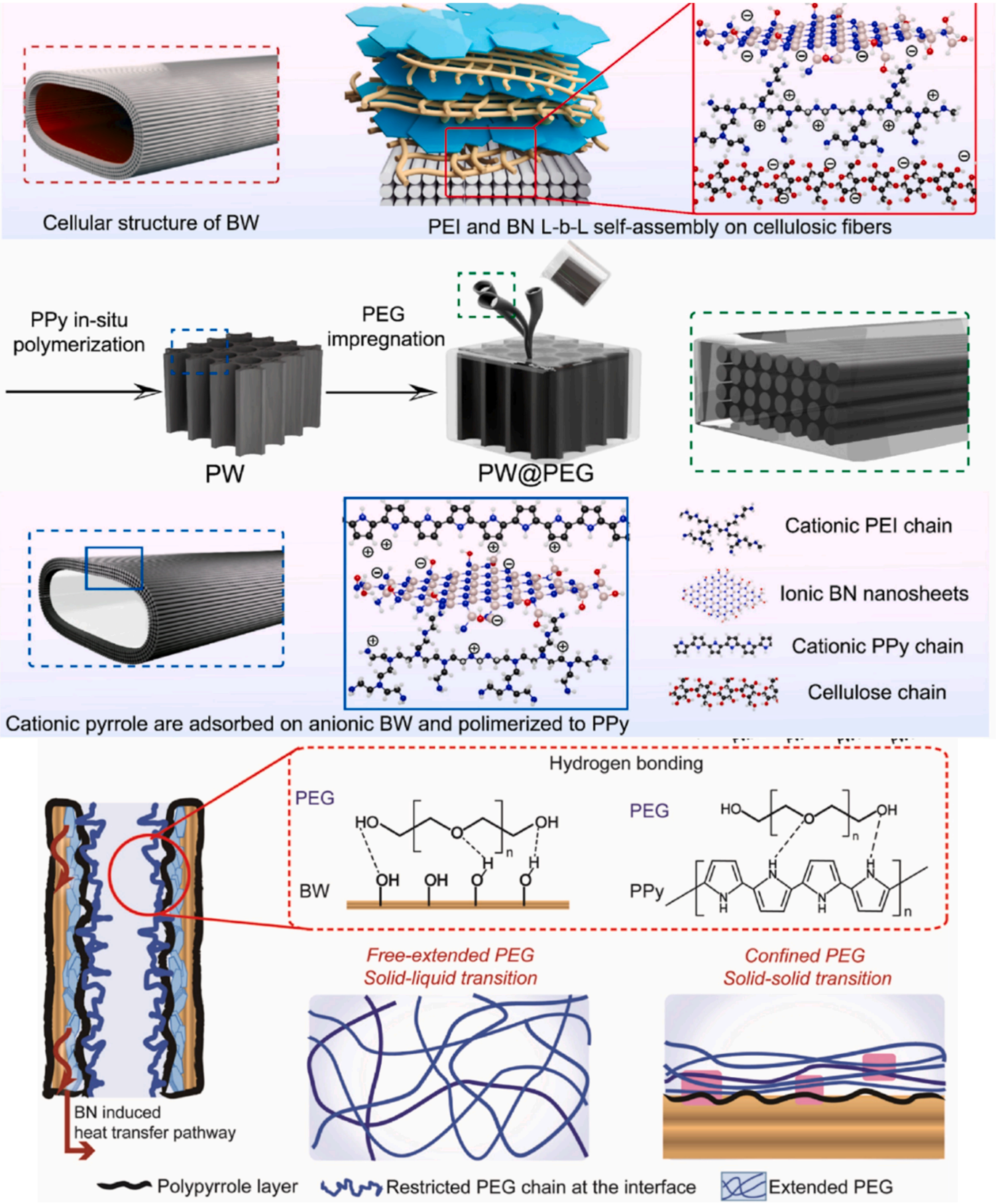

Fig. 4. Schematic illustration of bottom-up approach used for the preparation of PEG loaded PPy-wood, permission from [101].

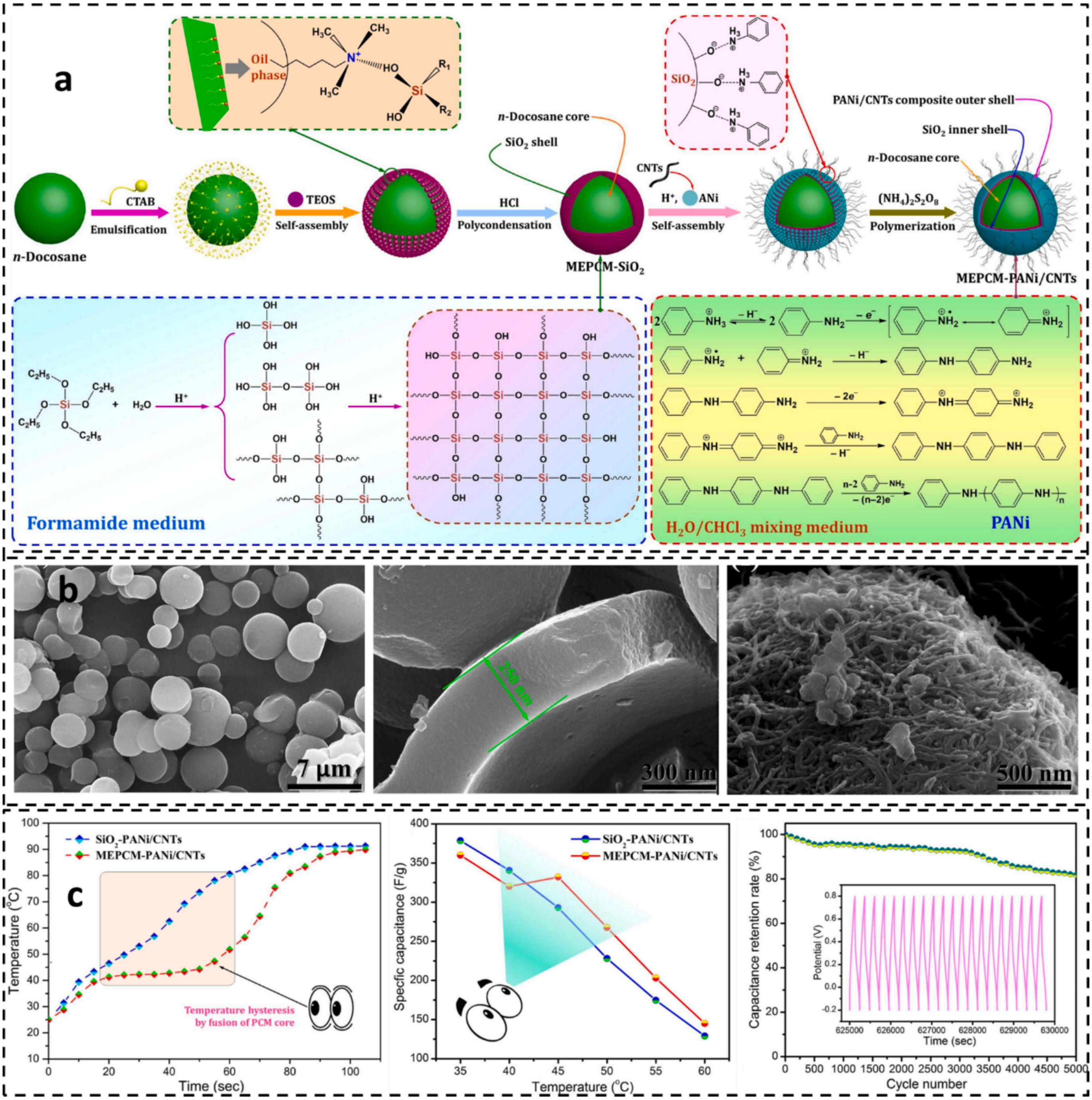

Fig. 5. A) Synthetic and reaction procedures for the PCM-PAni/CNTs microelectrode. b) SEM images of the microcapsules, their shell and surface structures. C) Phase change observation during heating process (left), specific capacitance versus temperature under a 1.0 A/g current density (middle), capacitance retention versus cycle number and inserted representative GCD curves at 45 °C and 2.0 A/g current density (right), permission from [113].

with MWCNTs, (Fig. 7), which resulted in enhanced conductivity, stretchability and thermal properties [116]. By varying MWCNTs and LA amounts, adjustable temperatures and enthalpy values up to 124.3 J/g were achieved. The composite fibers responded to electrical, infrared, and sunlight stimuli, with 83 % solar conversion efficiency. As stretchable conductors for powering a LED lamp, they exhibited proper reversibility, washability, and shape stability, making them ideal for conductive wearables and smart protective fabrics. The bottom-up method was employed to coat graphene oxide/graphene nanosheets aerogels with PEDOT:PSS, followed by vacuum-filling with PEG to form phase change composites [118]. The composites showed 211 J/g melting enthalpy and 1.12 W/m K thermal conductivity. When incorporated into photo-thermoelectric generators, they enabled constant output for over 25 min after 5 min of light exposure. Notably, the total energy output after switching off the light (8.4 V and 9.6 mW) surpassed that of light exposure period (2.7 V and 4.4 mW), offering photo-thermoelectric generators with effective function in emergencies. In another study, the PEDOT:PSS/MXene/PEG hybrids, with thermoregulation and electromagnetic shielding abilities, were developed through vacuum infiltration [120]. The PEDOT:PSS/MXene scaffold influenced PEG crystallization and induced denser stacking of PEG crystals. The composite showed high photo-thermal and electro-thermal efficiency, with effective electromagnetic shielding (29.8 dB) at just 1.22 wt% framework content. This solution is promising for photo-

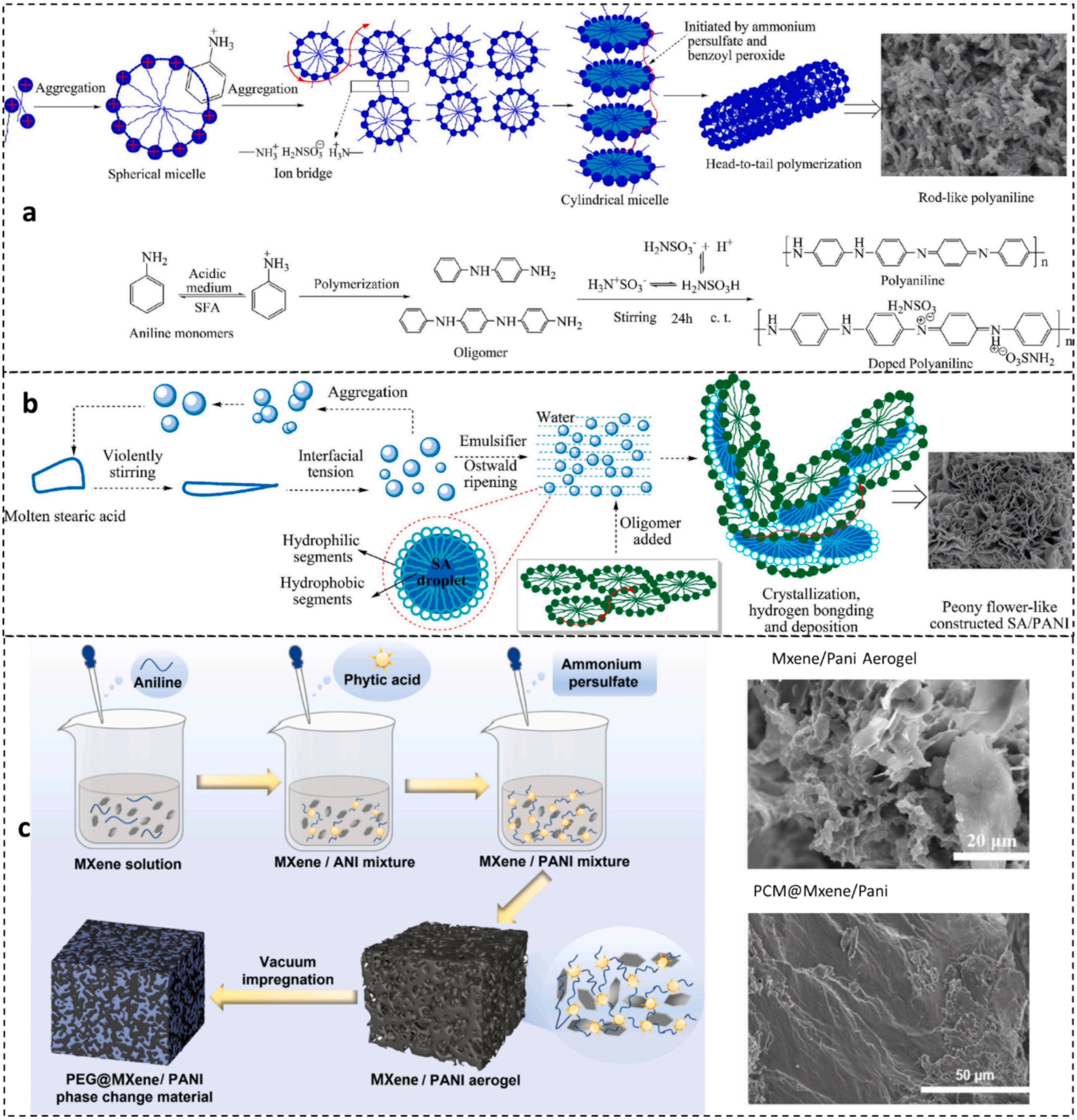

Fig. 6. A) Doping mechanism of PAni and b) the encapsulation of stearic acid with PAni, reprinting permission from [111]. c) Schematic illustration of bottom-up approach for the preparation of PEG@MXene/PAni aerogel, reprinting permission from [115].

thermal converters, heat buffers, and electromagnetic shielding in microelectronics. Fig. 7 is a representation of bottom-up and top-down methods used for incorporating PEDOT:PSS with PCM in aerogels and textiles.

Fig. 8 shows the encapsulation process of n-eicosane PCM with a SiO_2, a subsequent electroactive layer coating of PEDOT/AuNPs and a final immobilization of catalase on the surface [117]. The developed catalase-PCM microcapsules (CAT-MEPCM) utilize PCM's thermal regulation property for ultrasensitive H_2O_2 biosensing under harsh conditions with over 130 J/g latent heat. It was then employed for the modification of a glassy carbon electrode (GCE) in an electrochemical biosensor. The biosensor detected H_2O_2 with high sensitivity due to the conductive PEDOT/AuNPs layer together with covalently bonded catalase (Fig. 8 II, III). This integration enhances biocatalytic activity and electrochemical response by regulating the microenvironmental temperature, preventing catalase denaturation. Compared to traditional H_2O_2 biosensors, the developed sensor indicated higher sensitivity (42.85 μA/mM cm) and a lower detection limit (3.56 μM) at 50 °C, enabling precise H_2O_2 sensing in food and biological sources across a broad range of temperature. As shown in Fig. 8 II, the conductive PEDOT/AuNPs layer lowered the electron transfer resistance (R) (~20 folds) in the Au/PEDOT-MEPCM-enhanced GCE. Fig. 8 III indicates

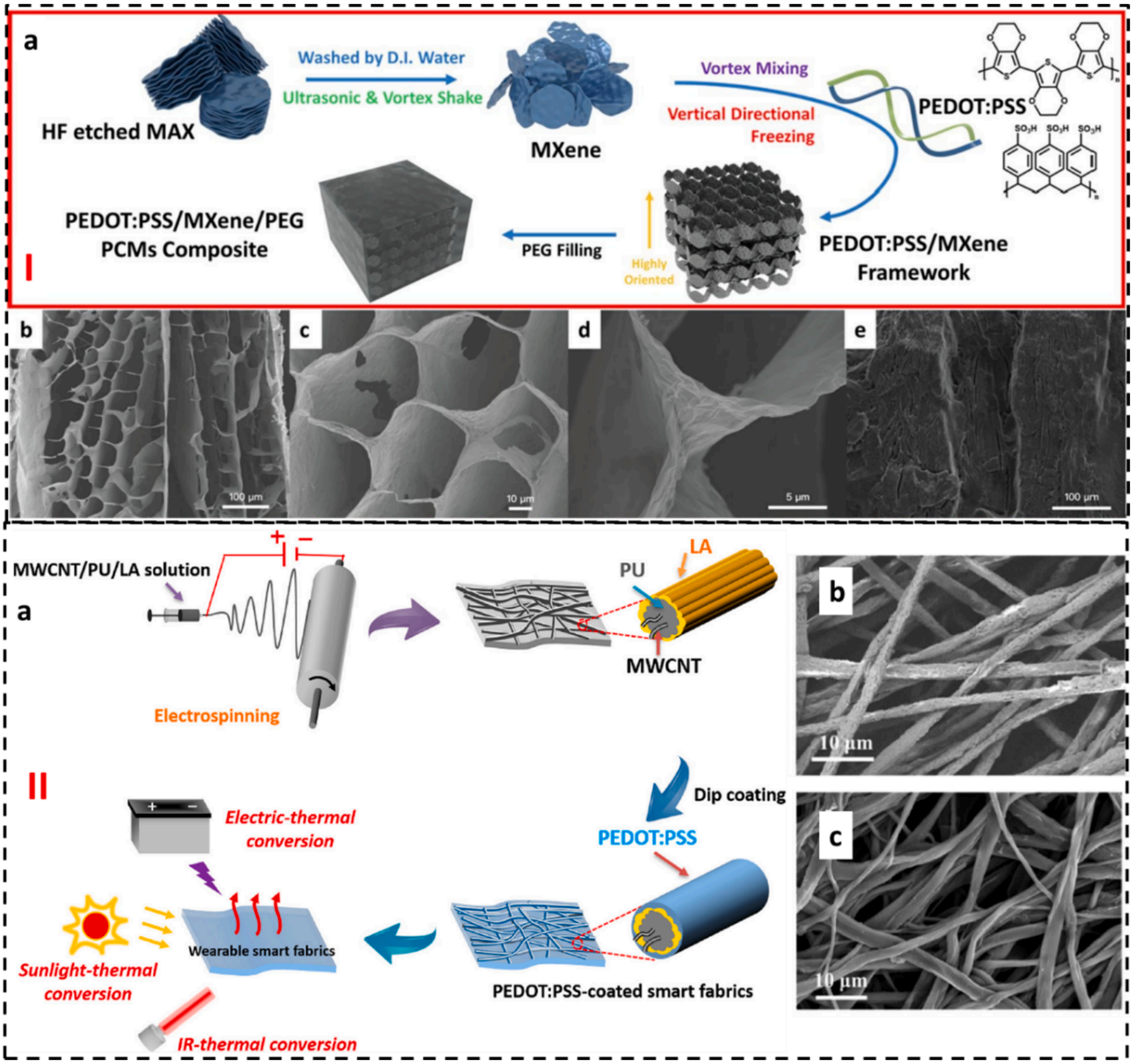

Fig. 7. I) a: Bottom-up approach used in the fabrication of PEDOT:PSS/MXene/PEG material. (b-d) SEM images of PEDOT:PSS/MXene scafold and (e) PEDOT:PSS/MXene/PEG composite, permission from [120]. II) a: Top-down approach used in the fabrication of PEDOT:PSS-coated MWCNTs/LA/PU modified textiles and b,c) corresponding SEM images before and after coating, reprinting permission from [116].

distinct redox peaks in the CV diagrams of sole GCE and GCEs enhanced with microcapsule. Due to insufficient conductivity, the SiO$_2$-MEPCM-enhanced and sole GCEs demonstrate weak peak currents, while the Au/PEDOT-MEPCM-enhanced GCE provides larger redox peak currents due to the conductive PEDOT/AuNPs layer enhancing electron transfer. The CP-PCM enhanced biosensor can be applied for the sensitive detection and precise measurement of H_2O_2 levels in food and biological samples in various temperatures.

New self-powered, multifunctional sensors incorporating CPs can offer a solution to the challenge of integrating temperature and pressure functions in portable and wearable dual-mode sensors. For example, a multifunctional sensor was developed by coating a melamine sponge with PEDOT and carboxylated CNT that could independently detect temperature and pressure changes, converting them to voltage and resistance signals via Seebeck and piezoresistive effects [127]. They provided high sensitivity due to 35.9 μV/K Seebeck coefficient and −3.35 % kPa^{-1} pressure sensitivity. Integrating PCMs with such dual-mode sensor can enhance its thermal regulation, sensitivity, energy storage, and multifunctionality, attracting even more interest for advanced applications including soft robots, electronic skin, and health monitoring.

The use of PCMs for personal thermal management offers better individual control and suitability for outdoor activities like sports, military, specialized jobs, and healthcare. This growing need has led to the rise of personal thermoregulation solutions, focusing on delivering the ideal thermal conditions for individual comfort [128]. Utilizing the top-down approach, ultra-stretchable PCM fibers were developed for thermal management in personal healthcare by embedding PCM microcapsules in elastic PU via wet spinning (Fig. 9) [49]. PEDOT:PSS bonded CNTs/graphene into a 3D network along the fibers, resulting in high sensitivity to multiple stimuli and efficient carrier transfer. The conductive fibers (1.96 × 10^4 S/m) maintained strong responsiveness, showing Joule heating and photothermal effects (Fig. 9 II) even under deformation, enabling self-powered thermal management. This approach offers enhanced flexibility and efficiency in regulating temperature, addressing critical needs in personal comfort and well-being applications.

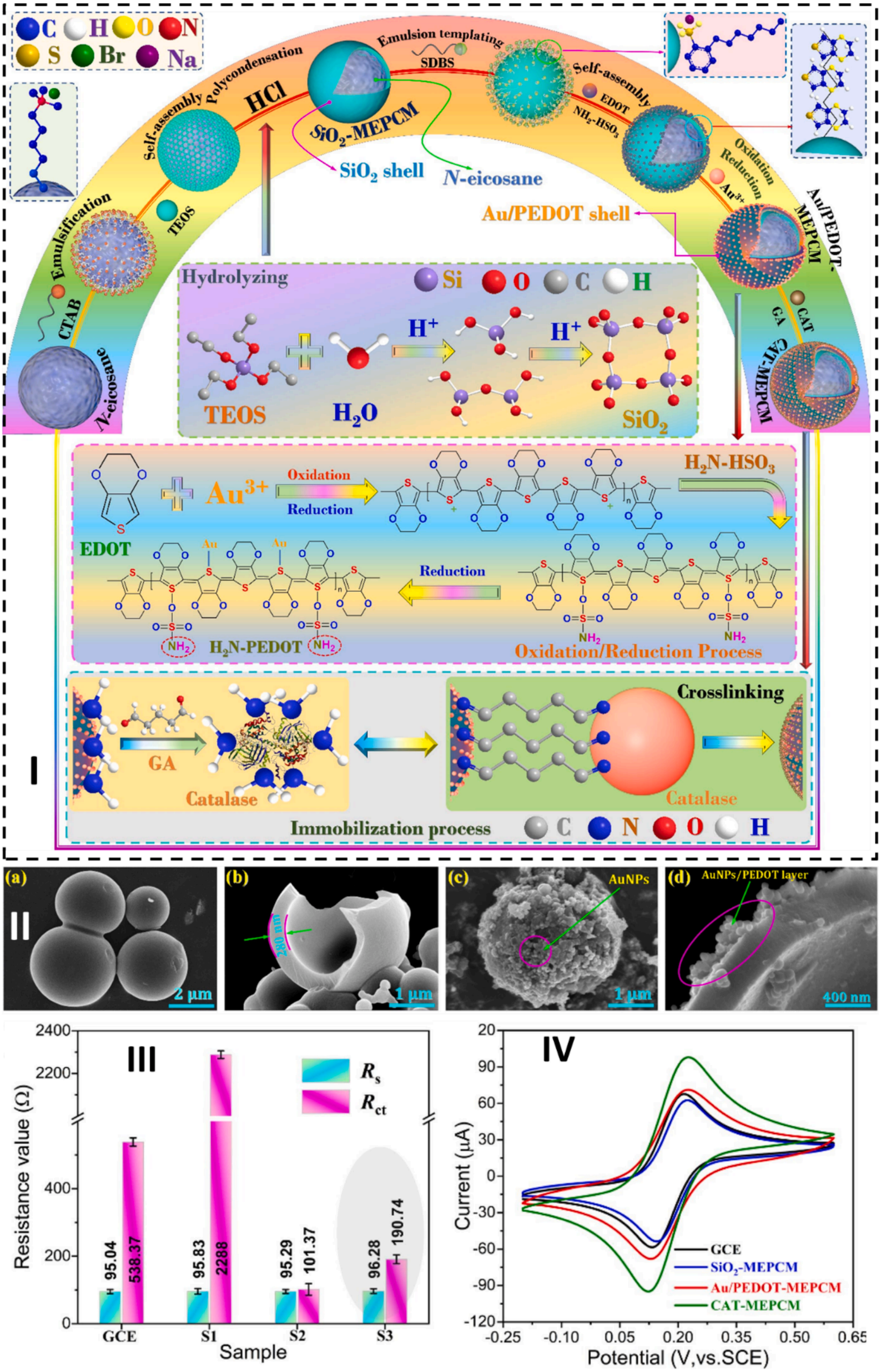

Fig. 8. I) Schematic presentation of preparation and reaction procedures of CAT-MEPCM. II) SEM pictures related to (a, b) SiO_2-MEPCM and (c, d) Au/PEDOT-MEPCM. III) Resistance performance related to GCE and the GCEs containing SiO_2-MEPCM (S1), Au/ PEDOT-MEPCM (S2), and CAT-MEPCM (S3). IV) CV graphs of GCE and refined GCEs in the $K_4Fe\ (CN)_6)$/KCl electrolyte, permission from [117].

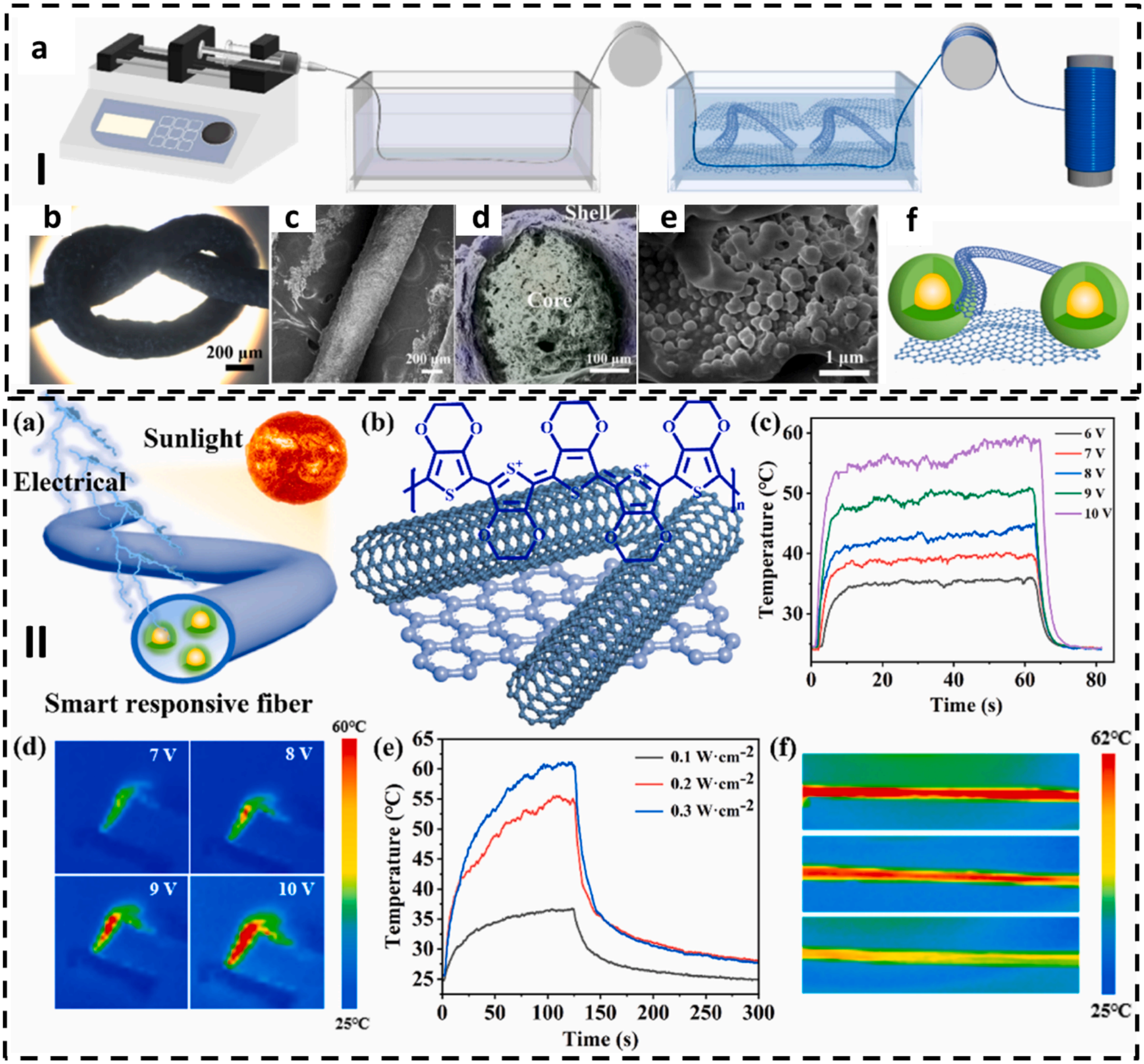

Fig. 9. I) a) The preparation route of multi-stimuli responsive PCM fibers. b) Optical microscopic image and (b, c, d) SEM images of the fiber (including the cross-section, the surface network consisted of CNTs/graphene connecting micro-PCM), and e) the corresponding schematic diagram. II) (a) The schematic representation of the fiber's reaction to different stimuli. (b) An illustration of CNTs` π-π interfacial interactions, graphene and π-conjugated structure of PEDOT. (c, d) The plots of fiber's reaction to electrical stimuli and their related IR images. (e, f) The plots of fiber's reaction to light stimuli and the related IR images, reprinting permission from [49].

5. Nano-dimensionality

Nano-dimensional additives (0D, 1D, 2D, 3D) of different types are increasingly explored for the conductivity enrichment of PCMs. Incorporating nano-dimensional additives into PCMs enhances heat transfer, overcoming the inherent issue of low thermal conductivity. This enables faster charging and discharging cycles, elevating the overall efficiency of energy systems. The effectiveness of thermal enhancement is influenced by the type, concentration, and uniform distribution of the additives. The role of such nanoparticles, mainly of carbon and metal nature, in thermal enchantment of PCMs has been covered in previous reviews [36,37]. However, the techniques utilized in the incorporation of nano-dimensional additives into PCMs have not been fully covered and missing from the literature. Different methods can be used to combine nano-additives with PCMs to enhance thermal conductivity. Melt blending [129] (or direct dispersion) where nano-additives are physically mixed into molten PCMs, is a simple and cost-effective technique applicable to all dimensional nano-additives (0D, 1D, 2D, 3D). Table 3 shows a variety of nanoparticles that have been mixed with PCM melts for thermal conductivity enhancement. However, achieving a uniform dispersion, especially for larger 1D and 2D structures like CNTs and graphene, can be challenging with this approach. Solution blending is also widely used for all dimensions, offering better dispersion by dissolving the additives and the PCMs in appropriate solvents and then blending them together, although solvent removal and uniformity can be problematic [37]. More complex methods like in-situ synthesis[130]

Table 3
Nano-dimensional additives used for PCMs` thermal conductivity enhancement and their incorporation techniques.

Nano-additives		Impact	Integration methods with PCMs	Ref.
0D	Metal nanoparticles	Effective enhancement of thermal conductivity for example, silver, gold, copper, and aluminium nanoparticles	Melt blending, Solution blending, In-situ synthesis	[142–147]
	Carbon dots	Offer good thermal properties, heat distribution capacity, and chemical stability		
	Metal oxide nanoparticles	Are used for their thermal conductivity and chemical inertness, for example, alumina, silica, and titania nanoparticles		
1D	Carbon nanotubes (CNTs)	Provide high aspect ratio and strong thermal conduction capacity, forming conductive heat transfer pathways	Melt blending, Solution blending Encapsulation Vacuum infiltration	[148–150]
	Metal nanowires	Improve thermal conductivity with their elongated structure and high conductivity For example, silver or copper nanowires		
	Carbon nanofibers	Generate continuous thermal pathways due to their elongated fibrous structure with a high aspect ratio		
2D	Graphene and graphene oxide	Highly efficient in enhancing heat transfer due to their strong heat transfer properties and high surface area	Melt blending, Exfoliation, Template-assisted synthesis, Vacuum impregnation	[151–153]
	Boron nitride nanosheets (BNNS)	Provide high thermal conductivity and electrical insulation		
	MXenes	Offer excellent thermal conductivity Especially explored in applications where other additives like graphene or metal particles may not be suitable		
3D	Metal-organic frameworks (MOFs)	Provide highly porous structure and thermal conductivity enhancement	Template-assisted synthesis, In-situ synthesis, Vacuum impregnation	[154,155]
	Graphene aerogels	Enable thermal bridges as a low-density 3D network of graphene with high thermal conductivity		
	Carbon-based foams	Enhance thermal conduction by developing continuous conductive pathways		

allow nano-additives to be formed directly in the PCM, improving dispersion but requiring precise control. Encapsulation [131] is another method that can be applied to encapsulate PCMs with conductive nanoparticles further enhancing stability and compatibility, although it can increase costs and processing complexity. Exfoliation [132] is specifically used for 2D materials like graphene and boron nitride nanosheets, while template-assisted synthesis and vacuum impregnation [133] are suited for 3D nano-additives, creating 3D structured conductive networks. The 3D nanostructures, like graphene foams or aerogels, are often synthesized using templates (e.g., scaffolds or molds). After synthesis, these structures are combined with PCMs through vacuum-assisted infiltration or impregnation capillary of the PCM melt into the porous 3D network. Common challenges across these methods include agglomeration, stability over time, and increased viscosity.

CPs can also be synthesized as nano-dimensional structures. Nano-dimensional CPs offer distinct advantages against their bulk counterparts, including enhanced surface areas, shorter charge/mass transport pathways, and enhanced mechanical properties for better strain tolerance [29]. These features are advantageous for energy conversion and storage, electronics, actuators, and sensors. Of particular interest are CP with continuous 3D porous frameworks, which further amplify the advantages of these materials for high-performance energy systems. The synthesis of nanoscale CPs requires modifications to the traditional methods used for bulk CPs. Although the core principles of polymerization—such as chemical, electrochemical, or oxidative polymerization—are similar, additional modifications are introduced to control the size and morphology at the nanoscale [134]. Various strategies have been developed for fabricating nano-dimensional CPs, including 1D structures like nanorods, nanowires, and nanotubes. These structures can be synthesized using either template-based methods (hard, soft, and wire templates) or template-free techniques (electrospinning, self-assembly, and nanosized patterning). Other techniques like interfacial polymerization and electrochemical approaches also yield diverse CP morphologies, including 3D porous structures [134,135]. However, despite their potential, there is limited research on using nanoscale CPs in hybridization with PCMs to enhance thermal conductivity, an area largely unexplored compared to the conventional use of bulk CPs. For example, 0D CPs nanoparticles, which are in spherical or near spherical forms and often synthesized through emulsion polymerization [136], can act as discrete nano-additives to elevate the thermal conductivity of PCMs. The 1D CPs in the forms of nanofibers, nanotubes or nanowires [134] provide a high aspect ratio and elevated surface area for enhanced heat transfer. The 2D CPs nanosheets [137] like PAni nanosheets [138] offer a large surface area advantageous in thermal management. Finally, 3D CPs networks, for example PPy-based 3D aerogels[139] and PEDOT foams [140,141], are porous frameworks that provide continuous pathways for heat transfer. Table 3 discusses suitable methods that can be adopted for integrating nano-dimensional CPs with various PCMs. For example, in-situ synthesis or melt blending may be more suitable for 0D/1D CPs, while infiltration/impregnation may perform the best for 3D aerogels and foams of nano-CPs.

6. Limitations and outlook

While CP-enhanced PCMs offer significant opportunities for applications in thermal management, energy systems, electronics, and smart wearables, their development is still restricted by limitations and unresolved research gaps. As tabulated in Table 2, research has predominantly focused on the use of PPy, PAni, and PEDOT for the conductivity enhancement of PCMs, leaving other CPs such as poly(p-phenylene) (PPP) [156], polythiophene (PT) [157], and polyacetylene [158] unexplored in this field.

Based on the evaluation of recent literature, three methods have been identified for integrating CPs with PCMs: bottom-up, top-down, and encapsulation approaches, as discussed in Section 4. Each approach offers distinct advantages and disadvantages, detailed in Table 1. It is important to compare the effectiveness of these three methods for different CPs and determine the most suitable approach for specific PCMs and target applications. Notably, the top-down approach remains relatively underexplored for CP-enhanced PCM synthesis, despite its potential to impart unique material properties. This method allows surface coating and functionalization with CPs, enhancing surface properties like hydrophobicity [7]. This post-stabilization modification offers flexibility in tuning the thermal and electrical properties of the final product. Additionally, CP coatings can improve composite durability during repeated thermal cycles. However, surface coating may primarily influence surface properties without significantly affecting the bulk material, limiting its overall enhancement. Coatings may also wear off or delaminate over time [79], especially in scenarios involving mechanical abrasion or mismatched thermal expansion coefficients with the substrate. However various strategies can be explored to overcome the challenges. For instance, implementing adhesion-promoting strategies can effectively address the delamination issues. These strategies may include surface modification, interface manipulation, or chemical bonding agents to enhance the adhesion between the coating and substrate, thereby improving durability and performance in such applications [79]. Further research is therefore needed to address these challenges and research opportunities.

Nanoscale CPs have remained unexplored in this domain despite their distinct advantages over bulk CPs, including higher conductivity, greater surface areas and shorter charge and mass transport pathways. For instance, the size-controlled synthesis of PPP nanoparticles was investigated [156], resulting various undoped-PPP (uPPP) morphologies and particle sizes ranging from ~ 30 to ~ 120 nm. Doping further increased conductivity, peaking at 74.34 S cm^{-1} with a doping agent-to-monomer ratio of 50:1. In another study, PPy nanoparticles with precisely controlled sizes were synthesized using a chemical oxidation polymerization method [159]. The study revealed that the surface area and charge carrier concentration significantly influence the capacitance performance of PPy nanomaterials in supercapacitor applications. Despite their effectiveness and potential, such nano-sized CPs yet to be explored for PCM enhancement.

Interfacial thermal resistance between nano-additives is also a major obstacle to improving thermal conductivity. While combining various enhancement strategies holds potential for higher conductivity, addressing interfacial resistance requires innovative structural designs. For instance, manipulating interfaces has been shown to improve thermal transport management [160]. Introducing two-dimensional material layers, such as MoS_2 or WS_2, between the PCM and its substrate can significantly reduce the energy required for phase transitions by confining energy within the PCM layer, effectively mitigating interfacial resistance [161]. Another effective approach involves the development of superlattice structures, which localize heat within the PCM's active region, reducing switching energy and improving phase change properties by minimizing interfacial resistance [162]. While such advancements highlight the critical role of structural innovations in overcoming the challenges associated with interfacial thermal resistance in nano-enhanced PCMs, similar approaches could also be incorporated in CP-enhanced PCMs.

More theoretical and experimental investigations are required to evaluate hybridization of CPs with other nano-additives effectively. Artificial intelligence (AI) and machine learning (ML) can facilitate this process by optimizing concentration ratios of hybrid CP nano-additives for maximum conductivity. While AI and ML-assisted tools have gained significant momentum in optimizing PCM for various applications [163–165], their potential remains underutilized in the development of CP-enhanced PCMs. Determining the optimal loading ratios of integrated CPs nano-additives for maximum conductivity enhancement typically relies on experimental methods, suggesting a potential for modeling approaches. Improving the dispersion and compatibility of CPs and integrating large domain 2D additives into 3D structures can enhance thermal conductivity for various PCM applications. Understanding the quantitative relationship between 3D network structures and conductivity is crucial for optimizing composite PCMs, and scalable production techniques are essential for their commercial application.

The thermal conductivity of nano-enhanced PCMs in their liquid state remains underexplored, with most studies focusing on the solid phase. High thermal conductivity in both states is crucial for PCM effectiveness, as it aids heat dissipation during solidification. However, liquid-phase thermal conductivity is generally lower than in the solid state, as observed with MWCNT-enhanced paraffin wax [166] and silicon dioxide-myristic acid PCMs [167], where structural disorder in the liquid reduces conductivity. The thermal conductivity of single-walled carbon nanotube (SWCNT)-enhanced paraffin was also studied in solid and liquid states. With just 0.25 wt% SWCNT, a 250 % increase was observed in the solid state and 10 % in the liquid state [168]. The conductivity contrast between states grew with higher SWCNT loading and was more significant than with exfoliated graphite nanoplatelets. Leong et al. [169] highlighted that natural convection suppression further limits heat transfer in the liquid phase. More experimental and numerical studies are therefore needed to better understand and optimize liquid-phase thermal conductivity for energy storage applications, specially related to CP-enhanced PCMs.

7. Summary

This review examines the most studied CPs and their role in enhancing the thermal and electrical conductivity of PCMs, comparing CPs with other commonly used conductive additives, such as carbon and metal nanoparticles. Focusing on energy efficiency, the review discusses the advantages, synthesis methods, and utilization of CPs in diverse energy-related fields. It highlights their applications across domains such as thermal management, energy systems, electronics, and wearables. Additionally, it provides a fundamental overview of electrical and thermal conduction in polymers and PCMs, as well as the effect of nano-dimensionality on conductivity enhancement.

Improving the conductivity of PCMs by integrating conductive additives requires a comprehensive understanding of the underlying mechanisms and influencing factors. Conductive additives like CPs are essential for addressing the low thermal and electrical conductivity of PCMs. Key factors such as structure, incorporation mechanisms, additive concentration, and the formation of continuous 3D conductive networks significantly impact conductivity enhancement. CPs that establish continuous phonon pathways can reduce interfacial and thermal resistance. Combining CPs with nano-additives of various dimensions can achieve higher conductivity at lower loadings, as dual nano-additive networks further minimize thermal resistance. Moreover, forming 3D-structured CP coatings on support matrices, such as carbon materials and graphene aerogels, effectively enhances PCM conductivity due to their intrinsic conductive 3D networks, which eliminate thermal contact resistance. CPs also enable multifunctional properties in PCMs, including Joule heating, electro-thermal conversion, and energy charging and storage through their electrical conductivity.

CP-enhanced PCMs show improved functionality and thermal conductivity due to the intrinsic electrical conductivity of CPs and their ability to form continuous conductive networks of within the PCM. This improvement stems from creating CP networks that facilitate efficient heat transfer during phase transitions, boosting the composite's overall thermal conductivity. Commonly studied CPs for this purpose include PPy, PAni, and PEDOT. Three synthesis strategies—bottom-up, top-down, and encapsulation—have been classified for integrating CPs with PCMs. Most studies have used the bottom-up approach, while the top-down method remains relatively underexplored despite its potential to impart unique properties, such as surface functionalization with CPs, enhancing surface characteristics like hydrophobicity and electrical conductivity.

Despite their potential and advantageous properties, such as high surface area and charge carrier mobility, nanosized CPs have been underutilized in PCM enhancement. Interfacial thermal resistance has been identified as a major obstacle, which can be addressed through innovative structural designs and interface manipulation. AI and ML tools could play a significant role in optimizing hybrid CP nano-additives for maximum conductivity. Additionally, further research is needed to understand the impact of CPs on PCMs in their liquid state. These efforts will address current research gaps and expand the applications of CP-enhanced PCMs across various energy-related fields.

CRediT authorship contribution statement

Maryam R. Yazdani McCord: Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Ari Seppälä:** Writing – review & editing, Validation, Resources, Investigation, Funding acquisition, Conceptualization. **Mahdi Pourakbari Kasmaei:** Writing – review & editing, Validation. **Julie B. Zimmerman:** Writing – review & editing, Validation, Conceptualization. **Orlando J. Rojas:** Writing – review & editing, Validation, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Maryam Roza Yazdani McCord reports financial support was provided by Research Council of Finland. Maryam Roza Yazdani McCord reports financial support was provided by Business Finland. Ari Seppala reports financial support was provided by Research Council of Finland. Orlando J. Rojas reports financial support was provided by Canada Excellence Research Chair Program. Orlando J. Rojas reports financial support was provided by Canada Foundation for Innovation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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