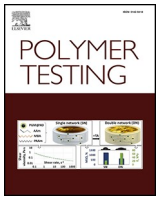
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A comprehensive review of research on organic-based phase-change thermal interface materials for thermal management of electric devices: Methods, performance, and applications

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ABSTRACT

Efficient thermal management is crucial for the optimal performance and longevity of electronic devices. With continuous advancements in technology and the ongoing miniaturization of electronic components, the requirement for effective heat-dissipation materials has become increasingly imperative. In this regard, organic phase-change materials (PCMs) have emerged as promising candidates for thermal management applications due to their unique properties. This paper explores the utilization of organic PCM-based heat dissipation materials for electronic devices, aiming to enhance their thermal performance and reliability. Through a comprehensive review of recent research and developments in this field, the paper discusses the fundamental principles underlying the thermal properties of organic PCMs and their suitability for electronic applications. The paper also discusses the challenges and opportunities associated with the integration of organic PCM-based materials into electronic devices, emphasizing the importance of addressing issues such as material compatibility, durability, and scalability. Overall, this review provides valuable insights into the utilization of organic PCM-based materials as efficient heat dissipation solutions for electronic devices, contributing to the advancement of thermal management technologies in the electronics industry.

1. Introduction

Electronic devices have become an indispensable part of modern life, permeating various aspects of daily activities and industrial operations. From smartphones [1–3] and laptops [4–6] to sophisticated electronic systems used in aerospace [7–9] and automotive [10–12] applications, the demand for compact, powerful, and energy-efficient devices continues to escalate [13,14]. However, the relentless pursuit of miniaturization and performance enhancement for electronic devices has led to significant challenges in managing the heat generated during operation [15,16]. Excessive heat accumulation not only compromises the performance and reliability of electronic components but also poses safety risks and limits further technological advancements [17,18] (see Fig. 1).

Effective thermal management is paramount for addressing these challenges and unlocking the full potential of electronic devices. Traditionally, metallic heat sinks and fans have been employed to dissipate heat away from electronic components through conduction and convection mechanisms [19,20]. While these methods have been relatively effective for conventional electronic systems, they face limitations in handling the escalating heat flux densities and spatial constraints imposed by miniaturized and densely packed electronic assemblies [21,22]. This has created a growing need for innovative heat dissipation materials capable of efficiently managing thermal loads in compact electronic devices [23,24].

Organic phase-change materials (PCMs) have emerged as promising candidates for addressing the thermal management challenges associated with electronic devices [25–27]. Unlike traditional heat sink materials, which rely on the conduction of heat through solid media, organic PCMs leverage the latent heat absorption and release mechanisms associated with phase transitions [28–30]. These materials undergo reversible phase transitions between solid and liquid states within a specific temperature range, absorbing and releasing large amounts of latent heat in the process. This unique property endows organic PCMs with a high thermal energy storage capacity and enables them to regulate temperature variations effectively in electronic devices.

The utilization of organic PCM-based thermal interface material

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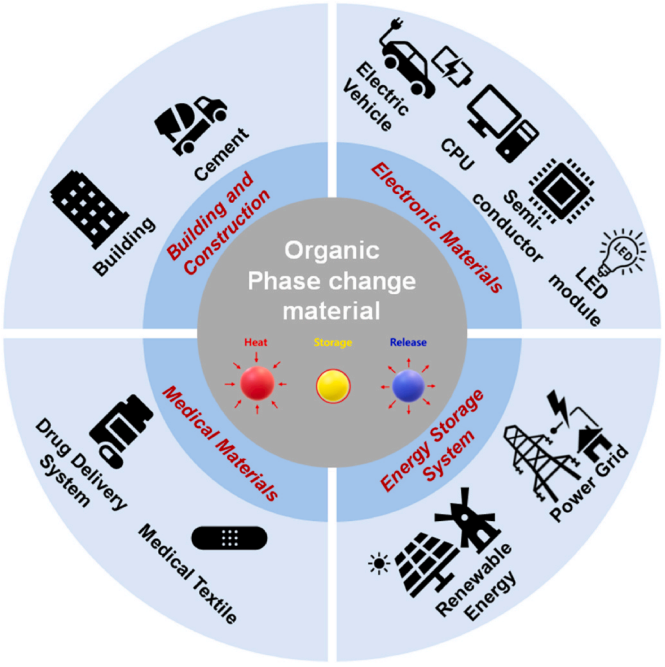


Fig. 1. Major application areas of PCMs.

(PCTIM) for electronic thermal management offers several advantages over conventional heat dissipation approaches [31–33]. First, organic PCTIM exhibit high thermal conductivity during the phase transition process, facilitating efficient heat transfer within electronic assemblies [34–36]. Moreover, the ability of organic PCMs to store and release thermal energy at constant temperatures ensures uniform temperature distributions across electronic components, mitigating hot spots and thermal stress [37,38]. Additionally, organic PCTIM are inherently lightweight and flexible; hence, they can be integrated into diverse electronic device architectures without compromising the form factor or performance [39,40].

Recent research has extensively explored the potential applications of organic PCMs in electronic devices. Researchers have investigated various aspects of organic PCM synthesis, characterization, and integration techniques to optimize the thermal performance and compatibility of organic PCMs with existing device designs. Strategies such as incorporating nanostructures and additives into organic PCM matrices have been employed to enhance the thermal conductivity and mechanical strength of PCMs, which has further expanded their applicability in demanding electronic applications.

Despite the promising prospects of organic PCMs, several challenges must be addressed to realize their widespread adoption in electronic devices. Material compatibility, durability, and long-term stability are critical considerations, particularly in harsh operating environments characterized by mechanical stresses, thermal cycling, and exposure to contaminants [41,42]. Furthermore, the scalability of manufacturing processes and the economic viability of organic PCM–based heat dissipation solutions warrant careful evaluation to ensure commercial feasibility [43,44].

In light of these considerations, this paper aims to provide a comprehensive overview of the current state of the art in organic PCM–based heat dissipation materials for electronic devices. By synthesizing and critically analyzing recent research findings and technological developments in this field, the paper seeks to elucidate the underlying principles, challenges, and opportunities associated with the integration of organic PCM–based materials into electronic devices. Through a systematic exploration of material properties, fabrication techniques, and potential applications, this paper endeavors to contribute to the advancement of thermal management technologies in the electronics industry, fostering innovation and sustainability in the design and manufacturing of electronic devices.

1.1. Application areas of PCMs

Due to their unique thermal properties and versatility, organic PCMs are gaining prominence across a wide array of industries. These materials, derived from organic compounds, undergo a phase transition from solid to liquid and vice versa, absorbing or releasing large amounts of latent heat in the process. This ability renders them highly effective in managing temperature fluctuations and therefore suitable for thermal energy storage applications. Below is a comprehensive list of the various fields where organic PCM-based thermal management materials are utilized:

- Building and construction: Organic PCM-based materials are extensively used in building envelopes, such as walls, ceilings, and roofs, to regulate indoor temperatures. By absorbing excess heat during the day and releasing it at night, they help maintain comfortable indoor environments, reduce energy consumption, and enhance the overall energy efficiency of buildings [45–49].
- Textiles and apparel: In the textile industry, organic PCM microcapsules or coatings are incorporated into fabrics to create thermoregulating clothing. Such garments offer enhanced comfort by actively responding to changes in environmental temperature, keeping the wearer cool in hot conditions and warm in cold weather [50–54].
- Automotive sector: Organic PCM-based materials are integrated into vehicle components such as seat cushions, headliners, and dashboard panels to provide localized temperature control, improving passenger comfort and energy efficiency [55–59].
- Electronics and electrical devices: The electronics industry benefits from the ability of organic PCMs to prevent overheating in electronic devices. By embedding PCM-based heat sinks or thermal interface materials into electronic gadgets, such as laptops, smartphones, and LED lights, thermal runaway risks are mitigated, which prolongs device lifespan and ensures optimal performance [60–65].
- Cold chain logistics: Organic PCM-based solutions play a crucial role in cold chain logistics for temperature-sensitive products, including pharmaceuticals, food, and vaccines. PCM-based packaging materials help maintain the desired temperature ranges during storage and transportation, safeguarding product quality and integrity throughout the supply chain [66–68].
- Energy storage systems: Organic PCMs serve as effective thermal energy storage media in various systems based on renewable energy, such as solar and wind power. By storing excess energy during peak production periods and releasing it in response to high demand, PCM-based energy storage systems contribute to grid stability and energy efficiency and facilitate the integration of renewable energy sources [69–73].
- Medical and healthcare sectors: Organic PCM-based materials find applications in medical devices, temperature-controlled shipping containers for medications, and therapeutic products such as cooling vests for patients with multiple sclerosis or fever management patches. By ensuring precise temperature control for the critical healthcare needs in such applications, organic PCM-based materials enhance patient comfort and treatment efficacy [74–76].

In summary, the versatile nature and thermal properties of organic PCM-based materials enable their widespread application across diverse industries, contributing to enhanced energy efficiency, environmental sustainability, and technological innovation.

1.2. Classification of PCMs

Organic PCMs encompass a diverse range of compounds with unique

thermal properties, which are valuable for thermal management applications. These materials can be classified into four main categories based on their chemical composition and molecular structure: paraffin, polyethylene glycol (PEG), fatty acid, and sugar alcohol PCMs. Each category exhibits distinct characteristics and offers distinct advantages, which are essential for various types of thermal energy storage and heat dissipation applications. Table 1 shows the characteristics of each type of representative organic PCM.

Paraffin PCMs are hydrocarbon compounds characterized by linear or branched alkane chains, derived primarily from petroleum sources [77–79]. The chain length directly influences the temperatures of melting of fusion. Increasing the carbon atom improves the properties (T_m and latent heat) of the paraffin as mentioned Table 1. Due to their high latent heat of fusion and wide range of phase transition temperatures, these versatile PCMs are suitable for different temperature control applications. Paraffin PCMs are typically available in various chain lengths, which allows for customization of their phase transition temperatures to suit specific requirements. Its chemical stability, non-degrading in melting/solidification cycles, storage potential of 200 J/g, and affordable with less susceptibility to corrosion and environmental pollution are the most expensive characteristics [102]. Moreover, their compatibility with polymer matrices enables the fabrication of composite materials with enhanced thermal properties. Owing to their high energy storage capacity and thermal stability, paraffin PCMs are widely used in applications such as building insulation, solar energy storage, and electronics cooling. However, paraffin has a few drawbacks of poor heat conductivity and noticeable volume change while phase changing.

PEG is used as the latent heat storage medium and the graphite, SiO2, bentonite, diatomite, carbon additives, sugar, epoxy resin etc. are utilized as a form stabilizer and properties enhancer. PEG is a type of organic PCM is suggested as heat energy storage material because of its high latent heat, corrosion resistance, congruent melting behavior, and suitable melting temperature. Since PEG available with various molecular weights and melting temperature shown in Tables 1 and it can be utilized in different engineering applications. However, less thermal conductivity and leakage during phase transformation hampers the utilization of polyethylene glycol [103].

Fatty acid PCMs are derived from natural sources such as vegetable oils and animal fats, consisting of carboxylic acid functional groups attached to alkyl chains [80–82]. This amphiphilic molecular structure of fatty acid exhibit good interfacial interaction with most functional fillers, moderate latent heats of fusion, and their phase transition temperatures can be tuned by modifying the fatty acid composition. The presence of polar functional groups in fatty acid PCMs facilitates hydrogen bonding and improves their compatibility with other materials, enhancing their performance in composite systems. These PCMs show favorable thermal properties compared to paraffins and are low-cost, non-toxic, and non-corrosive. Fatty acid PCMs are particularly suitable for applications requiring temperature regulation within a moderate temperature range, such as thermal comfort regulation in buildings, refrigeration, and food packaging. Additionally, their biodegradability and renewable nature make them environment-friendly alternatives to conventional PCMs.

Sugar alcohol PCMs, also known as polyols, are organic compounds derived from the hydrogenation of sugars [83–85]. These PCMs exhibit unique phase change behaviors attributable to their molecular structure, which includes multiple hydroxyl (–OH) functional groups. Sugar alcohol PCMs offer advantages such as high thermal stability, non-toxicity, and biodegradability; thus, they are suitable for various applications involving thermal energy storage and heat management. Due to their abundant hydroxyl groups, sugar alcohol PCMs can form extensive hydrogen bonding networks, which induce higher melting point and latent heat than other organic PCM such as paraffin, PEG, fatty acid. Moreover, the strong interaction by hydrogen bonding between adjacent molecules enhances their thermal conductivity and compatibility with other materials. As a result, sugar-alcohol PCM shows high intrinsic thermal conductivity (>0.5 W/mK) than most polymer matrix. These PCMs are employed in applications such as thermal regulation in textiles, pharmaceuticals, and controlled-release systems, where non-toxicity and biocompatibility are critical considerations.

In summary, the aforementioned classification of organic PCMs provides a framework for understanding their diverse properties and applications in thermal management. Each type of PCM offers unique advantages and can be tailored to specific temperature ranges and performance requirements, which could enable the development of efficient and sustainable heat dissipation solutions for various industries. However, as mentioned above, all types of organic PCM have leakage problems, so this is an important problem that must be solved first.

Table 1
Characteristics of substances according to classification of organic PCM.

Type	Material	Melting temperature (°C)	Latent heat (J/g)	Thermal conductivity (W/mK)
Paraffin	C19-C25	32–50.6	222–255	0.15–0.3
Polyethylene glycol (PEG)	PEG 1000-10000	33–65.9	140–171.6	0.19–0.32
Fatty acid	Lauric acid	43.2	178	0.21
	Myristic acid	54.4	204.5	0.22
	Palmitic acid	65.9	213	0.2
	Stearic acid	69.3	220	0.2
Sugar-alcohol	Xylitol	92	240	0.52
	Sorbitol	95	175	0.59
	Erythritol	121	390	0.5–0.7
	Mannitol	167	326	0.56

2. Technical challenges and solution strategies in the PCM filed

2.1. Characteristics and challenges related to PCM

Organic PCMs represent promising heat dissipation materials owing to their distinctive thermal properties. By virtue of their high latent heat capacity, these materials can absorb and release significant amounts of thermal energy during phase transitions. Additionally, organic PCMs offer tunable phase transition temperatures, which can be adjusted to match the demands of specific operating conditions. Furthermore, their compatibility with composite materials facilitates the fabrication of heat-dissipation solutions with improved mechanical strength and thermal conductivity.

However, several challenges hinder the widespread adoption of organic PCM-based heat dissipation materials. First, these materials often exhibit low thermal conductivity [86,87], which can limit heat transfer efficiency within electronic devices and other heat-sensitive systems. This issue must be addressed by devising strategies to enhance the thermal conductivity of PCMs without compromising their desirable properties.

Second, organic PCMs may possess weak mechanical properties [88,89], such as stiffness and tensile strength, which can limit their suitability for structural applications or hinder their integration into complex device architectures. Hence, improving the mechanical properties of organic PCMs is crucial for ensuring the integrity and durability of heat-dissipation materials under various operating conditions.

Another significant challenge associated with organic PCM-based heat dissipation materials is the risk of liquid leakage [90,91]. When organic PCMs transition into their liquid state, they may leak or seep out of the encapsulation material, leading to operational failures and potential safety hazards. Preventing such leakage necessitates the development of effective encapsulation techniques and PCMs that can

withstand mechanical stress, thermal cycling, and prolonged use while retaining their structural integrity.

Furthermore, the scalability and cost-effectiveness of manufacturing processes for organic PCM-based heat dissipation materials remain important considerations. Scaling up production processes while maintaining cost efficiency is essential to meet the growing demand for thermal management solutions across various industries. Developing scalable manufacturing techniques and optimizing material formulations can help address these challenges and promote the widespread adoption of organic PCM-based heat dissipation materials.

In summary, while organic PCM-based heat dissipation materials offer significant advantages for thermal management applications, addressing the low thermal conductivity, inadequate mechanical properties, and liquid leakage–related issues associated with such materials is imperative for realizing their full potential. Advancing research and development efforts in these areas will facilitate the production of organic PCM-based heat dissipation materials capable of providing efficient and reliable thermal management solutions for diverse applications.

2.2. Characteristics and challenges related to phase change thermal interface material (PCTIM)

Recently, many researchers have been exploring the use of PCM as either the matrix or additives in TIM composites for next-generation thermal management of electronic devices (see Fig. 3). This research primarily focuses on the endothermic reaction that occurs within the melting point temperature range of the PCM. Fig. 4 shows a representative thermal management mechanism of PCTIM. When the PCM or additives within the matrix are exposed to a heat source, the solid crystalline PCM begins to melt as it absorbs heat energy. This delays or prevents temperature rise of the heat source and PCM composite. Additionally, if the PCM composites exhibit high thermal conductivity, they can also facilitate effective thermal dissipation. As a result, PCTIM show great potential for achieving more stable thermal management compared to conventional TIMs. However, addressing the leakage issue of PCMs during phase transitions poses a significant challenge in fabricating high-performance PCTIM.

3. Strategies for fabrication of high-performance PCM composites

To address the challenges discussed earlier, diverse strategies based on physical and chemical methods have been developed. Physical methods focus on enhancing the material's structure to improve thermal conductivity, such as through the introduction of porous fillers, while chemical methods are intended to enhance mechanical properties through reactions with thermosetting or thermoplastic polymers. These strategies contribute to the designing of more efficient organic PCM-based heat dissipation materials (Table 2).

3.1. Physical methods

3.1.1. Introduction of porous structured fillers

Enhancing the thermal conductivity of organic PCM-based heat dissipation materials is a pivotal requirement for unlocking their full potential across a broad spectrum of applications. Amidst a myriad of techniques available, the strategic incorporation of porous fillers into the PCM matrix represents an exceptionally promising avenue, offering a multifaceted approach for addressing the pervasive challenge of low thermal conductivity. These porous fillers, ranging from expanded graphite (EG) to graphene and aerogels, exhibit intricate structural attributes that endow them with unparalleled capabilities for facilitating efficient heat transfer within the material [92,93].

Among the key properties of these fillers is their remarkably high surface area-to-volume ratio, which creates an intricate network of pathways through which thermal energy can propagate efficiently. By ensuring the uniform dispersion of these fillers throughout the PCM matrix, the material's thermal conductivity experiences a quantum leap with minimal degradation of its intrinsic phase change properties. Moreover, the porous morphology of these fillers assumes a pivotal role in accommodating PCM expansion during phase transitions, thereby mitigating mechanical stress and fortifying the material's overall thermal stability. Consequently, the leakage problem with PCMs is greatly suppressed.

Meticulous selection and optimization of porous filler materials can circumvent the low thermal conductivity of organic PCM-based heat dissipation materials, thereby enabling their adoption in thermal

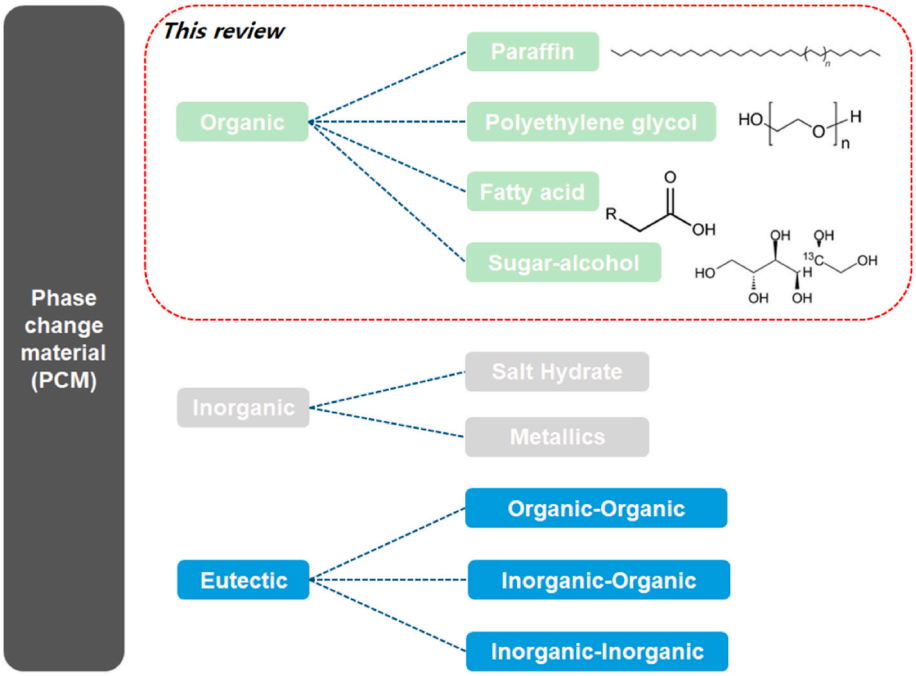


Fig. 2. Classification of PCMs and types of organic-based PCMs.

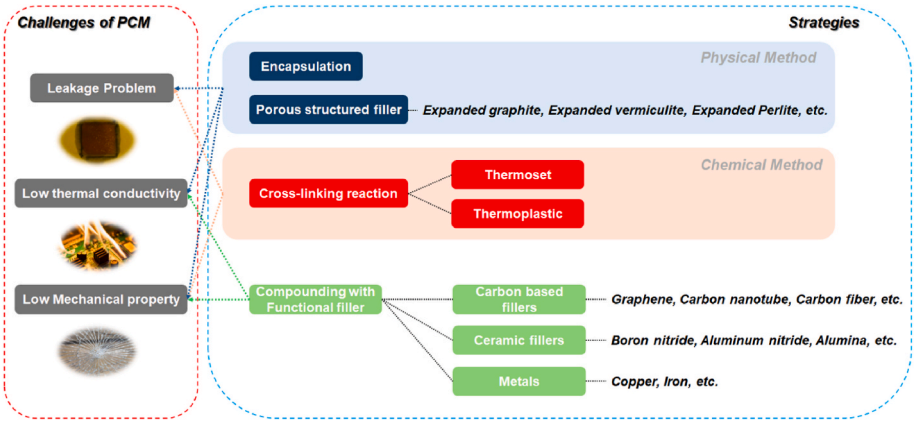


Fig. 3. Technical challenges with PCMs and strategies for fabricating high-performance PCM composites.

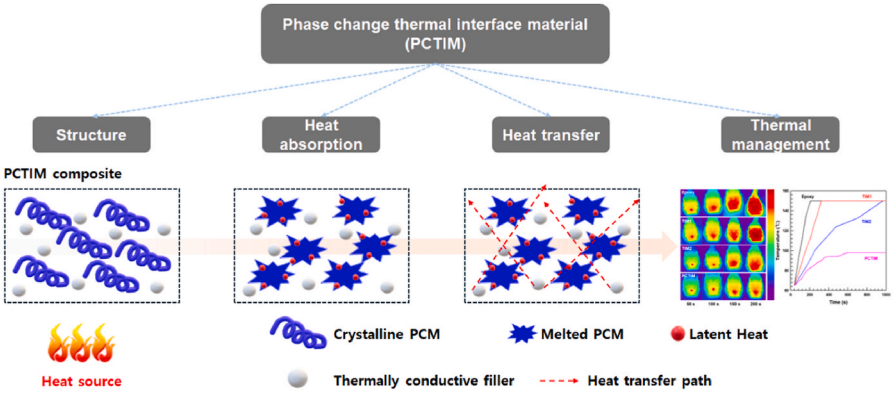


Fig. 4. Thermal management mechanism of PCTIM.

Table 2
Properties of PCM composites according to fabrication strategies.

Composite	Filler fraction	Strategy	Melting temperature (°C)	Latent heat (J/g)	Thermal conductivity (W/mK)	Ref.
PW/LMA/EG	LMA 4.55 wt% EG 9 wt%	Porous filler	70.08	225.09	5.842	[94]
PC-EG/(PW/TPP-3:1)	–	Porous filler	50.8	94.1	0.41	[95]
PS/CaCO3/HDA	–	Encapsulation	86	33.96	0.491	[96]
ESBGPs	–	Encapsulation	31.39	139.01	0.974	[97]
SABPA/EG/DBN	EG 5 wt%/DBN 50 wt%	Chemical method	60	93.51	7.21	[98]
ETBPA/BNCNF/BN	60 wt%	Chemical method	120	98.4	13.09	[99]
ETPLA/CF-OH/AlN	50 wt%	Chemical method	163	170.7	4.25	[100]

management applications across diverse sectors.

Zheng et al. have demonstrated the untapped potential of organic PCMs for efficient thermal energy storage and passive thermal management applications (Fig. 5). These materials boast attributes such as corrosion resistance, high heat storage capacity, and stable operating temperatures, which render them promising candidates for various thermal management tasks. However, their practical application is hindered by issues such as low thermal conductivity and susceptibility to leakage.

In addressing these challenges, the researchers introduced a groundbreaking approach for crafting high-performance shape-stable composite PCMs (CPCMs). This innovative method leverages paraffin wax (PW) as the energy storage material and EG as both a heat transfer enhancer and a supporting material. Notably, the incorporation of low-melting-point alloy (LMA) micro-particles, which have the same phase change temperature as PW, serves a dual purpose: augmenting latent heat capacity and establishing a hybrid three-dimensional thermal conduction network at the microscale, which ensures synergy with EG.

The resulting CPCMs exhibit remarkable attributes in terms of energy storage capacity, thermal conductivity, and thermal cycling stability. Notably, with optimal LMA and EG loadings, the thermal conductivity of ternary CPCMs can reach 5.842 W/mK, representing a staggering 16.4-fold increase relative to pure PW; this is achieved without compromising volumetric latent heat.

This pioneering work offers a novel and promising avenue for the development of CPCMs characterized by superior storage properties and rapid thermal response.

Zhang et al. reported on the prevalence of PCMs in thermal management strategies aimed at stabilizing temperatures and fortifying the fire safety of operational electronic devices (Fig. 6). PW is frequently utilized as a PCM owing to its commendable attributes, such as high latent heat, consistent performance, eco-friendly nature, and economical viability. However, the utility of PW is overshadowed by challenges such as susceptibility to leakage and heightened flammability concerns.

To overcome these limitations, the researchers devised an innovative method involving the creation of a porous framework using

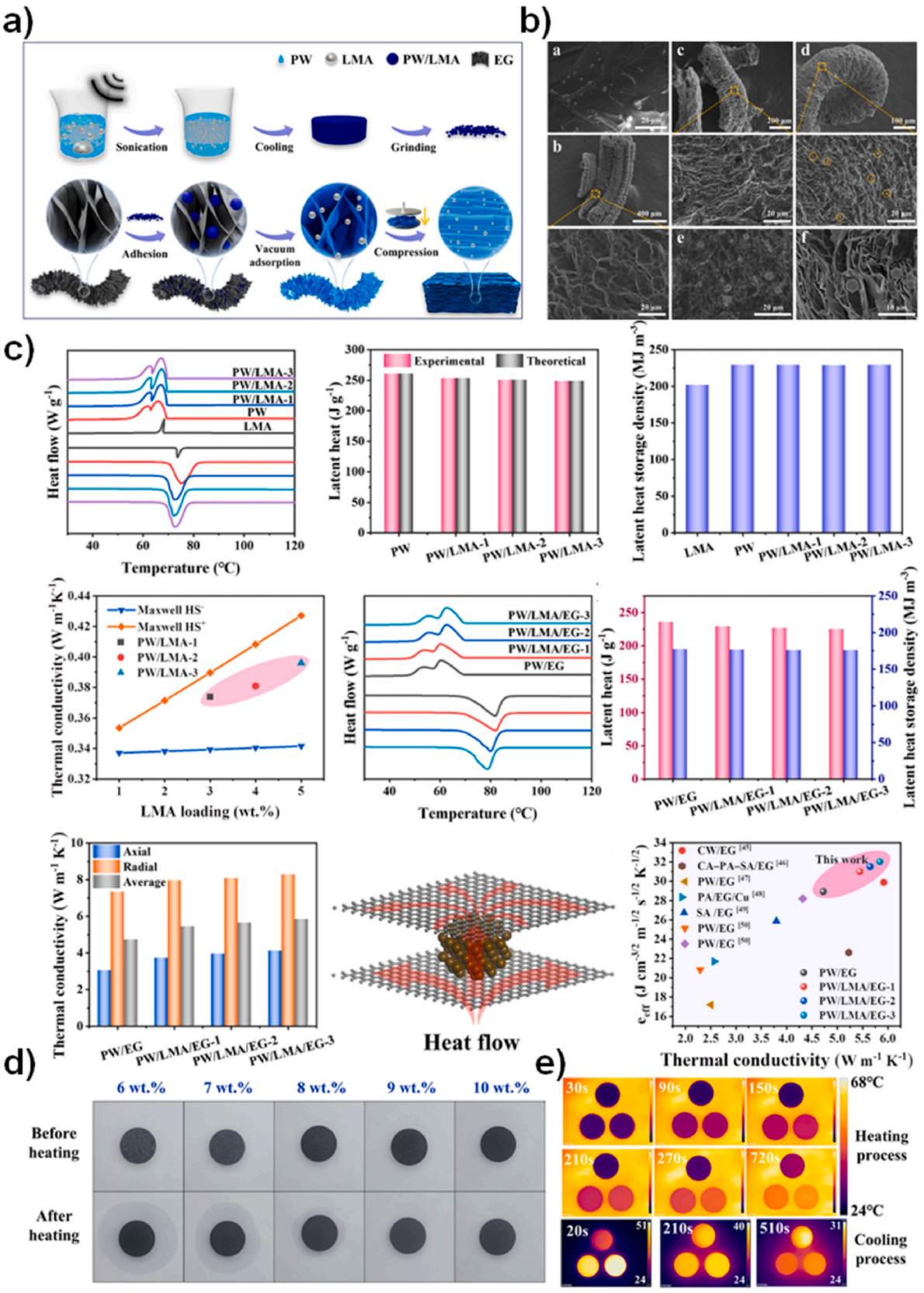


Fig. 5. (a) Schematic illustrating synthesis of PW/LMA CPCM and PW/LMA/EG CPCM. (b) scanning electron microscopy (SEM) images of PW composites. (c) Thermal properties of PW/LMA and PW/LMA/EG CPCMs: differential scanning calorimetry (DSC) curves, latent heat storage density, and thermal conductivity of LMA, PW, and PW/LMA CPCMs. DSC curves of PW/EG CPCM and PW/LMA/EG CPCMs. Latent heat of melting; latent heat storage density; and axial, radial, and average thermal conductivities of PW/EG CPCM and PW/LMA/EG CPCMs. Heat transfer path between LMA and adjacent graphite sheets. (d) Leakage-proofing test of PW/EG CPCMs. (e) Thermal responses of PW/LMA/EG-3 CPCM [94].

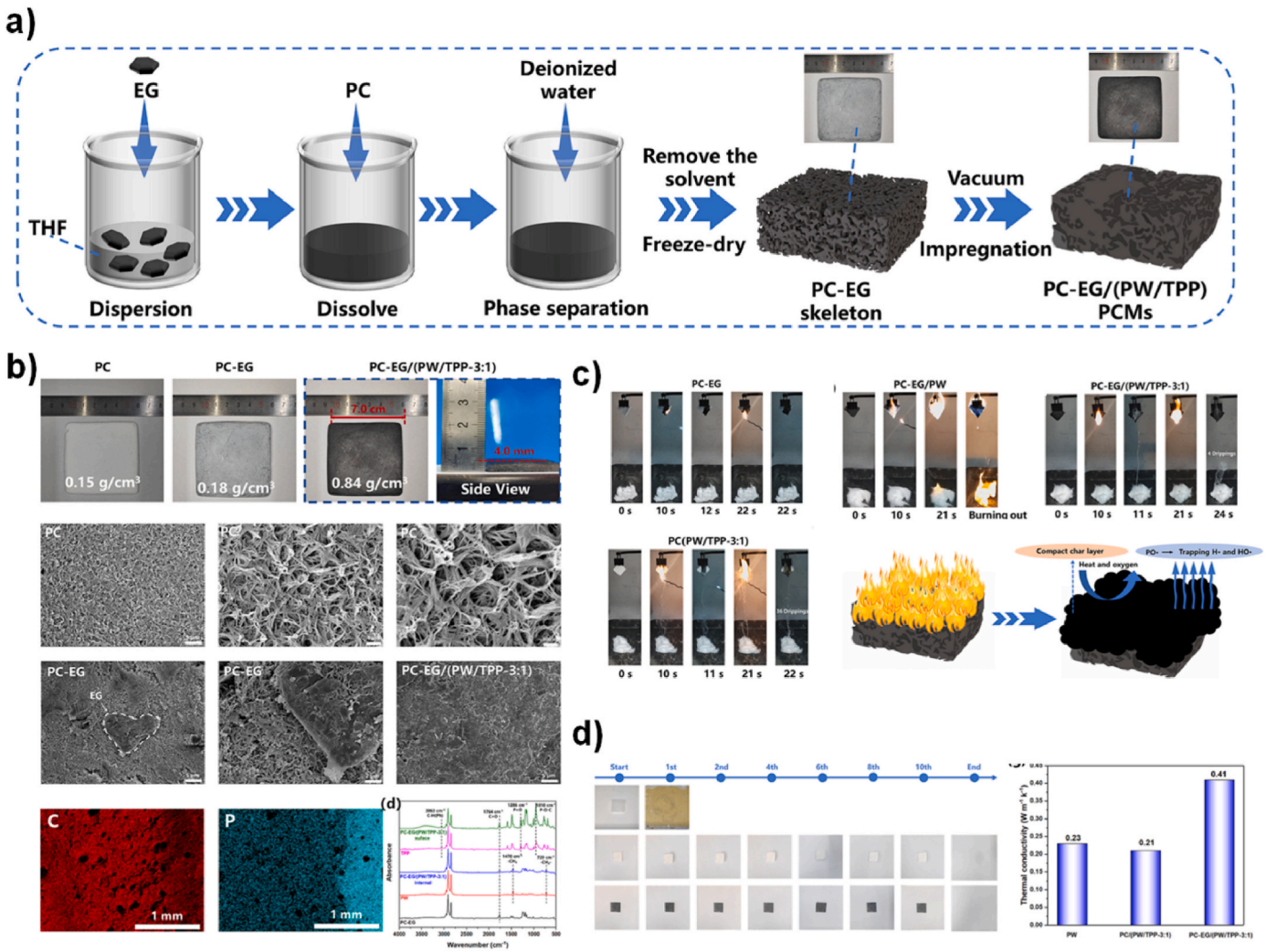


Fig. 6. (a) Preparation of PC-based PCMs. (b) Digital photographs of PC, PC-EG, and PC-EG/(PW/TPP-3:1); SEM images of PC, PC-EG, and PC-EG/(PW/TPP-3:1); EDS mapping of the surface of PC-EG/(PW/TPP-3:1); FTIR spectra of composites. (c) Vertical burning test of PC/EG skeleton; PC/EG/PW; PC/EG/(PW/TPP-3:1); PC/(PW/TPP-3:1); proposed flame-retardant mechanism. (d) Sample shape changes over 10 heating–cooling cycles; thermal conductivity of PW and PC-based PCMs [95].

polycarbonate (PC) and EG. This framework served as the foundation for the development of PCMs with enhanced flame retardancy and reduced PW leakage, achieved by encapsulating PW and triphenyl phosphate (TPP) within the PC-EG structure. Comprehensive assessments were conducted to evaluate the phase change behavior, fire safety, and thermal regulation capabilities of these PC-based PCMs.

The findings revealed that PC-based PCMs loaded with PW/TPP in a ratio of 3:1 exhibited a mere 0.26 % leakage rate and a latent heat of 94.1 J/g. Notably, compared with PC-EG/PW samples, the PC-EG/(PW/TPP-3:1) counterparts displayed substantial reductions in both peak heat release rate (58.8 %) and total heat release (39.1 %). Additionally, the inclusion of TPP doubled the char residue from 5.0 % to 10.0 %. Furthermore, the fabricated PCMs showcased remarkable thermal management performance in pouch-cell operation.

This study pioneers a groundbreaking approach leveraging the PC/EG framework to bolster flame retardancy and structural integrity in PCMs. It introduces a viable methodology for producing PC-based PCMs with exceptional flame retardancy, structural robustness, and high phase change efficiency, thereby showing immense potential for the advancement of next-generation thermal management materials.

3.1.2. *Encapsulation*

The reliability and longevity of organic PCM-based heat dissipation materials hinge on the efficacy of the encapsulation techniques designed to preclude liquid leakage and uphold material integrity. Encapsulation, in essence, entails enveloping PCM particles or droplets within impermeable barriers or coatings that prevent leakage of the liquid phase. Encapsulation methodologies range from microencapsulation to spray drying and sol–gel encapsulation, each offering distinct advantages in terms of uniformity, durability, and scalability.

Microencapsulation, for instance, represents the encapsulation of individual PCM droplets within microcapsules, which eliminates the risk of leakage. Additionally, spray drying disseminates PCM particles within a protective matrix, culminating in the formation of a robust shell that protects the material against mechanical and thermal stresses. Sol–gel encapsulation, on the other hand, involves the formation of a gel-like coating around PCM particles, which affords unparalleled adhesion and stability.

Furthermore, the exploration of cutting-edge encapsulation materials with self-healing or self-sealing properties represents a viable direction for devising more reliable and durable PCM-based heat dissipation solutions. The use of robust encapsulation methodologies tailored to the unique demands of specific applications can yield organic PCM-based heat dissipation materials that are immune to liquid leakage and can maintain their performance over protracted durations.

Jiang et al. assessed the merits of PCMs, which offer remarkable

energy storage density and maintain stable temperatures during phase transitions, thereby being ideal for applications in industrial waste heat recovery (Fig. 7).

The researchers employed hexadecylamine (HDA) as a PCM, alongside polystyrene (PS) and calcium carbonate ($CaCO_3$) as wall materials, to fabricate PS/$CaCO_3$/HDA microencapsulated PCMs (MEPCMs) through ultrasonic emulsification. After fabrication, the PCMs underwent comprehensive characterization and property assessment using various analytical techniques, such as transmission electron microscopy, SEM, X-ray diffraction analysis, X-ray photoelectron spectroscopy, Fourier infrared spectroscopy, thermogravimetry, and DSC.

The outcomes showcased the successful fabrication of PS/CaCO3/HDA MEPCMs exhibiting a microcapsule structure with a diameter of approximately 1.5 μm. Notably, the thermal conductivity of these MEPCMs was measured to be 0.491 W/(m·K), marking a significant 3.41-fold increase over pure HDA. Furthermore, the observed maximum latent heat of the PS/$CaCO_3$/HDA MEPCMs reached 33.96 kJ/kg, with no instances of leakage recorded.

These results underscore the stability, high thermal conductivity, and exceptional thermal storage capacity of PS/$CaCO_3$/HDA MEPCMs, particularly at lower temperatures. The findings from this study significantly elucidate the properties of MEPCMs and facilitate their development for diverse thermal energy storage applications.

Zou et al. highlighted the immense potential of PCMs in thermal management applications. Nonetheless, developing PCMs with sufficiently small dimensions and dependable thermal management capabilities tailored for regulating the temperature of miniature electronic components remains a persistent challenge (Fig. 8).

The researchers pioneered a method of granulating PCM gel particles (PCMGPs) as a viable solution. This inventive technique involves a two-stage process encompassing PCM droplet formation followed by sol–gel transition. The resultant PCMGPs boast micron-scale dimensions, rendering them apt for seamless integration into microelectronic components. Moreover, the infusion of boron nitride (BN) bolsters the thermophysical attributes of the PCMGPs further.

The PCMGPs exhibit a notable phase transition enthalpy of 139.01 J/g, coupled with a remarkable enhancement (175 %) in thermal conductivity to 0.9739 W/mK and exemplary form stability, without any leakage. The size congruence, dependable heat retention, and augmented heat transfer enable the use of these PCMGPs in optimizing the thermal conditions of a smartphone's CPU across diverse operational scenarios.

The findings reveal a substantial reduction in the CPU's operational temperatures and the mitigation of thermal shock under elevated workloads. Specifically, the CPU's operational temperatures decreased by 5.4 °C and 8 °C under low and high operational loads, respectively.

This micromachining methodology could be pivotal in micro-scale PCM preparation and heat conduction, facilitating the advancement of thermal management technologies.

3.2. Chemical methods

3.2.1. Reaction with thermosetting polymers

Chemical methodologies provide a multifaceted avenue for augmenting the mechanical properties and stability of organic PCM-based heat dissipation materials. By incorporating thermosetting polymers, such as epoxy resins or phenolic resins, into the PCM matrix to induce crosslinking reactions, a three-dimensional network structure can be generated. These interwoven networks serve as highly effective mechanical reinforcements, offering a pronounced enhancement in the

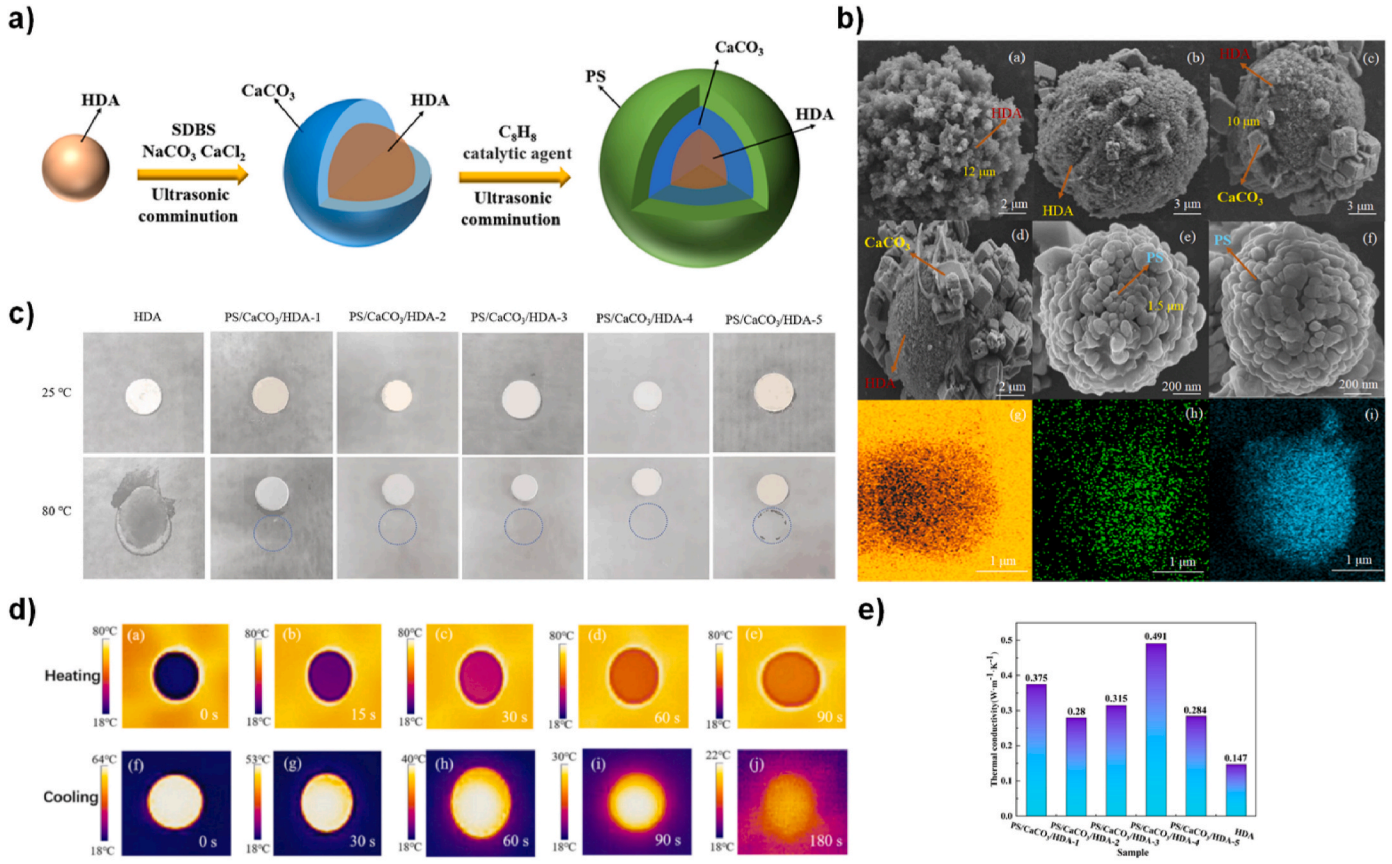


Fig. 7. (a) Flowchart of prepared PS/$CaCO_3$/HDA microcapsule. (b) SEM images of HDA, $CaCO_3$/HDA, and PS/$CaCO_3$/HDA, along with elemental distribution of C, N, and O in PS/$CaCO_3$/HDA. (c) Leakage experiment of HDA with PS/$CaCO_3$/HDA. (d) Infrared thermal images of PS/$CaCO_3$/HDA MEPCMs. (e) Thermal conductivity test of HDA and PS/$CaCO_3$/HAD [96].

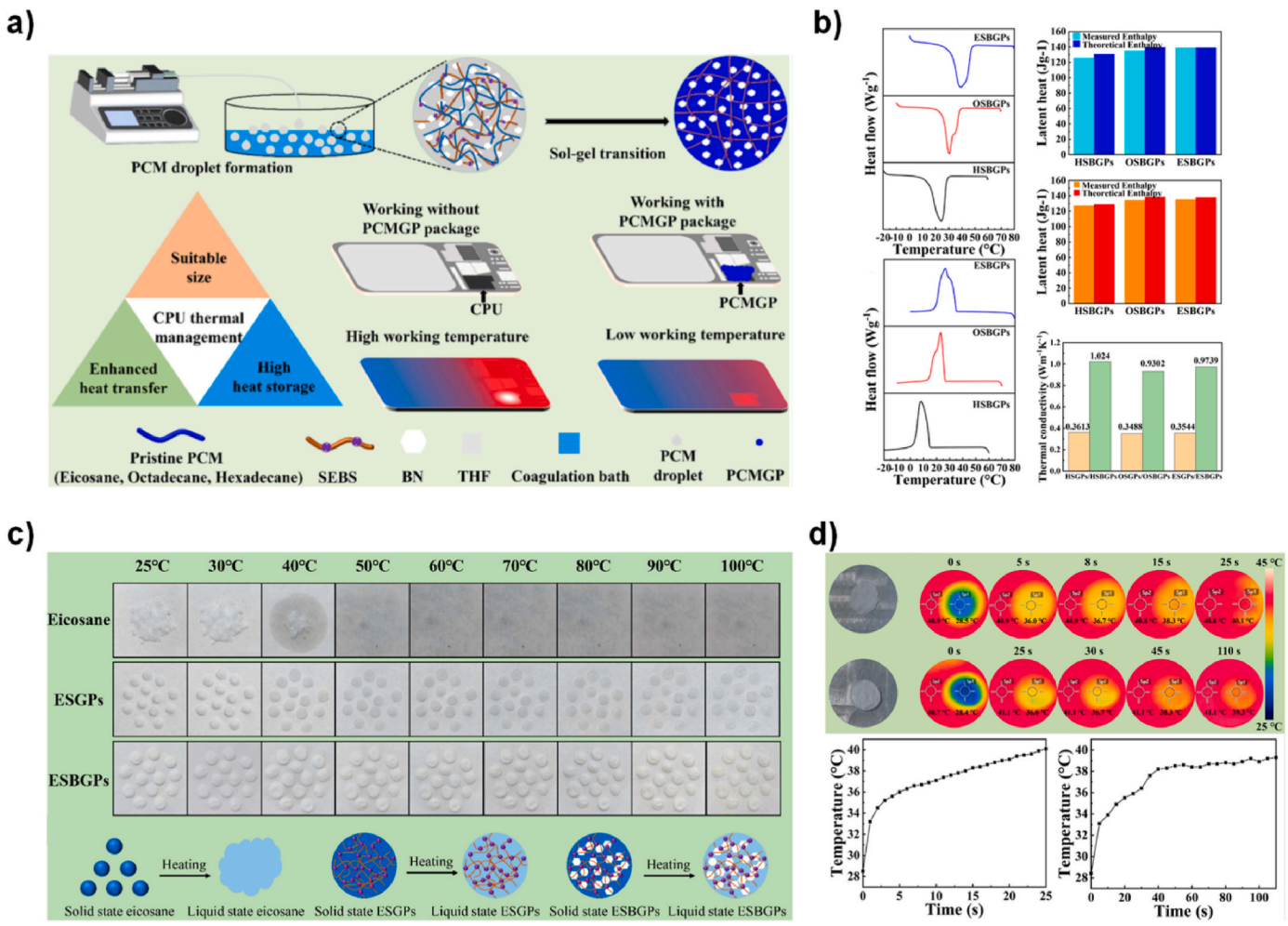


Fig. 8. (a) Schematic illustrating the fabrication of PCMGPs composed of BN micro-flakes, SEBS, and organic PCMs and their application in thermal management. (b) Thermal analysis of HSBGPs, OSBGPs, and ESBGPs: DSC thermogram of HSBGPs, OSBGPs, and ESBGPs in the heating and cooling processes. Measured and theoretical fusion enthalpies of HSBGPs, OSBGPs, and ESBGPs. Measured and theoretical solidification enthalpies of the composites. (c) Form stability analysis. (d) Experimental setup of ESGPs and ESBGPs in the heat transfer process [97].

material's strength, toughness, and durability.

Moreover, the chemical interplay between PCM molecules and the thermosetting polymer matrix ensures better compatibility while reducing the risk of phase segregation or leakage during thermal cycling. Thus, harnessing the innate reactivity of thermosetting polymers can enable organic PCM-based heat dissipation materials to achieve exceptional mechanical prowess and long-term stability by overcoming the hurdles associated with inadequate mechanical strength and liquid leakage.

Lee et al. synthesized stearic acid (SA)-grafted bisphenol A (SABPA) through an epoxy–acid reaction and mixed it with paraffin (Fig. 9). The SA grafted on the BPA prevented the phase separation between BPA and paraffin, while the curing of BPA reinforced the mechanical properties of the matrix. The investigation involved measuring the latent heat and mechanical properties of composites containing SABPA and paraffin in varying ratios, with an additional leakage test conducted to verify the enhancement in thermal stability. The findings revealed that the 1:1 SABPA/paraffin composite exhibited a latent heat of 180.85 J/g (80 % of that of pure paraffin), along with significantly improved tensile stress (206 %) and strain (269 %) compared with pure paraffin. Importantly, no leakage was observed at the melting point of paraffin (58–62 °C).

The fillers were hybridized by alkylating and impregnating BN into the pores of EG. The alkylation of BN addressed the low affinity between BN and the carbon chains in the EG and SABPA/paraffin matrix. Furthermore, the impregnation of BN facilitated the formation of efficient heat conduction paths along the EG structure within the matrix.

The PCM composites (SABPA/paraffin/EG-DBN) were fabricated using three distinct methods: mixing, hot-pressing, and injection molding. To assess the efficacy of these strategies, the thermal and mechanical properties of the composites, along with their heat management performance, were evaluated. The results demonstrated remarkable enhancements, including an ultra-high through-plane thermal conductivity of 7.21 W/mK (a 3333 % increase over paraffin), a tensile strength of 3.72 MPa (a 470 % improvement over paraffin), and an elongation-at-break of 7.72 % (a 568 % enhancement over paraffin).

Based on the results of the heat management test, the SABPA composite notably delayed the temperature increase in electrical devices, highlighting its effectiveness in practical applications. These findings underscore the potential of the developed strategies for advancing thermal management technologies.

Lee et al. employed esterification to fabricate a novel PCM, erythritol-grafted bisphenol A (ETBPA) (Fig. 10). Additionally, they developed a porous ceramic filler network using carbonized cellulose nanofiber (CNF) via freeze-casting and carbonization (see Fig. 2).

The PCM composites, namely ETBPA/BNCNF/BN and ETBPA/AlNCNF/AlN, were synthesized utilizing three distinct methods: melt mixing, hot-pressing, and injection molding. Based on the results, the hot-pressed BN-based composite exhibited an extraordinary thermal

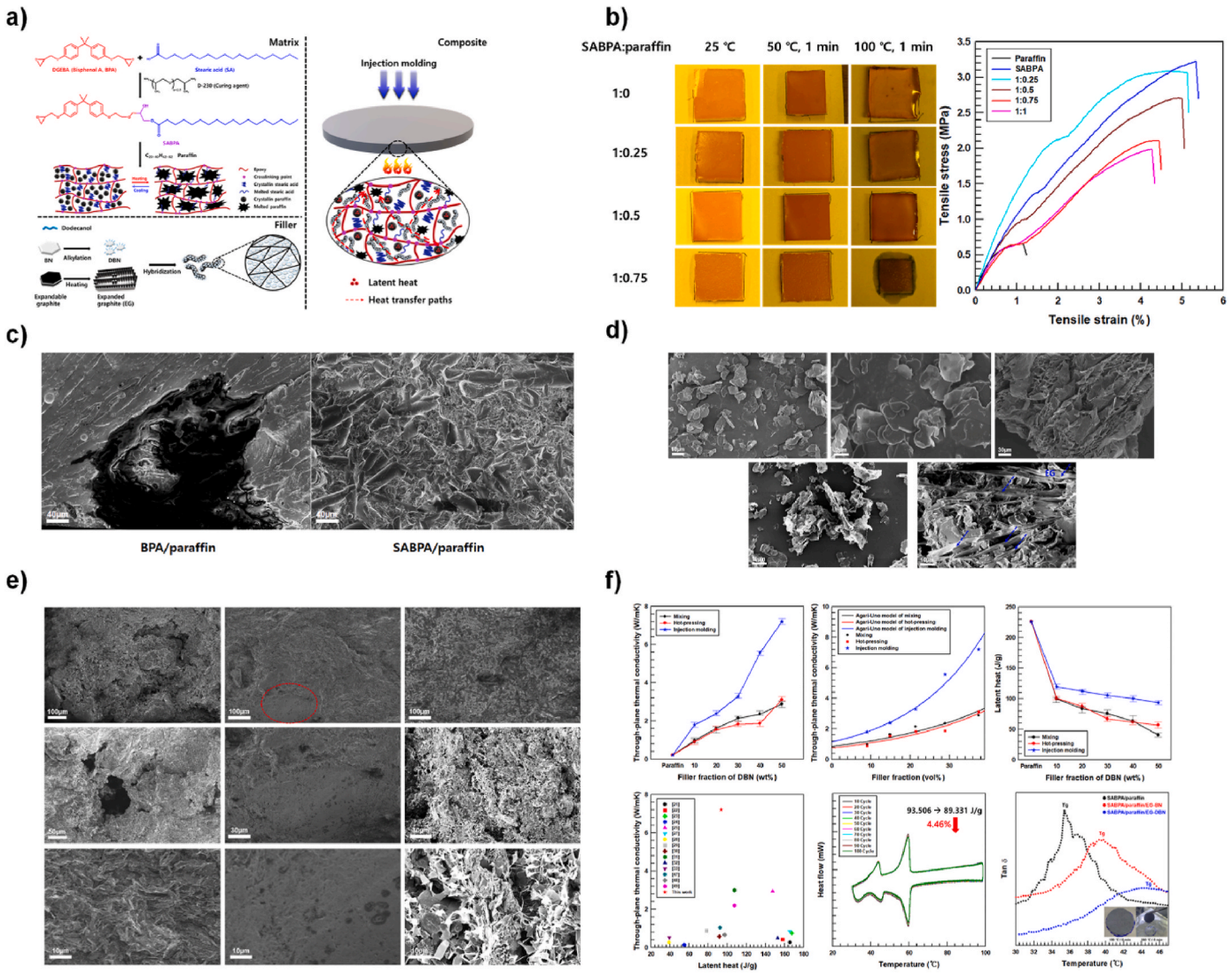


Fig. 9. (a) Schematic of SABPA/EG-DBN PCM composites. (b) Leakage test and mechanical properties of SABPA/paraffin mixed in various ratios. (c) Morphology of mixed matrix before and after SA grafting on BPA. (d) Morphology of fillers: raw BN, DBN, EG, EG-raw BN, and EG-DBN. (e) Cross-sectional images of SABPA/paraffin composites prepared using different fabrication methods. (f) Thermal properties of SABPA/paraffin composites [98].

conductivity of 13.09 W/mK, representing a remarkable 2518 % enhancement compared with pure ET. This enhancement was particularly pronounced along the through-plane direction. Moreover, the composite displayed a notable 300 % increase in tensile strength, which reached 3.2 MPa, alongside a latent heat of 98.4 J/g.

The fabricated ETBPA/BNCNF/BN composites achieved efficient thermal management, which is attributable to their combined heat absorption and dissipation properties. These properties are ascribed to their high thermal conductivity and substantial latent heat capacity. The findings underscore the potential of these PCM composites in advancing research on thermal management systems as well as in practical applications.

3.2.2. *Reaction with thermoplastic polymers*

An alternative pathway with promising prospects entails the chemical conjugation of organic PCMs with thermoplastic polymers, which instigates the formation of covalent or physical crosslinks between PCM molecules and polymer chains. This process, epitomized by copolymerization or blending, yields a homogenous PCM–polymer composite material with enhanced mechanical properties and exceptional thermal stability. Through meticulous selection of thermoplastic polymers characterized by elevated melting points and robust adhesion to PCM molecules, the resultant composite material could achieve superior strength, flexibility, and resistance to liquid leakage.

Moreover, by fine-tuning the compatibility between the PCM and the polymer through calculated selection of the polymer composition and processing parameters, the material properties can be tailored to meet the exacting demands of varied applications. Chemical reactions with thermoplastic polymers thus furnish organic PCM-based heat dissipation materials that can overcome the limitations imposed by insufficient mechanical strength and liquid leakage, thereby achieving their latent potential in thermal management applications.

The fabrication process, starting from the synthesis of the bio-based thermoplastic PCM (ETPLA) through *in situ* polymerization, to the preparation of thermally conductive composites using a hybrid filler system of oxidized carbon fiber (CF–OH) and aluminum nitride (AlN), is well-described (Fig. 11). The researchers also conducted a thorough characterization of the materials, including chemical analysis, morphological studies, thermal conductivity measurements, mechanical testing, and flame retardant evaluation.

The results are impressive, highlighting significant improvements in thermal conductivity, mechanical properties, and flame retardancy compared with existing PCM composites. The ETPLA/CF-OH/AlN composites exhibited a high through-plane thermal conductivity of

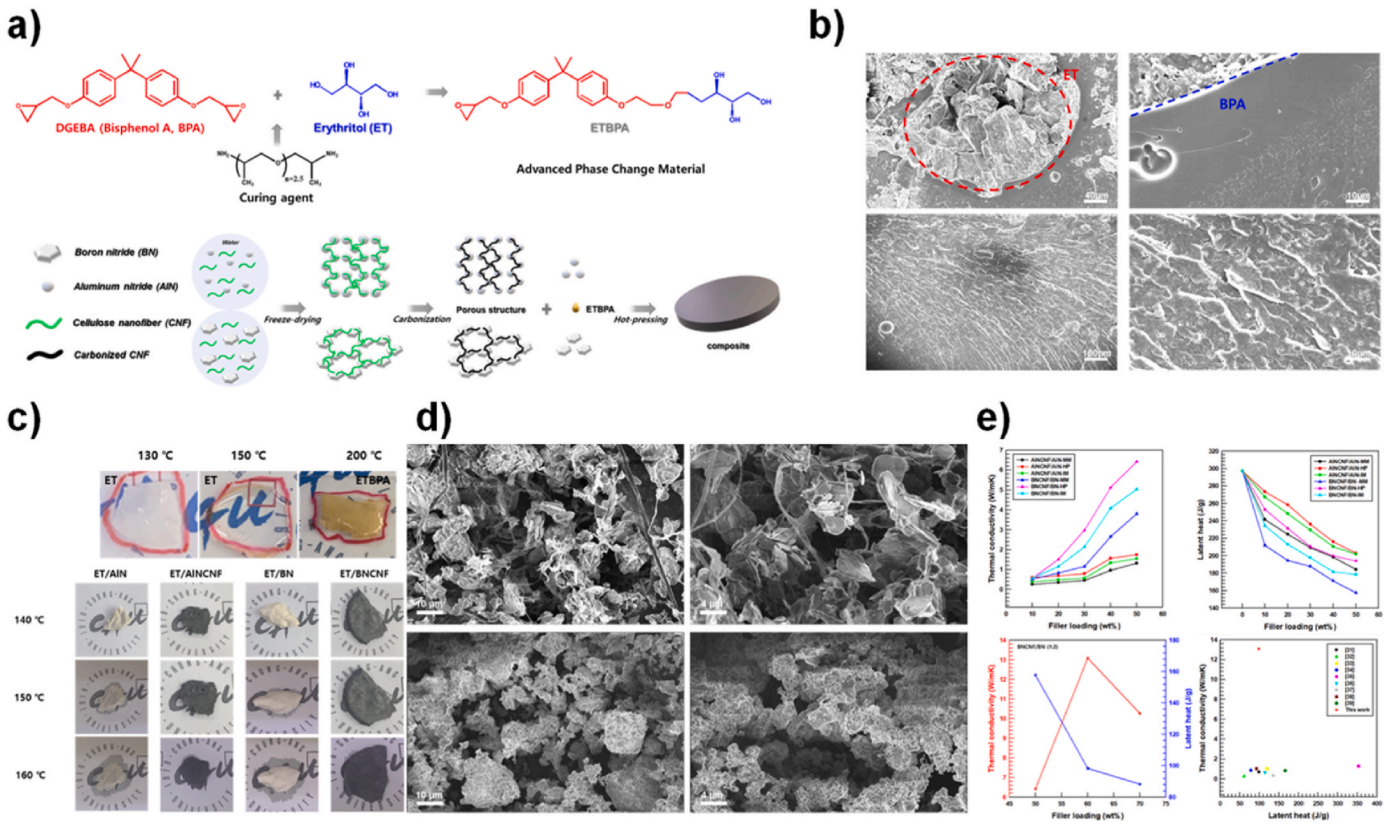


Fig. 10. (a) Fabrication of erythritol-grafted bisphenol A (ETBPA) composites. (b) Cross-sectional field-emission SEM (FE-SEM) images of ET, BPA in the physically mixed ET/BPA, and ETBPA. (c) Leakage test of ET, ETBPA, and ET/porous ceramic filler composites. (d) FE-SEM image of BNCNF and AlNCNF. (e) Thermal properties of ETBPA composites [99].

4.25 W/mK, along with enhanced tensile strength and elongation at break, in addition to achieving a V-0 rating in the UL-94 flame retardant test. The leakage test is a crucial step in evaluating the integrity of ETPLA assemblies. By subjecting the assemblies to varying conditions, such as changes in pH or temperature, their stability and potential for unintended release of cargo molecules can be assessed. This test ensures that ETPLA-based drug delivery systems maintain their integrity under physiological conditions, thereby enhancing their effectiveness and safety in biomedical applications.

Furthermore, in thermal management systems for lithium-ion batteries (LiBs), the composites achieved superior performance compared with commercial heat dissipation pads (CHP). The PCM composites effectively managed heat, maintaining lower temperatures in the LiB during discharge and inducing a delay in temperature rise under rapid thermal shock conditions.

4. Application

Due to their efficient heat dissipation properties, organic PCTIM have garnered significant attention considering their potential for application in various electronic devices. By applying various methods to improve the performance of PCTIM described above, we can learn about the efficient thermal management performance of PCTIM through representative examples of actual electronic devices. PCTIM offer a promising solution to the thermal management challenges faced by modern electronic devices such as central processing units (CPUs), batteries, and solar panels.

In the context of CPUs, organic PCM-based TIMs can effectively mitigate heat generation during operation, thereby enhancing the overall thermal performance and reliability of the processor. When applied between the CPU and the heat sink, these materials facilitate efficient transfer of heat away from the CPU, preventing overheating and maintaining optimal operating temperatures. Similarly, in batteries, which are prone to thermal runaway and degradation under high temperatures, organic PCM-based TIMs can play a crucial role in dissipating the heat generated during charging and discharging cycles. Incorporating these materials into the battery pack or directly into the battery cells can improve the thermal management, thereby extending the battery lifespan and enhancing safety. Moreover, organic PCM-based TIMs have the potential to enhance the performance of solar panels by improving heat dissipation efficiency. Solar panels operating under high temperatures experience reduced efficiency and may suffer from accelerated degradation. By integrating PCM-based TIMs into the backsheet or encapsulant layers of solar panels, the heat dissipation can be enhanced, which would improve overall performance and longevity. Overall, the application of organic PCM-based TIMs in electronic devices such as CPUs, batteries, and solar panels represents a potentially viable solution for addressing the associated thermal management challenges and thus improving device performance, efficiency, and reliability.

Lee et al. investigated the heat management performance trends of different composites, drawing comparisons between SABPA/paraffin/EG-DBN and CHP composites (Fig. 12). Traditionally, a higher thermal conductivity in heat dissipation composites is correlated with a swifter temperature increase. However, an intriguing observation emerged during Lee et al.'s analysis.

At a heat source temperature of 100 °C, despite the SABPA/paraffin/EG-DBN composite exhibiting higher thermal conductivity than CHP (3.6 W/mK), its temperature increase rate was lower. This phenomenon can be attributed to the PCM's heat storage effect, which momentarily delays the temperature rise during the phase change process. Additionally, the composite's ability to simultaneously store and dissipate heat, in combination with its high thermal conductivity and latent heat,

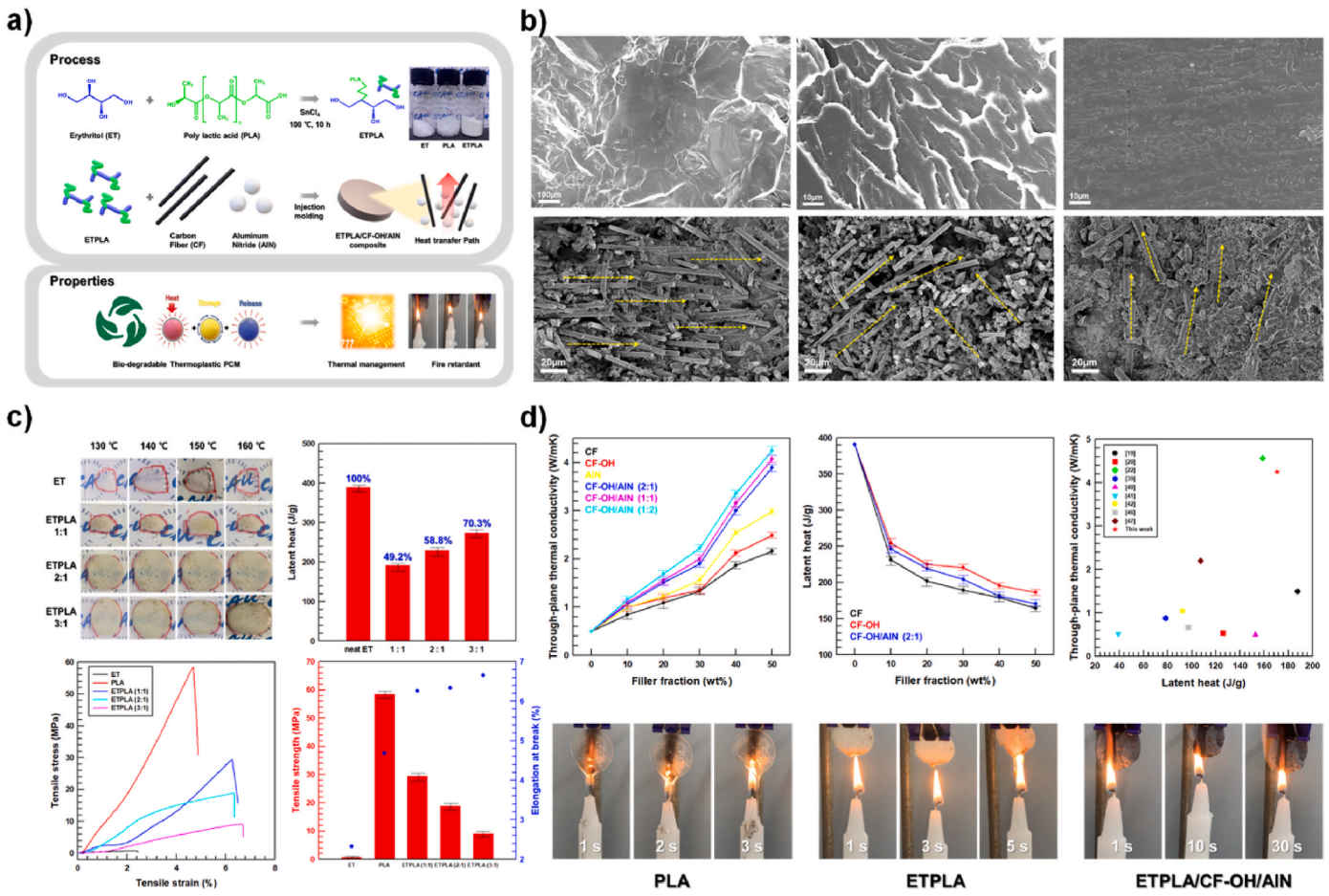


Fig. 11. (a) Schematic illustrating the manufacturing process of PCM composites. (b) Cross-sectional views showcasing the structure of both the matrix and the composites. (c) Evaluation of leakage resistance, latent heat capacity, and mechanical characteristics of ETPLA across different ratios. (d) Examination of thermal conductivity and latent heat properties of ETPLA composites with varying filler ratios and loadings, including a comparative analysis with other reported PCM composites, along with fire retardancy testing results for the composites [100].

culminates in an equilibrium state at a lower temperature.

These findings underscore the efficacy of thermally conductive PCM composites in managing heat efficiently, particularly in electrical devices such as smartphones (main board) and CPUs. Devices integrated with the PCM composite exhibited a lower temperature increase rate and equilibrium point than devices with the CHP composites did. This observation is attributed to the fact that the heat absorption by the PCM prevents sudden increases in temperature and thus delays thermal shock in devices. These findings highlight that, unlike existing thermally conductive composites, ultra-conductive PCM composites could enable efficient thermal management in electronic devices.

Lee et al. comparatively analyzed the heat management capabilities of ETPLA/CF-OH/AlN and CHP composites (Fig. 13). Practical application scenarios were simulated, involving batteries at an initial temperature of 30 °C being discharged at a 2 C rate [100].

As the battery was discharged, the temperature of both the battery body and the heat dissipation composites increased with time. Notably, the neat battery body reached a maximum temperature of 42.37 °C, surpassing the ideal operating temperature range of LiBs (20–40 °C).

Regarding the heat dissipation composites, a correlation was observed between higher thermal conductivity and swifter temperature rise. Based on an analysis of the heat source (LiB) over 600 s, the temperature increase rate of the PCM exceeded that of the CHP owing to the higher thermal conductivity (3.6 W/mK) of the former. However, despite the faster temperature increase, the LiB temperature was 2.1 °C lower when utilizing the PCM than when utilizing the CHP, and it was 3.4 °C cooler compared with that of the neat LiB. This highlights the efficacy of thermally conductive PCM composites in LiB applications.

Furthermore, a heat shock test using a hot plate corroborated the delay in the temperature increase. Below the melting point of ETPLA/CF-OH/AlN (200 °C), the PCM exhibited a faster temperature increase than the CHP due to its higher thermal conductivity. However, beyond the melting point (140 °C), the PCM's temperature increase rate dropped despite its superior thermal conductivity; this is attributed to the heat storage effect within the phase change range. This combination of heat storage and dissipation, facilitated by high thermal conductivity and latent heat, allows the PCM to attain an equilibrium state at a lower temperature than the CHP can.

In summary, bio-based thermoplastic PCM composites serve dual roles in efficient thermal management: first, they act as general thermal dissipators below the PCM's melting point, and second, they function as heat reservoirs beyond the melting point, which is crucial for mitigating rapid thermal shock and ensuring device safety.

Meanwhile, by using organic PCM as an additive and mixing it with inorganic PCM, the supercooling problem of inorganic PCM can be solved. Cao et al. To improve the shortcomings of disodium hydrogen phosphate decahydrate ($Na_2HPO_4{\cdot}12H_2O$, DHPD), an inorganic hydrated salt PCM commonly used for thermal energy storage, a hybrid PCM composite was fabricated using PEG, an organic PCM (Fig. 14). Despite its widespread application, DHPD has limitations such as significant supercooling, phase separation, leakage, and limited photothermal conversion capability, which hinder its practical utility.

In this study, PEG was employed for the first time as a nucleating agent to mitigate the supercooling of DHPD, which was effectively

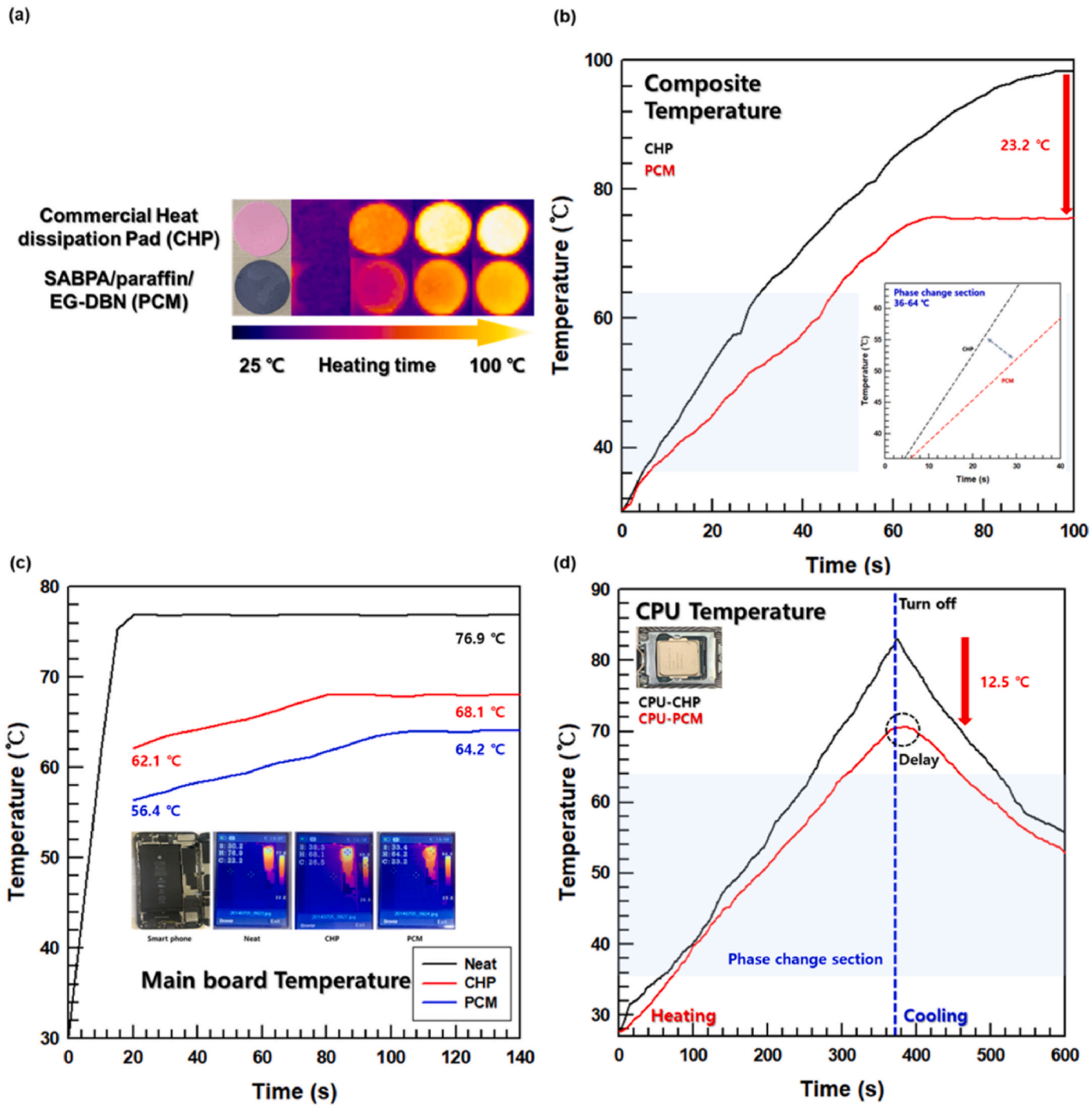


Fig. 12. (a) Infrared images and (b) temperature change graph of CHP and PCM composites with heating. Heat management performance of different composites on (c) main board and (d) CPU of smartphone [98].

eliminated. Additionally, ultraviolet light (UV)–curable polyurethane acrylate (PUA) was utilized as an encapsulation medium to suppress phase separation and leakage within the DHPD-PEG PCM (DHPDP).

Furthermore, polydopamine (PDA), a proficient solar absorber, was utilized to enhance the photothermal conversion performance of DHPDP@PUA/PDA phase change composites (DPPCCs). Based on the results, the addition of 2 % PEG4000 effectively reduced the supercooling degree of DHPD to 0.37 °C. Moreover, the DPPCCs exhibited favorable tensile strength (1.9 MPa) and elongation-at-break (31.1 %). Remarkably, even after undergoing 200 cycles, the enthalpy decreased by only 2.6 %.

Furthermore, the photothermal conversion performance of the DPPCCs was significantly better than that of the DHPDP@PUA. These findings highlight the potential for efficient thermal energy storage. Thermal infrared images of the DPPCCs captured during photothermal testing confirmed the absence of any liquid phase leakage, underscoring the efficacy of these substances.

In conclusion, the study highlighted the potential of DPPCCs for efficient thermal energy storage in solar energy systems, with potential applications in building energy conservation and intelligent temperature-regulating fabrics.

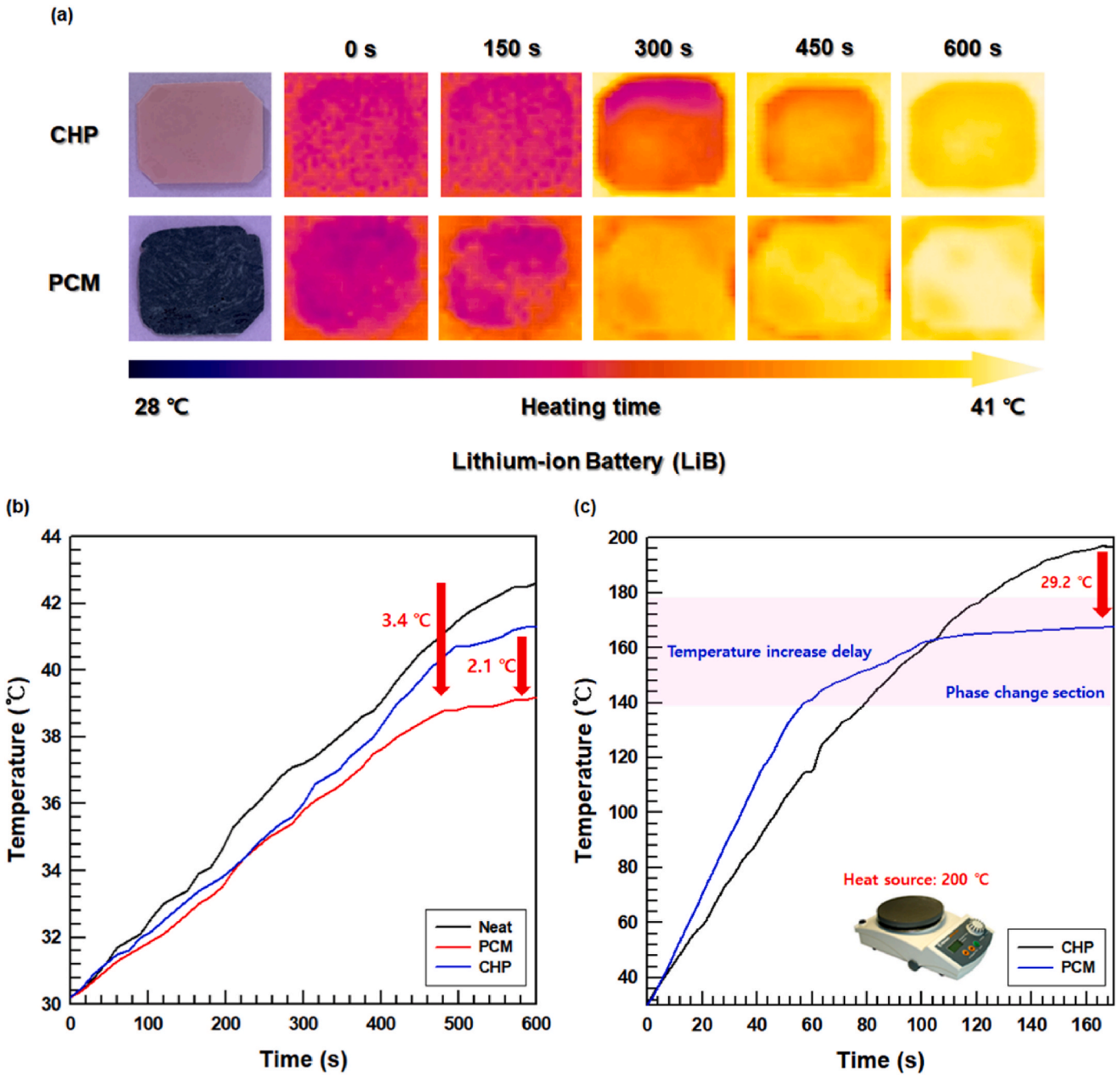


Fig. 13. (a, b) Comparison of LiB thermal management between different composites. (c) Temperature change of composites under heat shock test with a heat source (200 °C).

5. Conclusions and prospects

Recently, methods using a new type of TIM that utilize the endothermic reaction of PCM have been proposed as an efficient thermal management method for next-generation electronic devices. In this review, we explored recent advancements and trends in the utilization of organic PCMs as heat-dissipation materials in electronic devices. Due to their high latent heat storage capacity, tunable melting temperatures, and compatibility with flexible substrates, organic PCMs are attractive options for thermal management applications. In this paper, we discuss the fabrication method, characterization technology, and heat dissipation performance evaluation of TIM using organic PCM in various electronic devices, including microprocessors, LEDs, and battery modules.

In conclusion, organic PCMs represent a potentially viable option as efficient heat dissipation materials for electronic devices. The tunable properties of organic PCMs allow for customization according to specific device requirements, while their compatibility with flexible substrates enables innovative design approaches. However, further research is necessary to overcome challenges related to thermal conductivity enhancement, cycling stability, and scalability for practical implementation. If these issues are addressed, organic PCMs can play a significant role in advancing the thermal management capabilities of future electronic systems and can thus contribute to improved performance, reliability, and energy efficiency.

With reference to future prospects, the field of organic PCM-based thermal management is poised for continued growth and innovation. Emerging trends such as bio-derived PCMs, self-healing materials, and

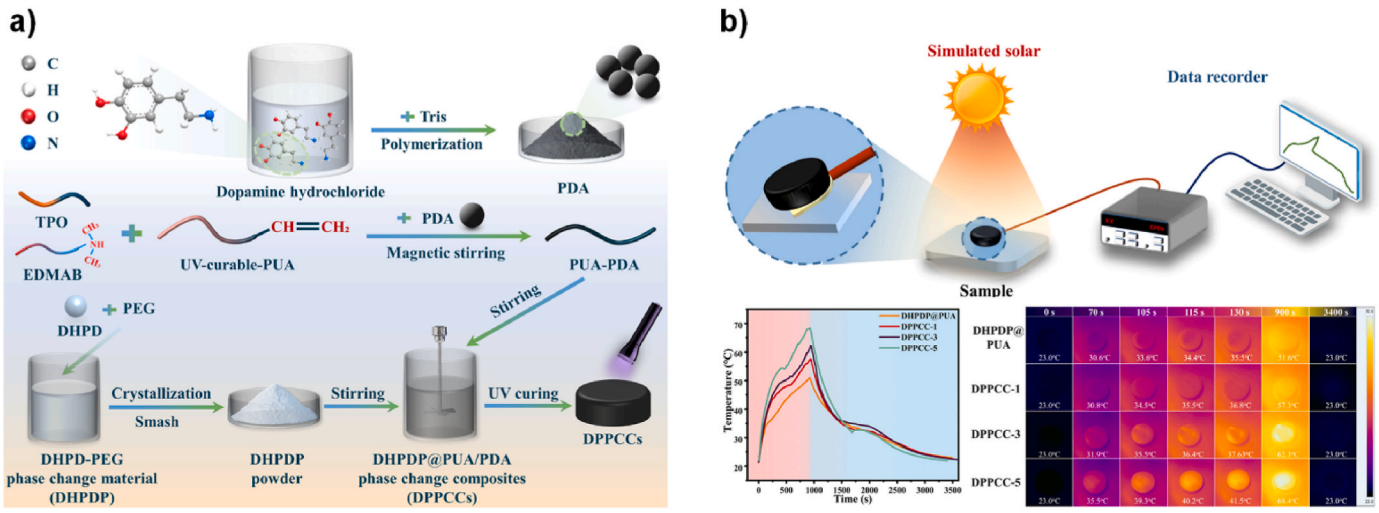


Fig. 14. (a) Flowchart illustrating preparation of DPPCCs. (b) Diagram of experimental device, along with temperature–time curves and thermal infrared images of shape-stabilized DPPCCs with different PDA mass fractions obtained from photothermal simulation experiments [101].

additive manufacturing techniques hold promise for further enhancing the performance and widening the applicability of organic PCM solutions. Additionally, interdisciplinary collaborations between material scientists, engineers, and device designers will facilitate the translation of research findings into commercially viable products. With the ongoing efforts, organic PCMs are expected to become integral components of next-generation electronic devices, enabling breakthroughs in areas such as data centers, automotive electronics, and renewable energy systems.

CRediT authorship contribution statement

Wondu Lee: Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. **Jooheon Kim:** Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.none.

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Data availability

The data that has been used is confidential.

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