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Polyrotaxane based phase change materials-A critical review

Xiao-Mei Yang [a,b,*], Baoqing Shentu [a], Zhongjie Zhai [b], Junhuan Zhao [b,*], Guang-Zhong Yin [c,d,**]

[a] College of Chemical and Biological Engineering, Zhejiang University, Hangzhou 310027, China
[b] Zhejiang Ruico Advanced Materials Co., Ltd., Huzhou 313018 Zhejiang Province, China
[c] Advanced Sustainable Polymers Lab, Escuela Politécnica Superior, Universidad Francisco de Vitoria, Ctra. Pozuelo-Majadahonda Km 1.800, Pozuelo de Alarcón, Madrid 28223, Spain
[d] IMDEA Materials Institute, C/ Eric Kandel, 2, Getafe, Madrid 28906, Spain

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ABSTRACT

Polyrotaxane-based phase change materials (PLR-PCMs) are increasingly recognized as versatile and efficient solutions for thermal energy storage, offering benefits such as outstanding form stability, leakage proof performance, flexibility, and multifunctional capabilities. This mini-review delves into the synthesis and processing techniques, functionalization strategies, and diverse applications (such as, thermal management in electronics and thermoelectric conversion) of PLR-PCMs, with a focus on recent progress in improving the flexible PCM properties and exploring innovative applications in energy storage, electronics, and thermoelectric conversion. Moreover, it provides a critical assessment of the advantages over conventional flexible PCMs, disadvantages, and future outlooks of PLR-PCMs, underscoring their transformative potential in advancing energy management and fostering sustainable technological development.

1. Introduction

Phase change materials (PCMs) have garnered significant attention due to their ability to absorb, store, and release large amounts of latent heat during phase transitions, typically between solid and liquid states.[1] This unique property makes PCMs highly effective for thermal energy storage applications, where maintaining a stable temperature is critical, such as in building system, [2] electronic heat management, [3] battery [4] and industrial thermal management. [5] As global energy demands rise and the need for sustainable energy solutions grows, PCMs present a promising approach to harvest and storage solar energy and improve energy efficiency across multiple sectors.[6].

However, traditional PCMs have limitations that restrict their broader applicability. Almost all the organic PCMs, such as paraffins and fatty acids, tend to leak during the solid–liquid transition, exhibit low thermal conductivity, and have limited mechanical stability, which reduces their effectiveness in applications requiring structural flexibility and durability [7,8]. These issues have spurred the development of flexible PCMs, which combine the thermal storage advantages of conventional PCMs with enhanced mechanical properties and adaptability. Flexible PCMs are especially valuable in emerging applications such as wearable electronics, flexible devices, and thermoregulating textiles, where both effective thermal management and material flexibility are essential.[9].

Flexible PCMs are engineered through various approaches, such as incorporating PCMs into flexible support matrices [10] or modifying their structure to enhance form stability and elasticity.[11] Methods include embedding PCMs within elastomeric polymers, [12] encapsulating them within micro- or nanostructures,[13] and introducing additives to improve properties like leakage resistance, flame retardancy and thermal conductivity. [14–16] A particularly innovative approach to flexible PCMs recently is the use of polyrotaxane (PLR)-based PCMs (PLR-PCMs). PLR is a supramolecular structure composed of cyclic molecules threaded onto a polymer chain, with bulky end-groups preventing the rings from detaching.[17] This unique architecture provides a stable, flexible matrix that can encapsulate PCMs, effectively enhancing structural integrity and reducing leakage during phase transitions.[18] The interlocking rings of PLR add flexibility, allowing PLR-PCMs to withstand repeated thermal cycling with minimal deformation. These characteristics make PLR-PCMs especially suitable for applications that demand both efficient thermal regulation and material resilience, such as in mobile electronics, and energy-efficient building

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* Corresponding authors at: Zhejiang Ruico Advanced Material Co., Ltd., Huzhou 313018, Zhejiang Province, China.
** Corresponding author at: Advanced Sustainable Polymers Lab, Escuela Politécnica Superior, Universidad Francisco de Vitoria, Ctra. Pozuelo-Majadahonda Km 1.800, Pozuelo de Alarcón, Madrid 28223, Spain.
E-mail addresses: xiaomei_yang@126.com (X.-M. Yang), gm@hzruico.com (J. Zhao), amos.guangzhong@ufv.es (G.-Z. Yin).

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materials. [18,19].

Recent studies have shown that PLR-PCMs can be further enhanced by functionalization, for example, the integration of thermally conductive fillers like graphene [20] or boron nitride [19] has been reported to improve heat dissipation, while the addition of flame retardants like phytic acid [21] increases fire safety, broadening their range of applications. Given that PLR is an excellent shape-stable PCM with unique structural and performance characteristics, this review aims to discuss the recent advances in flexible PLR-PCMs, their preparation and functionalization, and their applications in various fields. By examining the potential and challenges of flexible PLR-PCMs, we provide insights into future directions and innovative approaches in the new generation of thermal energy storage materials.

2. Preparation and processing of PLR-PCMs

The preparation of PLR-PCMs was carried out entirely in an aqueous phase (Fig. 1a), which minimizes environmental impact and aligns with

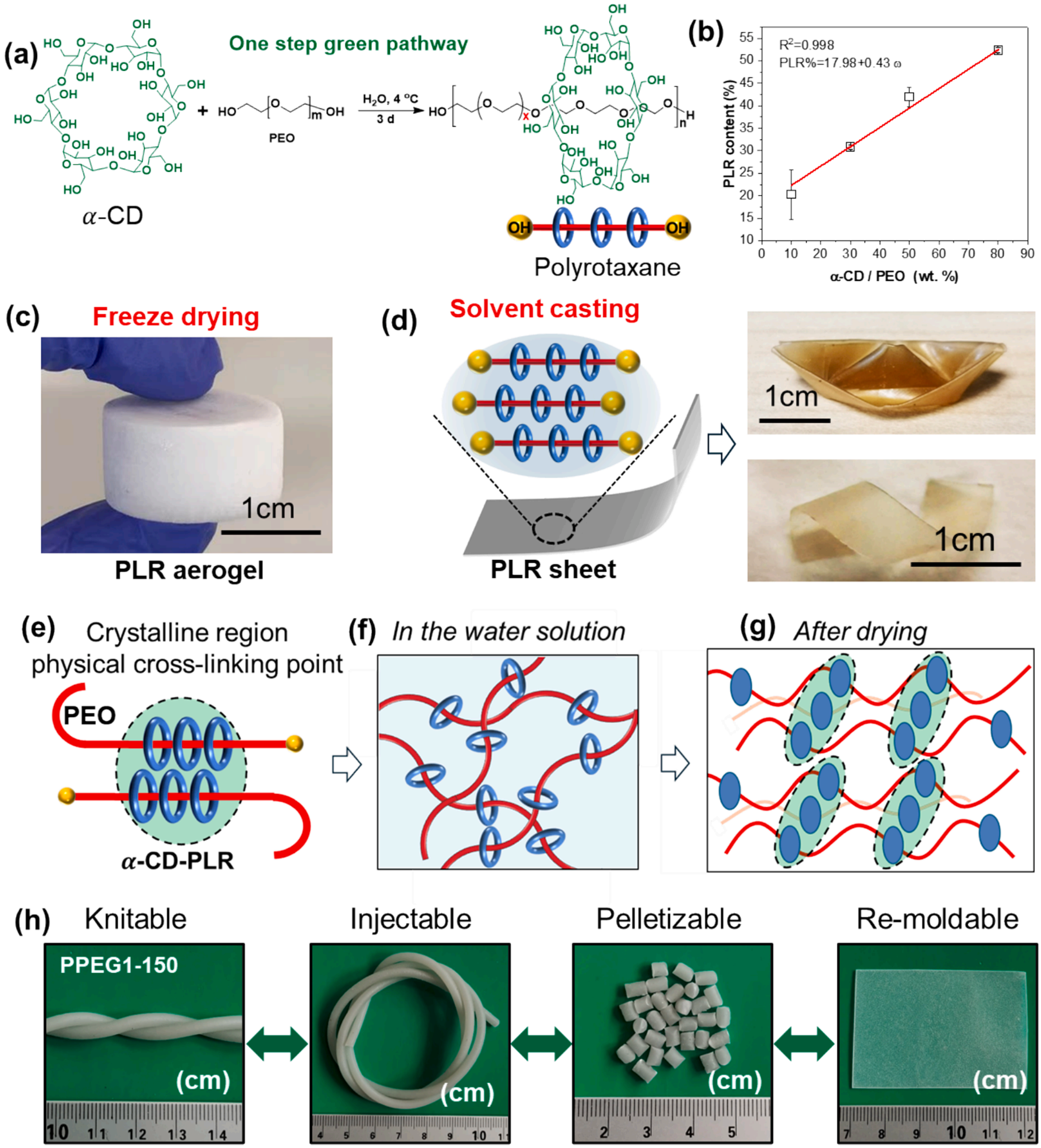

Fig. 1. (a) One step green pathway to synthesize PLR-PCM, (b) the measured PLR contents of the solid that can't be removed by Dichloromethane treatments (Redrawn from reference [22], Copyright (2021) Elsevier), (c) Image of PLR aerogel obtained by freeze drying, (d) the sheet illustration obtained from solvent casting at room temperature and the images (Folded miniature boat and twisted specimen strips) to illustrate the flexibility of the PCM, (e) α-CD crystalline region forming physical cross-linking point, (f) micro scale structure illustration of PLRs in the solution, (g) Schematic diagram illustrating the role of α-CD crystallites in polyrotaxane materials as physical crosslinking points, enhancing shape stability and leakage resistance, and (h) The extruded filaments intertwine with each other, the flexible filaments can be knotted, the extruded filaments can be granulated, and the granulated samples can be further hot-pressed into sheets with adjustable thickness. (Redrawn from reference [23], Copyright (2022) Elsevier).

green chemistry principles. To balance flexibility and mechanical strength, we selected polyethylene oxide (PEO) 900 k for PLR-PCM synthesis. This approach avoids the need for complex chemical end-capping.[24,25] Dissolution experiments using a good solvent (Dichloromethane) for PEO indicate the presence of a significant number of soluble components (PEO chains) in the PLR sample (Fig. 1b). As shown in Fig. 1c, aerogel products can be easily obtained by freeze-drying the aqueous solution of PLR. Sheet-like solid materials can be fabricated through solvent casting at room temperature. These sheets demonstrate outstanding flexibility, shape retention, and resistance to leakage above the melting temperature. (Fig. 1d) Experimental results indicate that PLR-PCM exhibits excellent leakage-proof performance and cycling stability, suggesting a relatively stable molecular structure. We speculate that below the melting point (>90 °C) of α-CD on the PLR chain, α-CD remains strongly associated (Fig. 1e), ensuring effective physical crosslinking. This, in turn, supports the shape stability and leakage resistance of PLR-PCM (Fig. 1 e-g). Following synthesis, various molding methods can be applied to shape and process the PLR-PCMs. Common molding techniques include freezing drying, hot press and solvent casting.

(1) Freeze-drying method involves freezing the PLR-PCM or PLR-PCM with functional fillers, followed by the removal of the solvent through sublimation under reduced pressure, creating a highly porous structure. In our previous report [26], freeze-drying was successfully used to prepare PLR-based foam and functional foam alloys (Fig. 1c), resulting in 3D foam structures with exceptional PCM encapsulation properties. The foams achieved porosity levels ranging from 94.65 % to 97.83 %, ensuring efficient adsorption and encapsulation of Polyethylene glycol (PEG). The prepared PCMs demonstrated ultra-high latent heat values between 171.6 J/g and 191.7 J/g, highlighting their excellent energy storage capacity. The structural stability and uniformity of the foam alloys were notably improved with the introduction of PLR, which enhanced compatibility between PEG and the foam matrix via hydrogen bonding interactions. Overall, the freeze-drying method has proven to be an efficient and versatile approach for fabricating form-stable, lightweight, and highly porous PCM supports, aligning with goals of environmental sustainability by using biomass polymers.

(2) Hot Press is a key fabrication method that involves applying heat and pressure to the PLR-PCM mixture due to its thermoplastic nature, enabling it to be molded into specific shapes while ensuring uniform dispersion of additives. One of the typical works focused on fabricating PLR/PEG-Boron Nitride (BN) nanocomposites using a blending method with water as the only solvent, followed by a critical hot-pressing step to optimize thermal conductivity and ensure shape stability. In this process, PEG is dissolved, and BN nanosheets are dispersed into the solution, followed by the addition of dissolved PLR to form a homogeneous mixture. After freezing drying (for a better fillers dispersion), the precursor undergoes hot pressing, which ensures compactness, structural integrity, and uniform distribution of components, while enabling precise control over the composites' shape and size.

(3) Solvent casting method offers several advantages in the preparation of PCMs, especially the solid sheet (Fig. 1d). In our previous work, the solvent casting method was effectively employed to prepare Pentaerythritol phosphate (PPP)-modified polyrotaxane (PLR-P) PCMs, phytic acid (PA)-modified polyrotaxane (PLR-PA) PCMs, and polyhedral oligomeric silsesquioxane (POSS)-grafted polyrotaxane (POSS-PLR) PCMs, using water as the solvent, ensuring a sustainable process. This method enabled the good dispersion of PPP and PA. This will be further elaborated in the following section on flame-retardant modification. Given the multi-hydroxyl properties of PLR, surface grafting modification can be achieved through chemical reactions. Taking POSS-PLR PCMs as another example, it is clearly observed that the samples can be curled, folded, and bent after preparation using the solvent casting method, showcasing their ultra-high flexibility. Furthermore, hydrophobicity improved remarkably, with water contact angles (WCA) increasing from 71° (PLR) to 123° (POSS-PLR). These results collectively underscore the solvent casting method as a versatile, efficient, and sustainable approach for developing high-performance, flame-retardant, hydrophobic, and shape-stable thermal energy storage materials. [18,21,27] Notably, a key advantage of PLR-based PCMs lies in their ability to undergo conventional thermal processing, enabling facile reshaping and repeated reprocessing. In addition, they typically exhibit excellent flexibility. As illustrated in Fig. 1h, PLR-based PCMs are compatible with extrusion, pelletizing, and multiple cycles of hot-press molding.

3. Functionalization of PLR-PCMs

The multifunctionality of PLR-PCMs is achieved through systematic advancements in flame retardancy enhancement, high phase change enthalpy optimization, and thermal conductivity improvement. These improvements are derived from an intricate balance of chemical modifications, nanofiller integration, and structural design innovations, which synergistically enhance their thermal management properties, and fire safety.

3.1. Flame retardancy enhancement in PLR-PCMs

Flame retardancy in PLR-PCMs is a crucial property for ensuring safety and reliability, especially in applications like building energy saving, thermal energy storage and electronic device thermal management. Improvements in flame retardancy are primarily achieved through chemical modifications, nano fillers integration, and biomass-derived additives.

(1) Acid Source-Based Flame Retardants. Acid source additives, such as Pentaerythritol Phosphate (PPP, Fig. 2a) and Phytic Acid (PA, Fig. 2**b**), act through both a gas phase and a condensed-phase flame-retardant mechanism. When subjected to high temperatures, these additives decompose to produce phosphoric acid derivatives, which catalyze the formation of a dense and stable char layer on the material's surface. This char layer acts as a thermal and physical barrier, preventing further decomposition and reducing heat and oxygen transfer. In PPP-modified PLR composites, char residue increased significantly from 1.1 % to 22.9 %, resulting in a 52.42 % reduction in pHRR and a 39.11 % reduction in THR. Similarly, PA-modified composites achieved a 54.2 % reduction in pHRR and a 34.0 % reduction in THR, while maintaining a char residue content of 16.5 %. Both materials demonstrated UL-94 V-0 ratings and high limiting oxygen index (LOI) values of 33.0 % and 28.2 %, respectively, highlighting their exceptional self-extinguishing behavior. Acid additives alter the pyrolysis mechanism of the polymer matrix, favoring char layer reinforcement over volatile release, which contributes to a more controlled combustion process. On the other hand, as illustrated in Fig. 2b-ii, the formation of phosphorus-containing radicals enables the efficient scavenging of flammable radicals, thereby decreasing the concentration of combustible species. Consequently, phosphorus-based flame retardants can also function effectively in the gas phase.

(2) Surface Grafting and Crosslinking Techniques. Surface modification, such as Polyhedral Oligomeric Silsesquioxane (POSS) grafting, serves a dual purpose by enhancing both fire safety and mechanical stability (Fig. 2c). As demonstrated in our previous work, POSS grafting introduces hydrophobic silsesquioxane cages, which act as thermal barriers and promote char formation during combustion. In POSS-coated PLR (POSS-PLR), the pHRR was reduced by 18.8 %, and the THR decreased by 19.1 %, compared to unmodified PLR. These improvements are attributed to the formation of dense char layers, facilitated by POSS's hybrid inorganic–organic structure, which effectively insulates the material during thermal decomposition.

(3) Nanofiller-Based Flame Retardants. Nanofillers, such as MXene, Graphene Nanoplatelets (GNP), and BN, can enhance the flame retardancy of PLR-PCMs to a certain extent by acting as thermal and physical barriers, which partially impedes heat and mass transfer

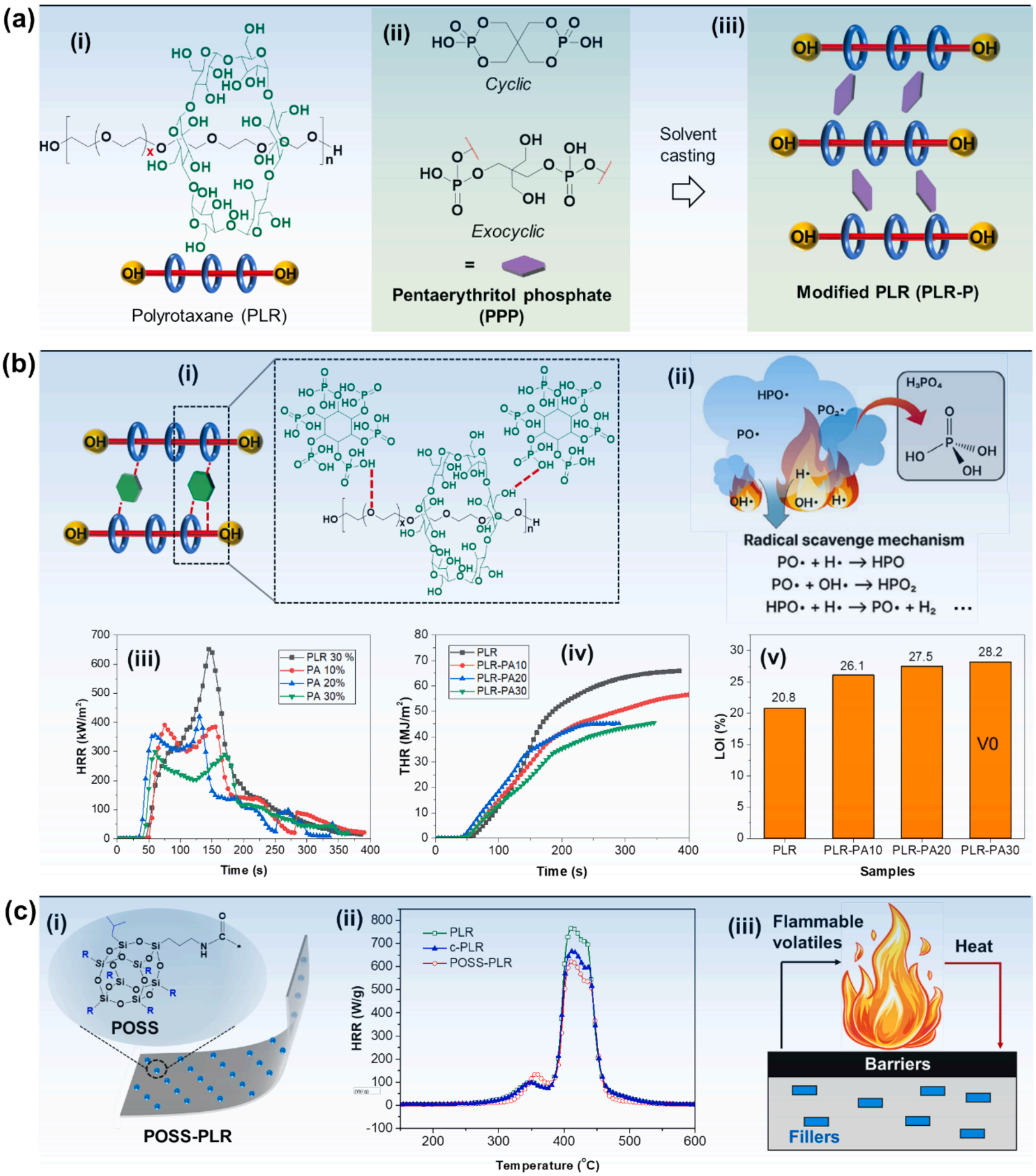

Fig. 2. (a-i) Chemical structure of PLR, (−ii) Chemical structure of the lab synthesized P-containing flame retardant, (−iii) Schematic of the microscopic molecular structure of the flame-retardant PLR-PCM. (Redrawn from reference [18], Copyright (2022) Elsevier) (b-i) Schematic of the microscopic structure of phytic acid blended PLR-PCM. (−ii) gas phase flame retardant mechanism illustration for P-containing PCMs. (Redrawn from reference [28], Copyright (2025) Wiley) (−iii) pHRR, (−iv) THR, (−v) LOI results of the PLR and PA functionalized PLR-PCM. (Redrawn from reference [21], Copyright (2022) Elsevier) (c-i) Illustration of obtained POSS surface coated PLR sheep and (−ii) the MCC curves for the 3 samples. (Redrawn from reference [27], Copyright (2024) Elsevier) and (iii) Schematic of the flame-retardant mechanism induced by the barrier effect of nanofillers.

processes, as illustrated in Fig. 2c-iii. These nanofillers reduce heat transfer and oxygen permeation, reinforce the char layer, and slow down heat propagation within the material matrix. MXene nanosheets create a dense char layer during combustion, blocking heat and oxygen transfer while preventing dripping. Their high surface area and functional groups ensure uniform dispersion and catalyze the early decomposition of cyclodextrin, promoting char formation. This results in a threefold increase in char residue at 800 °C compared to neat PLR-PEG composites, while maintaining thermal stability up to 250 °C, making MXene-enhanced PLR-PCMs ideal for applications in thermal management and electronic protection.[29] GNPs form a 2D protective barrier that reduces combustion intensity, facilitates stable char formation, and ensures uniform heat distribution. With GNP addition, the pHRR decreased by 4.3 % to 12.4 %, the THR dropped by 10.1 % to 17.0 %, and the char residue at 800 °C increased from 2.0 % (neat PLR-PEG) to 8.8 % (PLR-PEG-10GNP). These properties make GNP-enhanced PLR-PCMs suitable for energy storage, electronic cooling, and industrial thermal management.[20] Similarly, BN nanosheets enhance thermal barrier properties, limiting heat and oxygen transfer while maintaining the composite's integrity during degradation. In PLR-PEG-BN composites, the pHRR decreased by 16.6 %, the THR reduced by 25.5 %, and the char residue at 800 °C rose from 3.1 % (neat PLR-PEG) to 39.3 % (PLR-PEG-60BN). These advancements highlight BN's potential for thermal energy storage and fire-safe device thermal management.[19] Together, MXene, GNPs, and BN offer powerful solutions for improving the flame retardancy and thermal stability of PLR-PCMs.

(4) Flame-retardant mechanism. Based on the above analysis, it is evident that the flame-retardant performance of the PLR-PCM system primarily arises from the synergistic contribution of two or three of the following three mechanisms. **i)** The char-forming characteristics of α-CD. The flame retardant mechanism of α-CD is mainly due to its polyhydroxy structure. α-CD will be dehydrated and carbonized during combustion to produce a large amount of carbon residue. The more stable and compact the carbon layer, the better the matrix protection. [30] α-CD as a carbon source decomposes at high temperatures, and the catalytic system generates a stable carbon layer. [31] ii) The spatial barrier effect of the filler. It is believed that the barrier effect of the exfoliated layered nanomaterials is the flame retardant mechanism for polymer/layered fillers nanocomposites in common. [32] iii) P-based additives exhibit excellent flame-retardant effects through three primary aspects: *1)* radical scavenging, which leads to the formation of H_3PO_4, *2)* absorption of heat through dehydration and endothermic reactions,

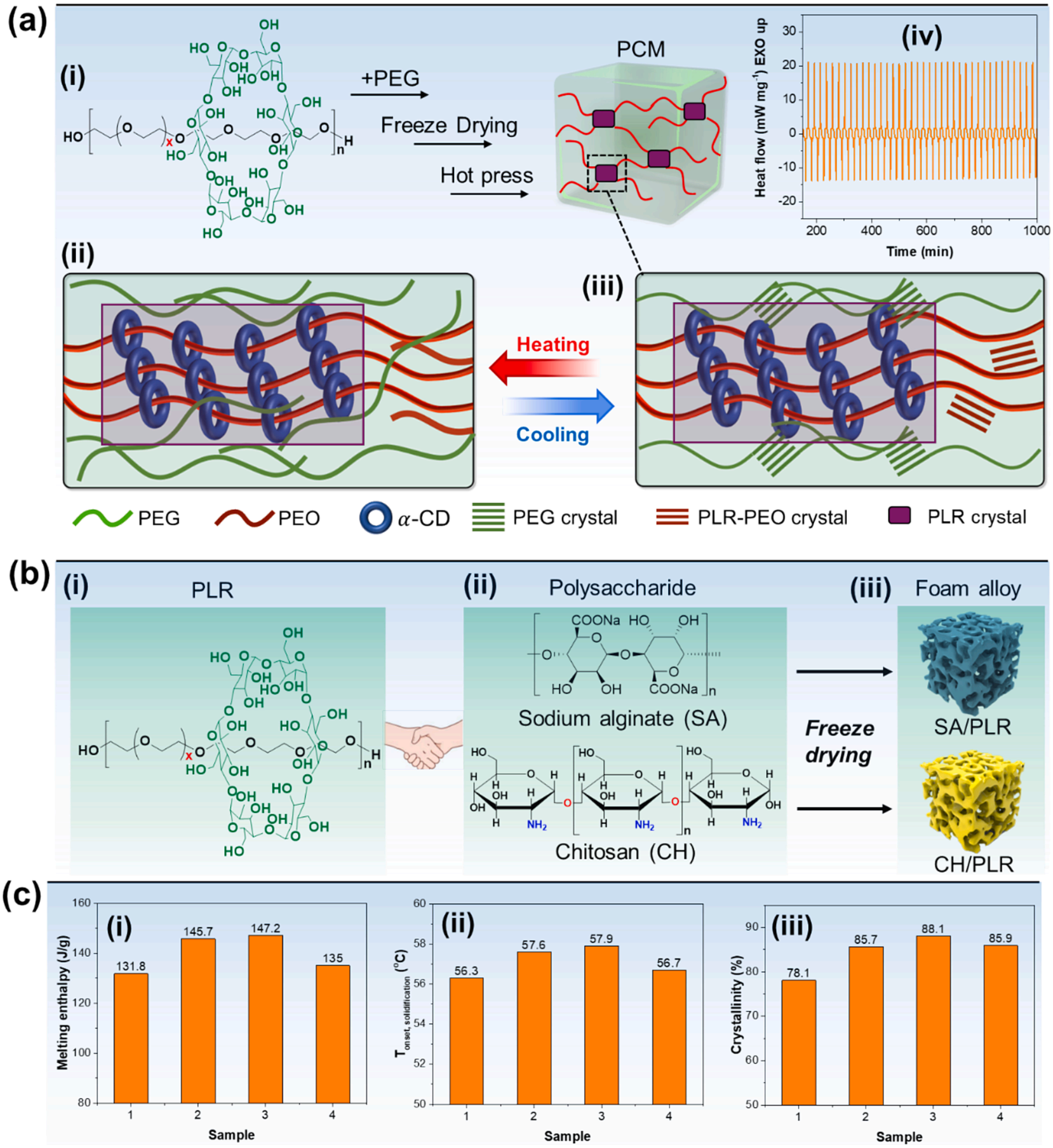

Fig. 3. (a-i) PLR encapsulated PEG with high enthalpy PCM was prepared through a solvent blending PEG-freeze-drying-hot pressing procedure. (−ii) Schematic of the molten state of PLR-PEG, (−iii) Microstructure schematic of PLR-PCM at room temperature, (Redrawn from reference [23], copyright (2022) Elsevier), (−iv) cycle performance obtained by Differential Scanning Calorimetry, and (b-i) Chemical structure of PLR, (−ii) Chemical structures of Chitosan and Sodium Alginate, (−iii) Schematic of the final foam alloy's porous structure. (Redrawn from reference [26], copyright (2022) Elsevier), (c-i) Melting enthalpy values, (−ii) onset solidification temperature, and (−iii) crystallinity of the sample 1: PLR-PEG6k, 2: PLR-PEG6k-3 %GNP, 3: PLR-PEG6k-5 %GNP, and 4: PLR-PEG6k-10 %GNP.

resulting in the production of H_2O and P_2O_5 layer, and *3)* reduction of oxygen concentration or blocking the contact between oxygen and fuel.[28].

3.2. High phase change enthalpy in PLR-Based PCMs

PLR-PCMs are renowned for their high phase change enthalpy, ensuring efficient thermal energy storage and release. This exceptional performance stems from the encapsulation of PEG within the PLR framework, which provides substantial latent heat storage and structural integrity (Fig. 3a). The enthalpy values of PLR/PEG composites range from 116.1 J/g to 162.2 J/g, depending on PEG content and molecular weight. For instance, a composite containing 77.8 wt% PEG (PPEG6-350) achieves an impressive phase transition enthalpy of 162.2 J/g.[23] PLR and PEG are perfectly compatible because they have the same chemical structure of the main polymer chain. Moreover, incorporating higher molecular weight PLR and support materials enhance enthalpy efficiency, exceeding 100 %, significantly boosting thermal storage performance. These materials also exhibit remarkable cycling stability, maintaining consistent phase change enthalpy and melting temperatures over more than 40 thermal cycles (Fig. 3a-iv), making them highly reliable for advanced thermal energy storage applications.

Additionally, various approaches have been employed to further enhance the enthalpy and performance of PLR-PCMs. (1) **Incorporation of Nanofillers.** Fillers such as MXene, GNPs, and BN can be integrated into PLR-based composites to act as crystallization nucleation agents, enhancing PEG crystallization and improving thermal energy storage. As shown in Fig. 3c, the incorporation of GNPs leads to an increase in the phase change enthalpy of the material within an appropriate filler loading range. This nucleation-induced crystallization behavior is substantiated by the elevated crystallization onset temperature and enhanced final crystallinity. In MXene-enhanced PLR-PEG composites, latent heat values remain high, ranging from 123.9 J/g to 159.6 J/g, depending on the MXene content and PEG molecular weight.[29] Similarly, GNPs and BN effectively maintain or improve phase change enthalpy. GNP-enhanced PCMs achieve latent heat values ranging from 132.9 J/g to 142.9 J/g,[20] while BN-reinforced materials demonstrate stable and high enthalpy values, even at increased filler concentrations.[19] The integration of these fillers strengthens interfacial bonding, minimizes energy losses during phase transitions, and ensures consistent performance over multiple thermal cycles. These advancements highlight the multifunctionality and efficiency of PLR-PCMs, making them ideal for applications in thermal energy storage, electronic cooling, and smart energy systems. Notably, the strategy of improving crystallinity using fillers only has a slight promoting effect at lower filler contents. (2) **Optimized Structural Designs.** Advanced encapsulation techniques and porous frameworks, such as PLR/Chitosan (CH) and PLR/Sodium Alginate (SA) composites, significantly enhance compatibility with PEG, resulting in ultra-high latent heat values ranging from 171.6 J/g to 189.5 J/g and improved energy storage efficiency.[26] The high porosity of these foam composites (up to 97.83 %) and their excellent compatibility with PEG facilitate strong hydrogen bonding interactions and promote efficient PEG crystallization. Additionally, these PCMs demonstrate exceptional stability, maintaining consistent latent heat and phase transition temperatures over more than 80 thermal cycles, making them highly suitable for long-term and demanding applications.

The high phase change enthalpy of PLR-PCMs enables their application in thermal energy storage systems, electronic device cooling, and smart building materials, ensuring efficient energy utilization and stable thermal performance. These materials seamlessly combine exceptional energy storage capabilities with enhanced thermal management, meeting the growing demand for high-performance and sustainable PCMs. The synergistic interplay of flame retardancy, thermal conductivity, and high enthalpy in PLR-PCMs underscores their potential as sustainable thermal management solutions. By incorporating acid source additives, nanofillers, and biomass-derived agents, these materials achieve outstanding safety, performance, and durability.

3.3. Thermal conductivity improvement in PLR-PCMs

The thermal conductivity of PLR-PCMs has been significantly enhanced by integrating advanced nanofillers such as GNPs, BN and MXene (Fig. 4). These nanofillers create efficient thermal pathways, addressing the inherent low conductivity of conventional PCMs and enabling rapid heat transfer and dissipation during phase transitions. Fig. 4a illustrates the blending schematic of PLR-PEG and GNP, showing the dispersion of graphene within the PLR-PEG matrix (Fig. 4a-ii), as well as the rectangular strips (Fig. 4a-iii) and flexible sheets (Fig. 4a-iv) obtained via hot pressing. Fig. 4b and 4c respectively present the dispersion schematics of BN and MXene in the PLR-PEG system, along with SEM images of their well dispersion. Additionally, the sheet-like samples obtained by thermoplastic molding also showing the excellent flexibility of the PCMs.

GNPs leverage their 2D structure to form continuous thermal pathways within the PLR-PEG matrix. Thermal conductivity improvements ranged from 60 % to 257 %, depending on GNP content, with 10 wt% GNPs providing significant enhancement while preserving the PCM's latent heat and structural integrity (Fig. 4d). These advancements ensure efficient temperature regulation in energy storage and industrial cooling applications. BN nanosheets demonstrated remarkable thermal conductivity enhancements in PLR-PEG composites, increasing values from 0.30 W/m·K (PLR-PEG) to 2.72 W/m·K (PLR-PEG-60BN), representing an 806.7 % improvement (Fig. 4e). BN's layered structure creates highly effective thermal pathways while maintaining structural stability and complementing the PCM's flame retardancy. This makes BN-enhanced PCMs multifunctional and highly efficient for advanced thermal management applications. MXene nanosheets, with their high surface area, intrinsic thermal conductivity, [33] and functional groups, [34] improve interfacial bonding within the PLR matrix. In PLR-PEG composites, MXene increased thermal conductivity by 137.5 % (PLR-PEG1k) and 44.3 % (PLR-PEG6k), ensuring effective heat transfer (Fig. 4**f**). This improvement supports applications like electronic cooling and thermal energy storage by maintaining uniform heat dissipation.[29] The improved thermal conductivity in these PLR-PCMs results from the strong interfacial bonding between the fillers and the matrix, uniform filler dispersion, and the ability of the nanofillers to form continuous and effective heat transfer networks. In the PLR-PCM system, the fillers are randomly distributed, making it suitable to be described by the Maxwell theoretical model. $k = k_{matrix}\left[\frac{k_{filler}+2k_{matrix}+2\varphi(k_{filler}-k_{matrix})}{k_{filler}+2k_{matrix}-\varphi(k_{filler}-k_{matrix})}\right]$, where k_{matrix} is the thermal conductivity of the PCM matrix, k_{filler} is the thermal conductivity of the filler, and φ is the volume percentage of the filler.[35] A typical nonlinear increase in thermal conductivity with rising filler content is observed, as illustrated in Fig. 4d–f. These enhancements make PLR-PCMs highly suitable for applications requiring efficient thermal management, such as thermal energy storage, electronic cooling, and industrial systems.

4. Application cases of PLR-PCMs

PLR-PCMs have emerged as a groundbreaking innovation in various application domains, thanks to their outstanding thermal management capabilities, mechanical flexibility, and environmental compatibility. These features make PLR-PCMs highly impactful across a range of cutting-edge technologies and applications as common reported, electronic chipset thermal management module, [36] battery thermal management systems. [37] Below are some key areas where they excel:

4.1. Thermal management in electronic devices

PLR-PCMs are ideally suited for advanced thermal energy storage

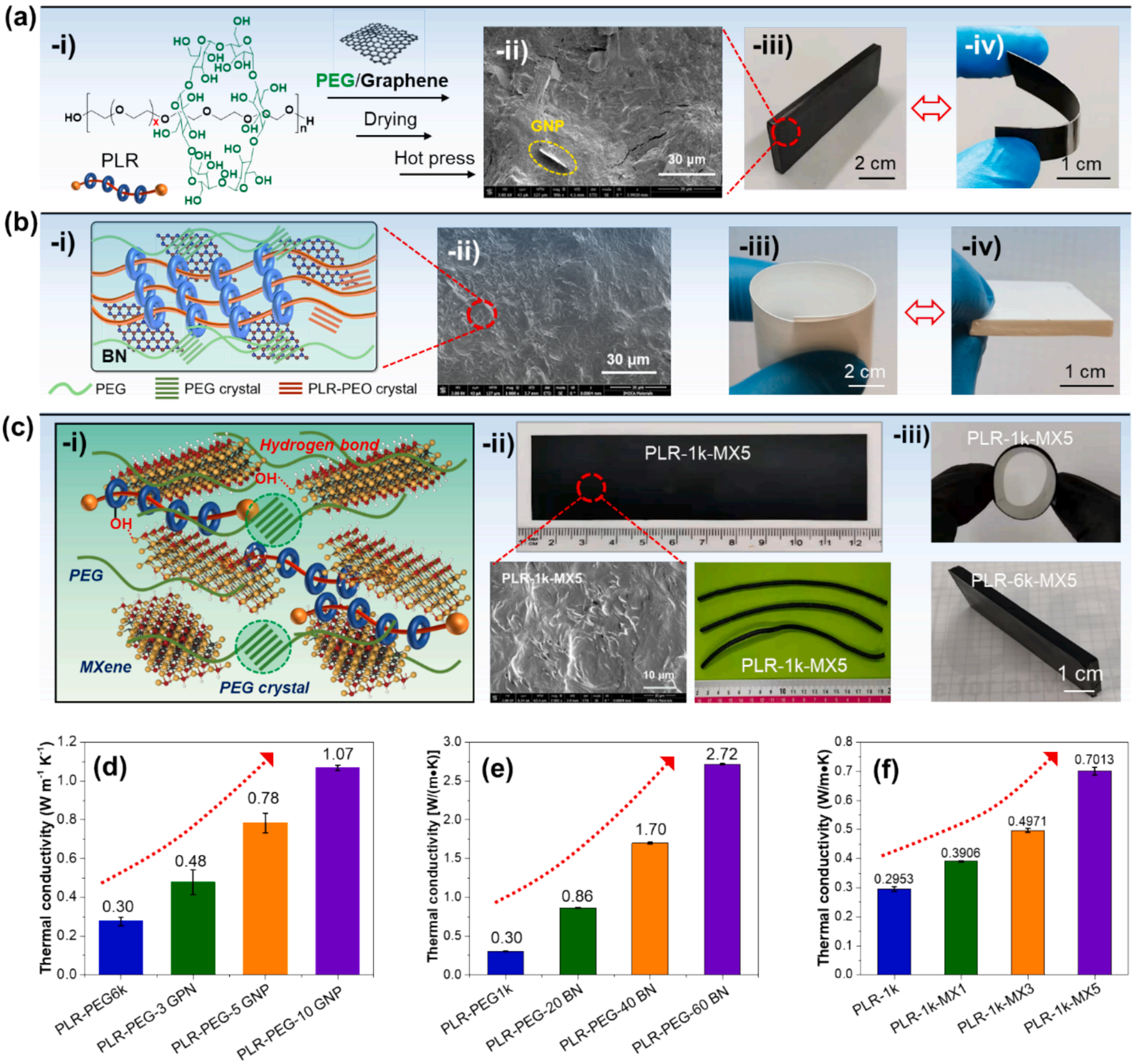

Fig. 4. (a-i) PLR and GNP blending modification illustration, (a-ii) SEM image of GNP functionalized sample. (a-iii) Hot-pressed rectangular sample. (a-iv) Hot-pressed thin sheet demonstrating a certain degree of flexibility. (Redrawn from reference [20], copyright (2023) Wiley) (b-i) Schematic of the BN-modified PLR system. (b-ii) SEM image of BN functionalized sample. (b-iii) Sample can be rolled up. (b-iv) Thick PCM sample obtained by hot pressing. (Redrawn from reference [19] copyright (2023) Elsevier) (c-i) Schematic of the microscopic structure of the MXene and PLR-PCM system. (c-ii) Flexible thin sheet obtained by hot pressing, SEM image and filament produced by extrusion. (c-iii) Sample flexibility demonstration—rolling and repeated hot pressing to achieve thickness control. (Redrawn from reference [29] copyright (2024) Elsevier) (d) Thermal conductivity modification results for the GNP system. (e) Thermal conductivity modification results for the BN system. (f) Thermal conductivity modification results for the MXene system.

systems, owing to their high latent heat values, excellent phase change stability, durable cycling performance and flexibility. These materials address the critical need for efficient thermal regulation in electronics, ensuring optimal functionality and reliability.

In particular, thermal management in electronics benefits significantly from PLR-PCMs integrated with advanced fillers such as graphene, MXene, and boron nitride. These fillers enhance thermal conductivity and prevent leakage, making PLR-PCMs exceptionally effective for dissipating heat in high-performance devices such as solid-state disks (Fig. 5a-b) and smartphones (Fig. 5c-d). By ensuring stable thermal regulation, PLR-PCMs mitigate overheating risks, prolong device lifespans, and support the demands of next-generation 5G technology. Typically, Fig. 5(a) demonstrates the temperature regulation effect of PCM on the test sample under a constant heat source. Experimental results show that the system containing PCM maintains a temperature 3–5 °C lower than that of the system without PCM, effectively ensuring thermal safety (Fig. 5b). It is worth noting that since heat generation in electronic devices can be continuous, the latent heat storage capacity of PCM is inherently limited. Once the PCM is fully melted, its thermal regulation capability significantly declines. As a result, PCM-based thermal management has a limited effective duration. Based on our previous work, the thermal management effect of PCM lasts approximately 30 min to 1 h, indicating that its most significant impact occurs within the first 30–60 min of device operation.

4.2. *Thermoelectric conversion*

PLR-PCMs also play a pivotal role in thermoelectric conversion by providing efficient thermal energy storage and release capabilities. Their ability to achieve “peak shaving and valley filling” in thermoelectric generators ensures consistent energy flow. The incorporation of thermally conductive fillers like boron nitride further optimizes heat

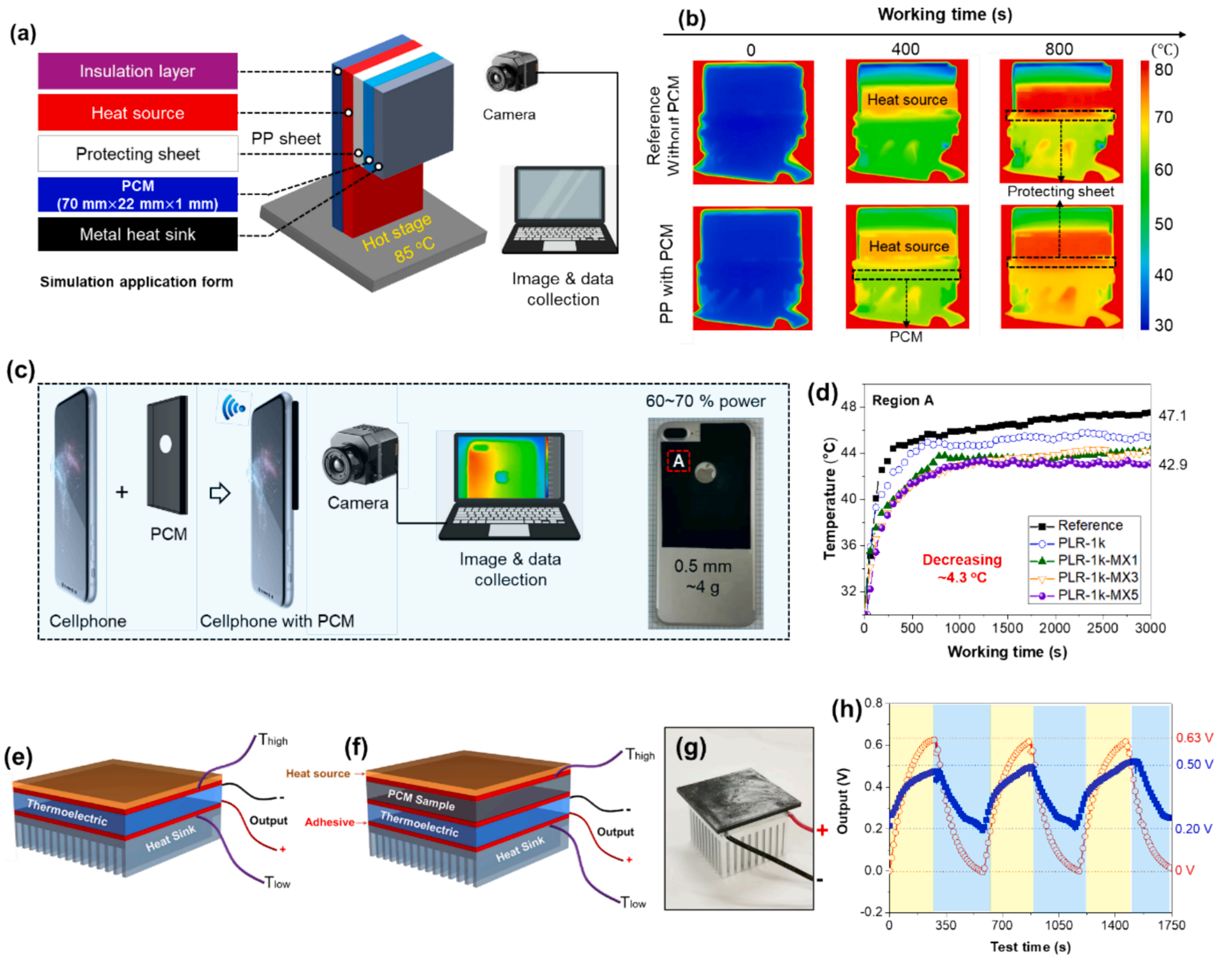

Fig. 5. (a) Real application form of the PCM for solid state disk (SSD) thermal management, and illustration of the experimental stack structure of the device, test process and sample test diagram: including sample heating, signal acquisition and data storage, (b) IR images of the devices during heating at 85 °C; ((Redrawn from reference [20], copyright (2023) Wiley)) (c) Phone with PCM and the temperature regulation illustration, and (d) temperature change curves of region A; (e) structure illustration of the test devices without PCM, (f) Packed structure illustration of device with PCM, (g) physical image of the test system and (h) Voltage output of the two devices with or without PCM (Redrawn from reference [20], copyright (2023) Wiley).

transfer, enhancing the efficiency and performance of thermoelectric systems. These attributes position PLR-PCMs as a vital component in industrial energy recovery and sustainable energy management applications. For a typical example, GNP-functionalized PLR-PCM (Fig. 5e-h) effectively enhances the stability of the output voltage in thermoelectric converters. Specifically, the voltage remains within the range of 0.2–0.5 V, whereas systems without PCM fluctuate between 0 and 0.63 V (Fig. 5h). Once the external heat source is removed, the device ceases to generate any voltage. This result clearly demonstrates the “peak shaving and valley filling” effect of PCM in electrical thermal management.

By merging high thermal efficiency with commitment to sustainability, PLR-PCMs are revolutionizing the landscape of thermal energy storage and management. Their unique combination of advanced thermal regulation, mechanical flexibility, and environmental compatibility is driving innovation and setting new benchmarks across diverse high-tech applications.

5. Advantages, disadvantages and outlooks

5.1. Advantages

In the context of emerging technologies, flexible and sustainable PCMs are gaining significant attention due to their potential to address the growing challenges of thermal regulation in advanced applications such as flexible electronics (including wearable devices, foldable screens) and chip-level thermal management. [38,39] For instance, researchers have developed PCM-embedded polymer matrices that combine high flexibility with excellent thermal conductivity, enabling effective heat dissipation in wearable electronic textiles. Additionally, hybrid PCMs with graphene or carbon nanotube fillers have demonstrated significant improvements in both mechanical robustness and thermal regulation, making them ideal for foldable displays and other flexible technologies.[10] PCM has also shown promising results in maintaining electronic devices, e.g., chip temperatures under peak operational loads while being lightweight and eco-friendly. [40,41] Moreover, the focus on sustainability aligns with global efforts to reduce environmental impact. The development of bio-based, recyclable, and low-carbon PCMs supports the transition towards greener technologies without compromising functionality or efficiency. [42].

The current key strategies in preparing flexible PCMs are shown in Fig. 6a, including (I) Carbon scaffold/PCM composites, (II) Elastic foam/PCM composites, (III) Microcapsule PCMs, (IV) Co-polymer, (V) Polymer (blending) based flexible PCMs and (VI) Core-Shell/PCM composites. [10] We have also outlined the main shortcomings of each method, as all clearly listed in Fig. 6a. For example, some key issues in the encapsulation process of (II) elastic foam/PCM composites include: Easy to leak during melting due to the open pore structure, and low thermal conductivity. (V) Polymer (blending) based flexible PCMs face major challenges such as high cost due to the multiple chemical synthesis steps, chemical wastes and relatively low enthalpy.

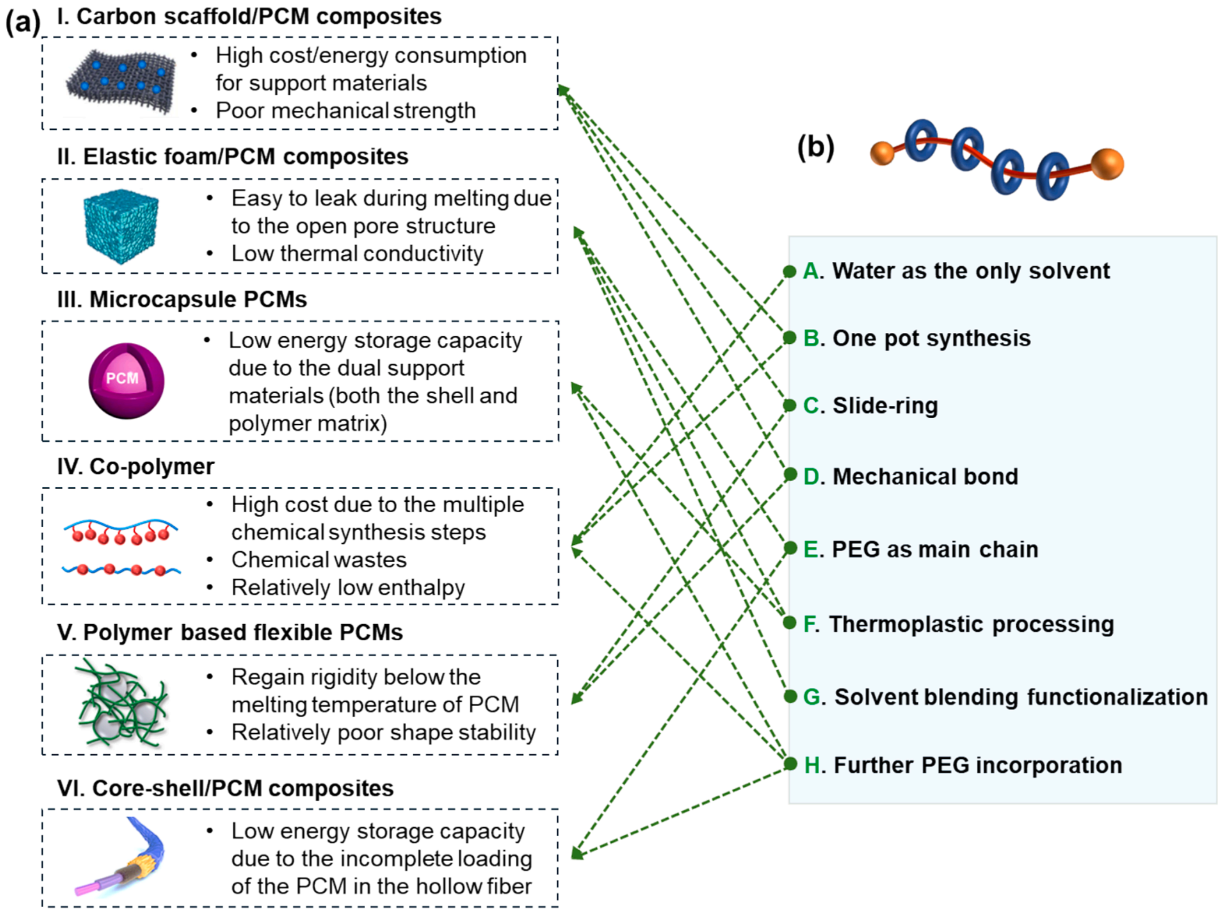

Fig. 6. (a) Common fabrication techniques for flexible PCMs and their significant drawbacks; and (b) schematic representation of the PLR structure along with its typical fabrication process and performance advantages, aiming at overcoming the limitations of traditional flexible PCMs fabrication strategies.

Table 1
Comparison between PLR-based PCMs and conventional flexible PCMs in terms of fabrication methods and key properties.

No.	PCM	Strategies or support materials	Enthalpy (J/g)	Thermal C. (W m^{-1} K^{-1})	Solvents used	Ref.s
1	Paraffin	Expanded graphite (EG), thermoplastic elastomer, twin-screw extrusion	124.6	2.2	–	[44]
2	Paraffin	Melting blending with thermoplastic polyeher ester elastomer + EG	112.5	1.267	–	[45]
3	Paraffin	Solvent blending + EG	121	1.7	Cyclohexane	[46]
4	Paraffin	Natural rubber + MXene	126.3	0.16	Water	[47]
5	Paraffin	olefin block copolymer (OBC) + EG, Melting blending	176.7	–	–	[48]
6	Paraffin	Styrene-b-(ethylene-co-butylene)-b-styrene triblock copolymer (SEBS) and expanded graphite (EG), thermal crosslinking	125–136.5	0.2–0.45	–	[49]
7	Oct	SEBS (Kraton G1650), Solvent blending	187.7	–	Cyclohexane	[50]
8	Paraffin	LDPO or OBC, (+EG) physically-blended strategy	102	1.64	–	[51]
9	PEG	Directional freezing and freeze-drying methods, CNT, Chitosan, PVA	152.7	0.50	Water	[52]
10	PEG	thiol-ene photo-crosslinking reaction	128.3	–	UV initiators and multifunctional thiol compounds	[53]
11	PEG	Toluene diisocyanate chain Extension	78.2–118.7	–	N, N-dimethylformamide (DMF)	[54]
12	PEG	4,4′-diphenylmethane diisocyanate, Chain extension, +Graphene oxide	157.6	0.983	DMF	[55]
13	PLR	–	104.29	–	Water	This review
14	PEG	PLR	162.2	–	Water	This review
15	PLR + PEG + GNP	PLR + GNP	132.9–142.9	0.48–1.07	Water	This review
16	PLR + PEG + BN	PLR + BN	91.16–123.50	0.86–2.72	Water	This review
17	PLR + PEG + MXene	PLR + MXene	123.9–145.4	0.4833–0.7013	Water	This review

PLR consisting of biomass cyclodextrin (CD) “wheels” and polymer “axles” is appealing mechanically supramolecular polymers.[43] As shown in Fig. 6b, PLR can be synthesized in water (**A**) via a one-pot (**B**) method, which can at least be used to balance costs, such as environmental and processing costs. The slide-ring (**C**) structure and mechanical bonds (**D**) significantly enhance its mechanical performance, while the flexibility and thermoplastic properties of high-molecular-weight PEO (**E**) enable thermoplastic processing (**F**) of the PLR-based PCM, and ensures excellent leakage resistance, high mechanical strength, and cost-effective post-processing flexibility. In addition, (**G**) solvent blending functionalization capacity enables the post-functionalization of PLR-PEG materials, such as enhanced thermal conductivity and flame retardancy; and (**H**) further PEG incorporation facilitates the fabrication of leak-proof, high-enthalpy flexible PLR-PEG PCMs. This innovative approach is expected to overcome the limitations of conventional strategies and pave the way for a new generation of flexible, high-performance PCMs. Additional point-to-point advantages and their corresponding improvements over traditional technologies are illustrated by the arrows in Fig. 6.

It can be observed in Table 1 that the conventional flexible PCMs in this series are prepared using melt blending or solution blending processes, and the phase change enthalpy values range between 100 and 170 J/g, with thermal conductivity varying from 0.2 to 2.2 $W\,m^{-1}\,K^{-1}$. Furthermore, Due to the physical blending of the PCM with the supporting material, it can be inferred that, the material’s leak resistance may not be well maintained at higher filler content. Another widely reported method for preparing flexible PCMs involves PEG-based flexible PCMs. The preparation of PEG-based flexible PCMs typically involves using flexible polymer-based supporting materials, and either chain extension of PEG with diisocyanates or chemical crosslinking of PEG (e.g., thiol-ene click chemistry) to create leak-proof PEG-PCMs. However, chemical modification of PEG often involves complicated processes and the use of large amounts of organic solvents, such as DMF. Additionally, PEG-based PCMs modified by chemical methods tend to have relatively low phase change enthalpy values (ranging from 78.2 to 157.6 J/g). For PLR-based PCMs, we know that they are synthesized in an aqueous phase, are flexible, and can easily achieve leakage proof and phase change enthalpy values of up to ~160 J/g. As PLR also serves as the supporting material, it contributes to thermal storage, allowing the thermal enthalpy efficiency during PEG encapsulation efficiency to exceed 100 %. Moreover, the thermal conductivity can be significantly enhanced by incorporating thermally conductive fillers, with reported values reaching as high as 2.72 $W\,m^{-1}\,K^{-1}$. In summary, PLR-based PCMs represent a novel approach in the PCM field and an exploration of supramolecular polymers for PCM applications. They offer significant advantages and have considerable potential for future development.

5.2. Disadvantages

Owing to its unique supramolecular structure and the stringent requirements for aqueous conditions during synthesis, research and development are predominantly confined to a limited selection of polymers such as PEG (polyethylene glycol) and PEO (polyethylene oxide), significantly restricting the diversity of available materials. During the experimental process, freeze-drying is commonly employed in the drying stage. While this method preserves material structure, it is associated with low efficiency and high costs (Including production cycle and raw material cost). Moreover, the final material’s practical application is constrained by its pronounced hydrophilicity, leading to performance degradation in high-humidity environments, thereby limiting its long-term stability and broader application potential across various fields. Based on the aforementioned content, the challenges currently faced by PLR-based PCM systems can be summarized as follows:

(1) Flame retardancy and safety concerns and low phase change enthalpy;
(2) relatively high cost; and
(3) poor water/humidity resistance.

5.3. Outlook

As the phase change energy storage materials evolve toward multi-functionality and high performance, Polyrotaxane-based PCMs exhibit significant potential due to their unique molecular architecture and tunable latent heat properties. Future research on these materials can be expanded in the following directions:

(1) Development of Flame-Retardant Phase Change Aerogels and highly flame retardant PLR based PCMs (Strategy for Challenge 1). The combination of polyrotaxane-based PCMs with aerogel technology offers a promising pathway for creating advanced functional materials with both high thermal energy storage efficiency and flame-retardant properties. Compared to traditional PCMs, polyrotaxane-based phase change aerogels may exhibit lower thermal conductivity and superior thermal insulation. Incorporating flame-retardant elements such as phosphorus, silicon, or nitrogen, or integrating inorganic nanomaterials, can further enhance their flame resistance and thermal stability. These materials hold great promise in areas such as building insulation, aerospace, and electronic thermal management, particularly in applications requiring high safety standards and excellent thermal performance. Additionally, the obtained PLR aerogel can serve as an excellent carrier for other types of PCM work substances, offering a new approach to designing 3D shape-stable and high-enthalpy PCMs.

(2) Low-Cost Phase Change Materials Based on β-Cyclodextrin (Strategy for challenge 2). β-Cyclodextrin (β-CD), a naturally occurring oligosaccharide, is widely regarded for its low cost, excellent biocompatibility, chemical modifiability, and lower cost than α-CD based PLR, making it an ideal building block for polyrotaxane-based materials. Synthesizing polyrotaxane-based PCMs using β-CD as the core unit can significantly reduce production costs while leveraging the cavity structure of β-CD [56,57] to fine-tune the phase transition temperature and thermal properties of the materials. Additionally, the formation of inclusion complexes between β-CD and polyethylene glycol (PEG) can improve thermal cycling stability and extend the service life of the material, facilitating large-scale applications in green building and energy storage systems. In addition, another approach to overcome the high cost is to develop high-value-added applications, such as thermal management for portable devices, enhancing the efficiency of solar thermal conversion devices, chip thermal management, and thermal management for perovskite solar cells, among other advanced applications.

(3) Crosslinking and Hydrophobic Modification (Strategy for challenge 3). One of the primary challenges for polyrotaxane-based aerogels is their poor stability in high-humidity environments due to their strong hydrophilicity and poor form stability if used as PEG support materials. Crosslinking and hydrophobic modification are effective strategies to address this limitation. Introducing chemical crosslinking can improve the mechanical strength and shape stability of the materials, reducing leakage and dimensional changes during phase transitions. Meanwhile, chemical modification with hydrophobic groups such as long-chain alkyl or silane compounds can significantly reduce surface hydrophilicity, enhancing long-term stability under humid conditions. This approach is expected to expand the application of polyrotaxane-based PCMs in flexible energy storage devices and waterproof functional materials.

In addition, advanced techniques such as in-situ Transmission Electron Microscope (for in-situ phase transition characterization of PCM), and Material Studio/data-driven approaches (for big data-assisted optimization and development of PCM) should be employed to systematically optimize and select the efficient design of high-performance PCMs, and even enable customized design, in order to achieve the optimal balance between process, performance, composition, and cost.

6. Conclusion

PLR-PCMs represent a groundbreaking class of materials with outstanding energy storage capacity and multifunctional properties. While challenges such as cost and processing complexity remain, ongoing advancements in material design, sustainability, and scalability are expected to overcome these barriers. With vast potential in renewable energy, electronics, smart materials, and eco-friendly applications, PLR-PCMs are well-positioned to address global energy and sustainability challenges. Continued research into polyrotaxane-based PCMs, particularly in areas like flame retardancy, cost reduction, and stability enhancement, will further expand their applications in energy storage, intelligent thermal regulation, and functional composite materials. By integrating structural design with multifunctional modification strategies, these materials are poised to become a cornerstone of next-generation phase change energy storage technologies, offering both academic and practical value.

CRediT authorship contribution statement

Xiao-Mei Yang: Writing – review & editing, Writing – original draft, Methodology, Conceptualization. **Baoqing Shentu:** Writing – review & editing, Supervision. **Zhongjie Zhai:** Writing – review & editing, Validation. **Junhuan Zhao:** Writing – review & editing, Supervision, Funding acquisition. **Guang-Zhong Yin:** Writing – review & editing, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization.

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.

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Data availability

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

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