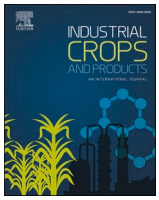
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Bio-based phase change composites consisting of collagen-grafted polyethylene glycol for efficient thermal management

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ABSTRACT

This study introduces a cutting-edge phase change material (PCM) engineered from collagen-grafted polyethylene glycol (CP) and reinforced with a hybrid blend of carbon fiber (CF) and aluminum nitride (AlN). This PCM composite exhibits an outstanding perpendicular thermal conductivity of 3.99 W/mK, an impressive 1800 % increase in tensile strength, and a latent heat capacity of 88.1 J/g. It undergoes an endothermic phase transition within the temperature range of 42–55 °C. When integrated into lithium-ion batteries, this PCM composite effectively regulates temperature fluctuations by leveraging its substantial latent heat capacity, outperforming conventional conductive polymer composites. Therefore, CP/CF/AlN composites offer an effective solution for advanced thermal management in electronic systems, as they deliver both efficient heat storage and dissipation.

1. Introduction

Electronic devices are now integral to modern life, influencing everything from daily routines to industrial processes (Jia et al., 2023; Liu et al., 2021a; Zhang et al., 2020). The trend toward miniaturized, high-performance, and energy-efficient technologies, such as smartphones (Naduvilakath-Mohammed et al., 2024), laptops (Zhu et al., 2024), or advanced systems in aerospace (Bai et al., 2023) and automotive industries (Lv et al., 2024), is ever-increasing (Coutinho et al., 2023; Ferretto and Viola, 2023; Nam et al., 2023; Liu et al., 2025). However, this drive for miniaturization and enhanced performance presents a critical challenge: heat management. Excessive heat buildup can compromise the functionality and dependability of electronic components, pose safety hazards, and hinder technological advancements (Maiorino et al., 2024; Dan et al., 2023; Huang et al., 2025; He et al., 2025).

To address this issue, effective thermal management is crucial. Traditionally, heat dissipation has relied on metallic heat sinks and fans, which manage thermal loads through conduction and convection. While these methods have been effective for traditional systems, they struggle to meet the demands of modern devices, which are characterized by higher heat flux densities and tighter spatial constraints. As electronic assemblies become increasingly compact and densely packed, the limitations of conventional cooling methods become evident. This has spurred the exploration of advanced thermal management solutions, including innovative materials designed to efficiently handle the heat challenges of compact electronic devices (He et al., 2023; Afaynou et al., 2023; Lin et al., 2024).

The growing emphasis on eco-friendly initiatives has led to substantial interest in bio-based materials. Among them, phase change materials (PCMs) stand out for their ability to manage thermal energy effectively. Substances such as paraffin (Hu et al., 2023), stearic acid (Hwang et al., 2024), and palmitic acid (Sun et al., 2024) can absorb and release significant heat during their transition from solid to liquid phases, positioning them as highly efficient solutions for thermal management.

Organic PCMs offer a distinct approach to thermal management when compared to conventional heat sink materials, which primarily conduct heat through solid mediums. These PCMs stand out because of their ability to absorb and release latent heat during phase transitions. They can transition from solid to liquid states within a defined endothermic temperature, enabling them to absorb or release significant

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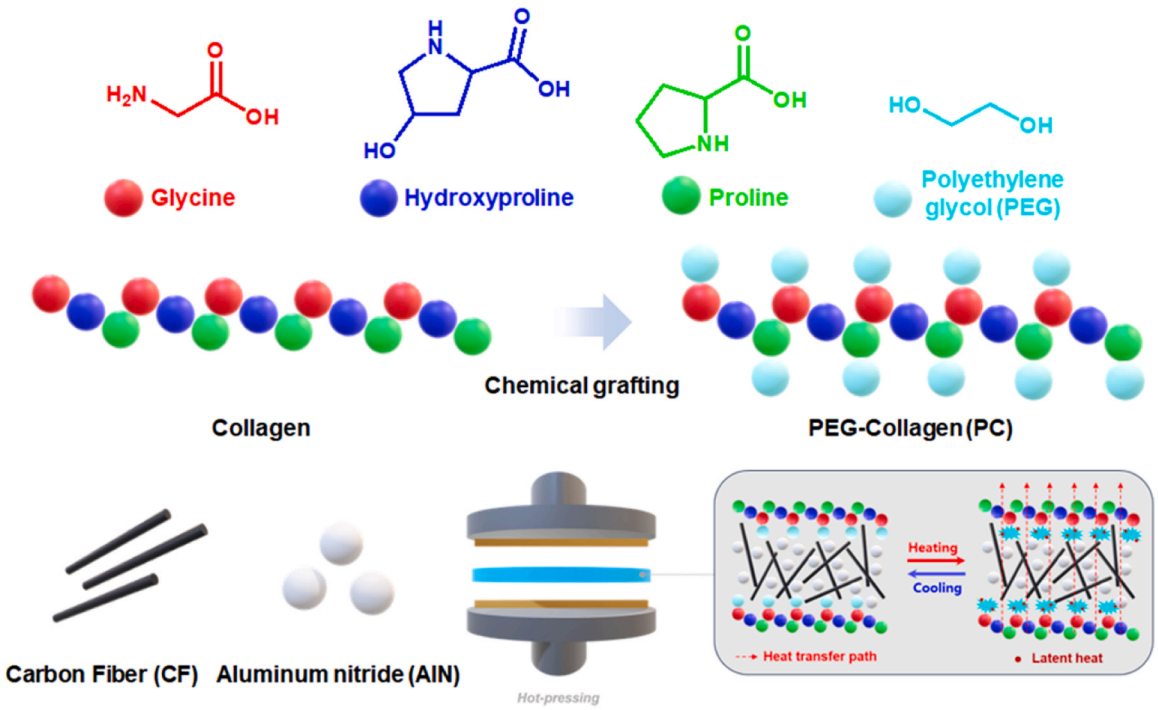


Fig. 1. Schematic illustration of the CP/CF/AlN composites.

amounts of heat. This unique characteristic provides them with an exceptional capacity to store thermal energy, making them highly effective at regulating temperature fluctuations in electronic devices (Cao et al., 2025a).

The integration of thermally conductive organic PCM-based composites into electronic thermal management offers clear advantages over traditional cooling methods. These materials exhibit improved thermal conductivity when undergoing phase transitions, facilitating effective heat transfer within densely packed electronic systems. Moreover, their ability to absorb and release heat at stable temperatures helps maintain a uniform temperature distribution across the components, enhancing overall thermal stability. This aids in eliminating hot spots and reducing thermal stress, thereby offering a reliable and uniform cooling solution for advanced electronic systems.

Recent research has increasingly focused on harnessing the capabilities of organic PCMs for electronic device applications. Efforts have been made to refine methods for synthesizing, characterizing, and integrating these materials, with the aim of optimizing their thermal properties and compatibility with existing device designs. To improve the thermal conductivity and mechanical durability of PCMs, studies have explored the addition of nanostructures and other additives to PCM matrices, thereby broadening their potential for high-performance electronics (Chen et al., 2024; Kharabati and Saedodin, 2024; Mai et al., 2025). However, despite their promising advantages, organic PCMs still face significant challenges that must be overcome before they can be widely implemented in electronic devices and thermal management solutions. These challenges include maintaining material integrity under harsh conditions such as mechanical strain, temperature fluctuations, and exposure to impurities. Additionally, assessing the practicality of large-scale production and the cost-effectiveness of these materials is crucial to determining their potential for widespread industrial adoption.

Another trend in PCM material research is related to bio-based materials. Biological macromolecules and biomass materials originate from diverse sources, including animals, plants, and microorganisms (Cao et al., 2025b). Bio-based PCM composites possess biodegradability, sustainability, and environmental friendliness, which has great potential as an eco-friendly material. Li et al. (2022) employed cellulase to effectively hydrolyze segmented cellulose in wood, resulting in the formation of abundant micropores. The resultant wood-derived porous carbon adsorbed PEG and exhibits good mechanical properties, high energy storage capacity (151.7 J/g), anisotropic thermal conductivity, and enhanced thermal stability. Zhu et al. (2022) prepared 3D porous carbon scaffolds consisting of SiC-coated biomass carbon fibers as a thermally conductive backbone and shape stabilizer for PW. The composite with a filler ratio of 10.4 wt% had a high thermal conductivity of 0.61 W/(mK) in the axial direction (>3 times that of PW) and 0.48 W/(mK) in the transverse direction. The composite also had good thermal cycling stability and high thermal storage capacity (186.0 J/g).

Accordingly, this research introduces a novel bio-derived PCM composite designed to provide improved thermal conductivity, enhanced mechanical strength, and stable latent heat retention. The material was synthesized through a reaction between collagen and polyethylene glycol (PEG), with carbon fiber (CF) and aluminum nitride (AlN) incorporated in varying proportions to establish a robust three-dimensional filler network (Composition information of the composites is presented in Table S1). The composite exhibits outstanding thermal stability, remaining structurally intact up to 140 °C without any signs of leakage. Performance evaluations in lithium-ion battery (LIB) modules confirmed its effectiveness in heat regulation. By combining efficient energy storage with heat dissipation, this advanced PCM composite holds great potential for next-generation thermal management technologies.

2. Results and discussion

2.1. Fabrication of the PCM composites and characterization of the CF–PEG matrix

Fig. 1 illustrates the fabrication process of the collagen-grafted PEG (CP) matrix and the oxidation process of the filler. The CP matrix was specifically designed to enhance the intrinsic weak properties of traditional PCMs. To address the issue of CF detachment caused by poor interfacial bonding, an esterification reaction was employed to create

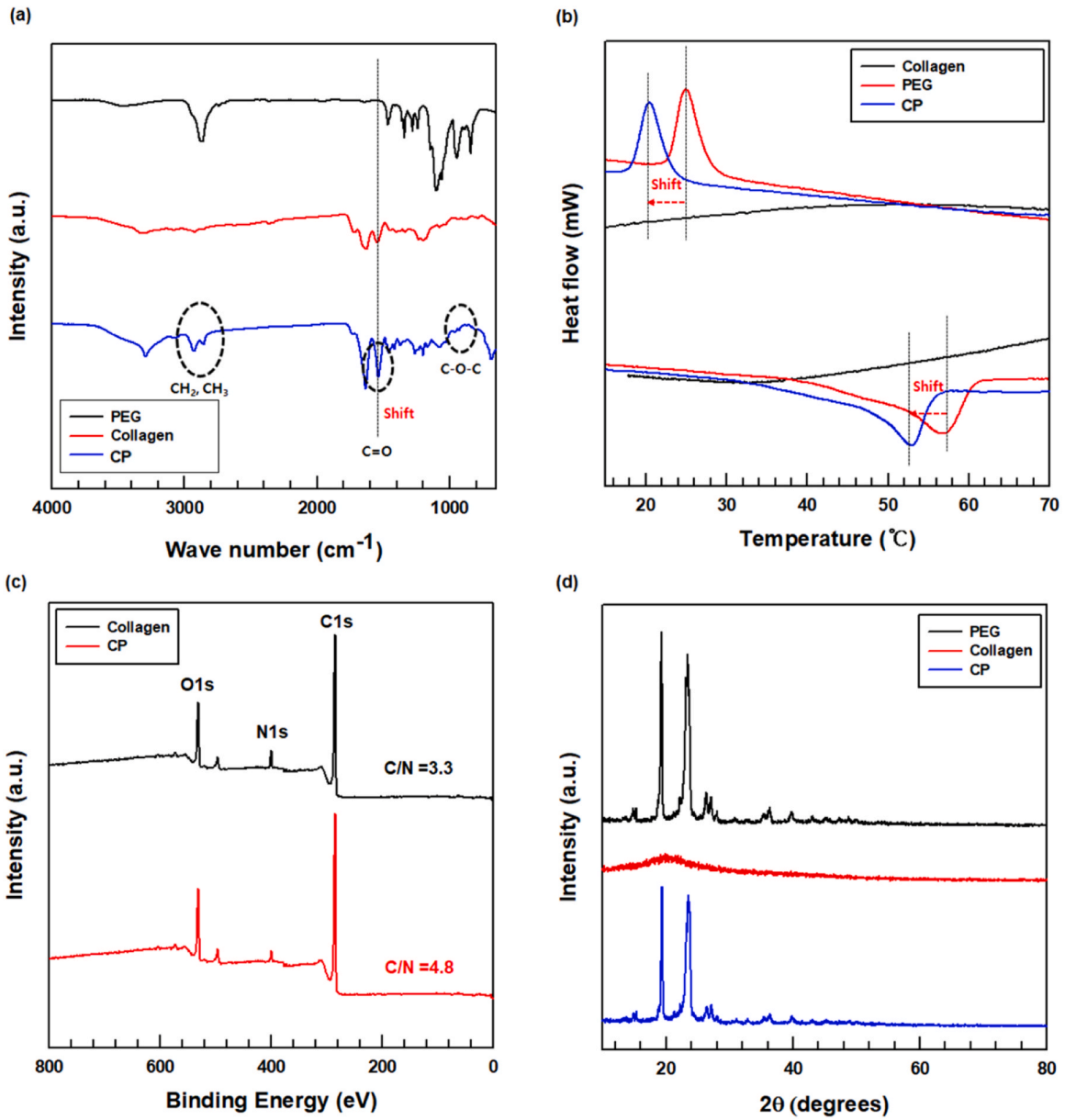


Fig. 2. (a) FT-IR spectra, (b) DSC curves, (c) XPS wide spectra, (d) XRD curves of the matrices during CP synthesis.

CF–PEG, which significantly improved the adhesion between the CF and the matrix.

Mechanism cross-linking mediated by glutaraldehyde can be explained through the reaction of the aldehyde functional groups with free nonprotonated ε-amino groups (-NH_2) through a nucleophilic addition-type reaction. The nucleophilic addition of an ε-NH_2 group of collagen to the carbonyl group (C═O) of an aldehyde forms a tetrahedral unstable intermediate called a carbinolamine. After that, the protonation of the -OH group is followed by the loss of a water molecule to form a conjugated Schiff base. Under these conditions, PEG is grafted onto collagen and then esterified (Farris et al., 2010).

To confirm the chemical change after synthesizing CF–PEG, FT-IR analysis was conducted, as depicted in Fig. 2(a). A sharp peak was observed at 1600 cm^{-1} , which is associated with the C═O stretching vibrations in the carboxyl groups of collagen. After esterification, this peak shifted to 1589 cm^{-1} , signaling the effective grafting of CF and PEG, in line with the expected esterification process.

To further investigate the changes in thermal behavior after synthesis, a DSC analysis was conducted. Fig. 2(b) provides a comparison of the endothermic temperature range of the materials based on CP synthesis. While PEG originally exhibited a melting point of 57 °C, the synthesized CP exhibited an endothermic transition between 42 °C and 55 °C. This decrease in melting temperature is likely due to a reduction in the number of carboxyl and hydroxyl groups during esterification, which previously formed hydrogen bonds that elevated the melting point of PEG. These results provide strong evidence that the CF and PEG were successfully integrated into the composite through the esterification process.

In XPS wide spectra (Fig. 2(c)), collagen has a abundant carbon, oxygen, and nitrogen derived from three types of protein such as glycine, hydroxyproline, and proline. After grafting process between PEG and collagen, due to the high carbon content in PEG, the C/N ratio of CP increased from 3.3 to 4.8 compared to collagen alone.

Meanwhile, despite collagen's amorphous structure, the typical crystalline structure of PEG within CP is maintained after CP synthesis (Fig. 2(d)). Therefore, the above analyses suggest that CP was successfully synthesized without any crystalline structure modification of PEG.

In the physically mixed collagen and PEG (Fig. 3(c)), there were two

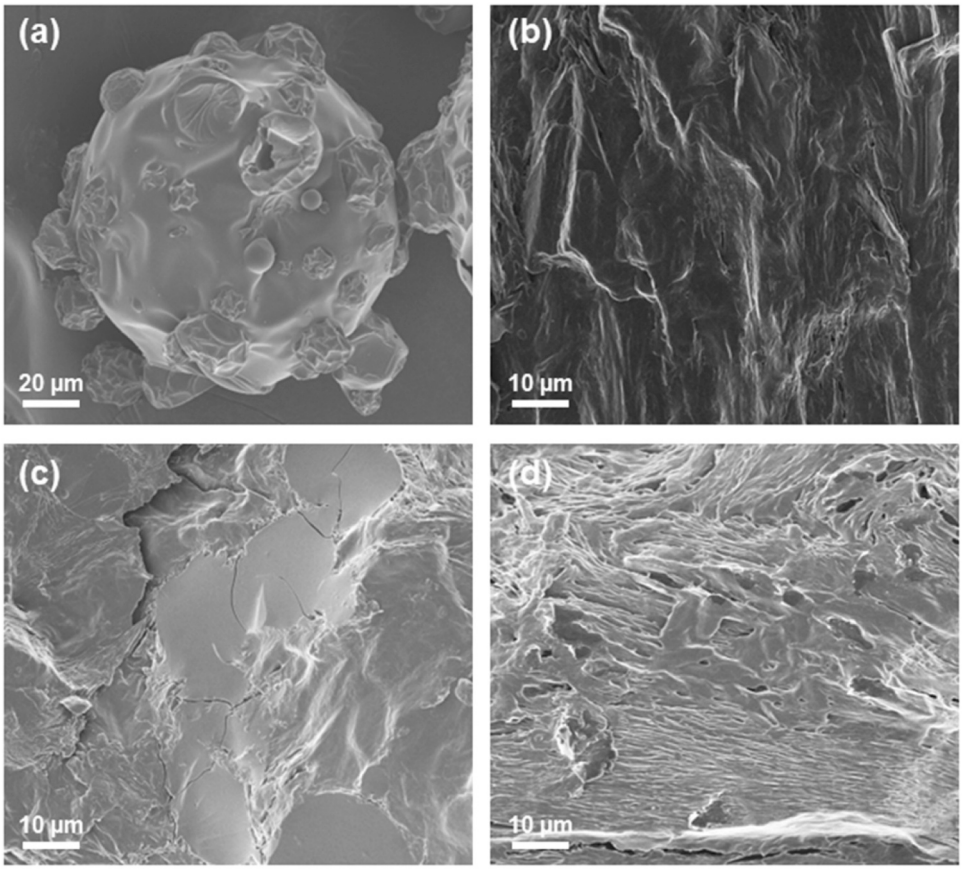


Fig. 3. Morphologies of (a) collagen, (b) PEG, (c) physically mixed collagen and PEG, and (d) CP.

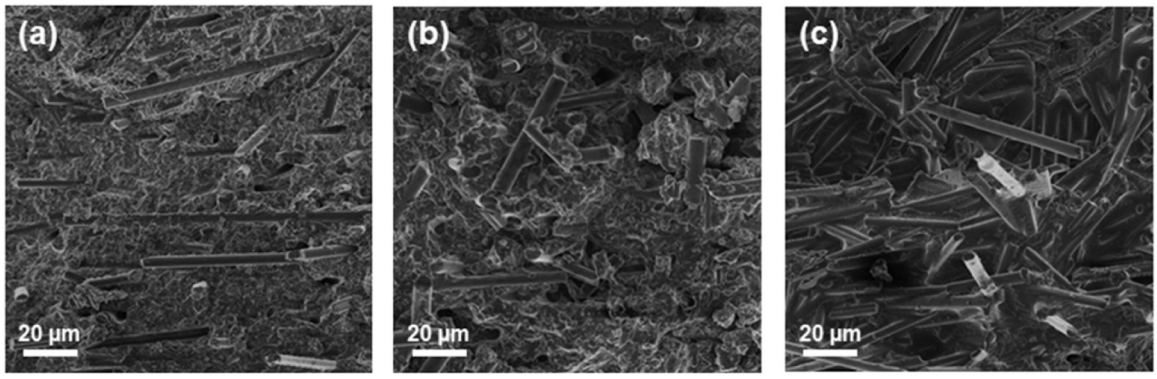


Fig. 4. Cross-sectional images of CP/CF/AlN with different CF-to-AlN filler ratios of (a) 2:1, (b) 1:1, and (c) 1:2.

separate phase interfaces comprising collagen (Fig. 3(a)) and PEG (Fig. 3 (b)). After the synthesis of CP, the samples exhibited homogeneous cross-sections without phase separation (Fig. 3(d)).

2.2. Morphology of the CP/CF/AlN composites

The CP matrix was reinforced with various ratios of thermally conductive fillers, CF (Fig. S1(a)) and AlN (Fig. S1(b)), as illustrated in Fig. 4. When the CF-to-AlN ratio was 2:1, the 2D CFs predominantly aligned along the in-plane direction (Fig. 4(a)), while the 1D AlN particles were uniformly distributed within the matrix. As the AlN content increased, the alignment of the CFs gradually shifted from the in-plane direction to a through-plane orientation. This shift was driven by the synergistic interaction between the 1D AlN particles and the 2D CFs (Fig. 6(b, c)). When the CF-to-AlN ratio reached 1:2, the CFs aligned nearly vertically, creating efficient heat-transfer pathways in the perpendicular direction.

The leakage tests demonstrated that the composite exhibited a significant improvement in structural stability under heat exposure, as shown in Fig. 5. To quantitatively assess sample leakage, calculations were performed using Eqs. (1) and (2):

$$\varepsilon = m_r/m_0 \times 100\%, \quad (1)$$

$$\gamma = (m_0 - m_r)/m_0 \times 100\%, \quad (2)$$

where ε represents the quality retention rate of the PCM, while γ denotes the leakage rate (Hu et al., 2023) (Fig. 6). Leakage and retention rates serve as key indicators of the resistance of a PCM to seepage. The retention rate of PEG decreased sharply once the drying oven

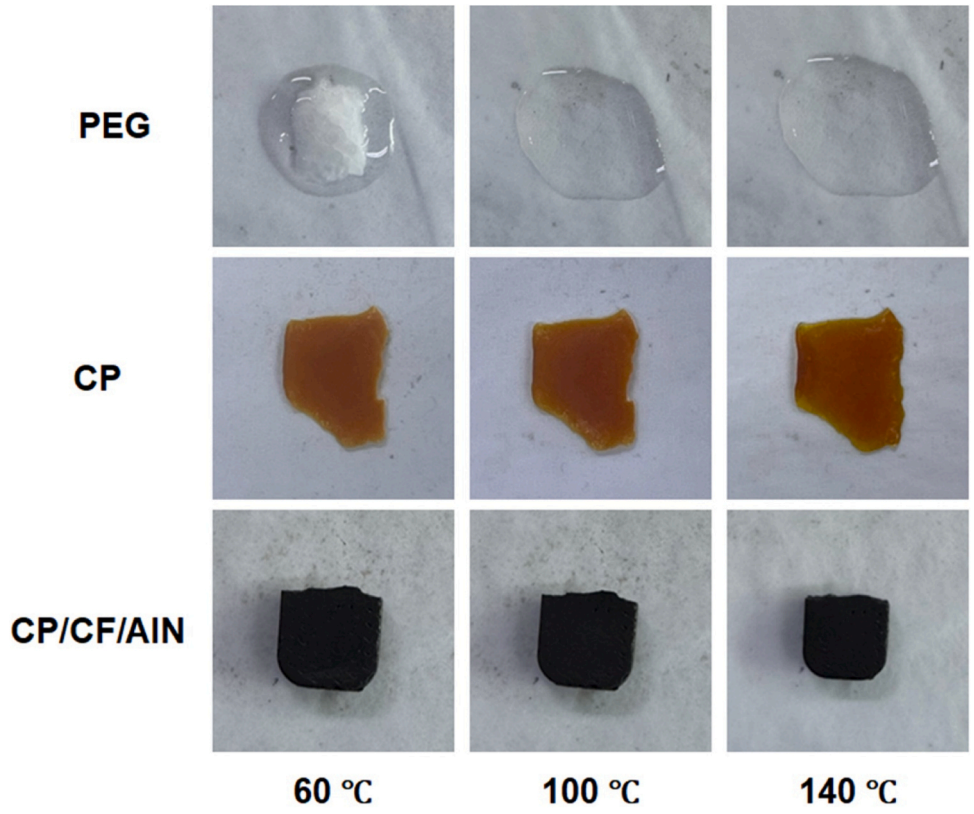


Fig. 5. Leakage test of PEG, CP, and CP/CF/AlN.

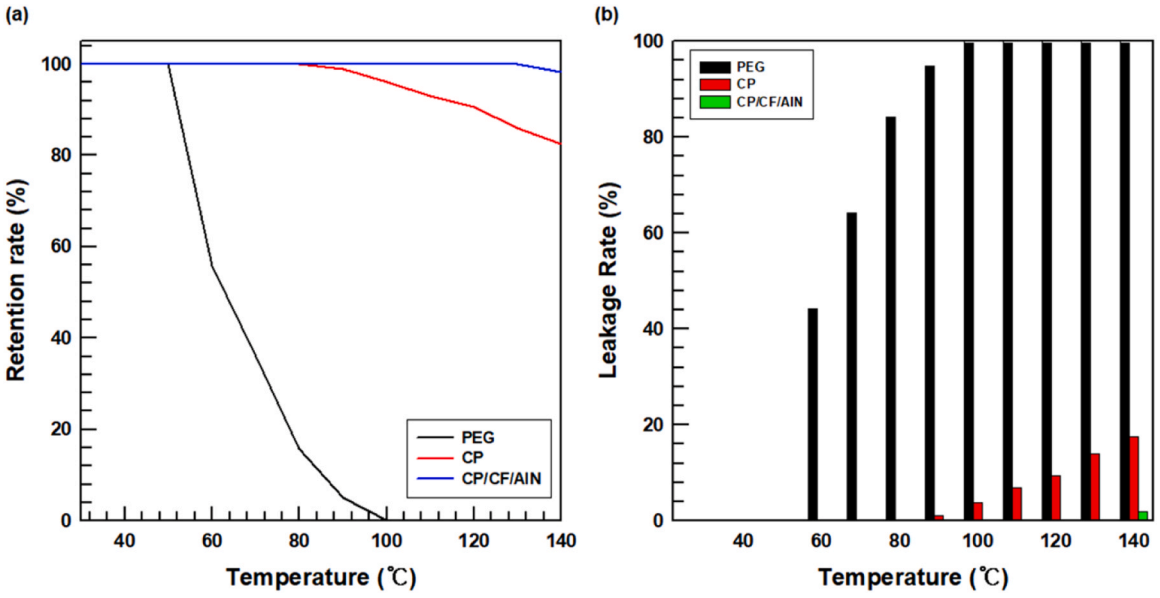


Fig. 6. (a) Retention rate and (b) leakage rate of PEG, CP, and CP/CF/AlN, as determined through leakage tests.

temperature exceeded 60 °C, which is its melting point. At 100 °C, PEG fully liquefied and lost its structural integrity entirely, resulting in a retention rate of 0 %.

In contrast, the CP composite maintained its stability up to 90 °C, exhibiting retention rates of 98.8 % at 100 °C and 82.4 % at 140 °C. The CP/CF/AlN composite exhibited even greater thermal resistance, with only minimal reduction in retention, maintaining a retention rate of 98.16 % at 140 °C. These results indicate that the chemical cross-linking between collagen and PEG greatly enhanced the thermal stability of the material.

2.3. Thermal properties and heat management performance of the PCM composites

The density, heat capacity, and thermal diffusivity of the CP/CF/AlN composites were thoroughly analyzed based on the different filler loadings and fabrication techniques. Thereafter, the through-plane thermal conductivity of the composite was evaluated. As shown in

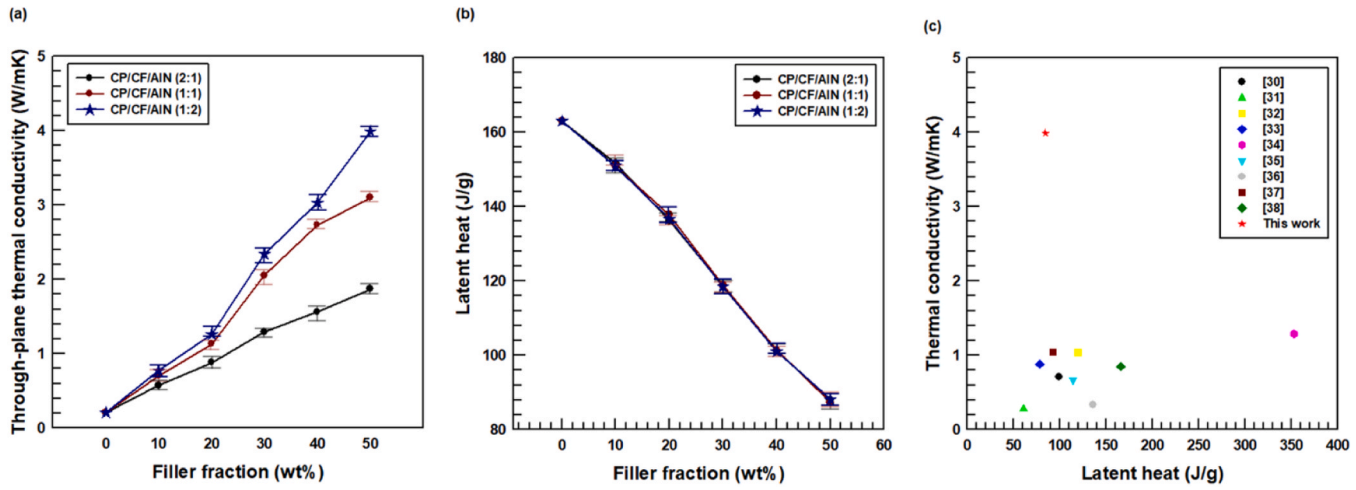


Fig. 7. (a) Thermal conductivity, (b) latent heat, and (c) comparison with previously reported values (Min et al., 2024; Zhang et al., 2021a, 2021b; Liu et al., 2021b; Zhou et al., 2022; Chibani et al., 2021; Wang et al., 2020; Puig et al., 2017; Nazeer et al., 2019).

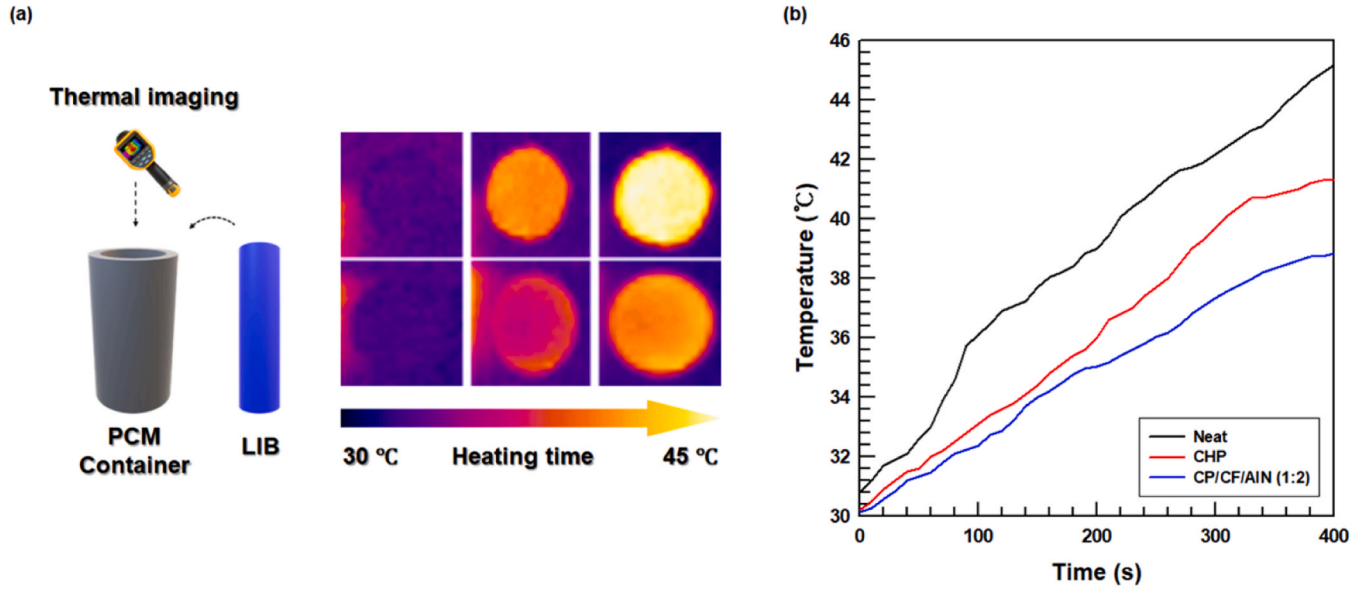


Fig. 8. Results of thermal management tests on an LIB module.

Fig. 7(a), the thermal conductivity varied depending on the fabrication technique. When 50 wt% CF and AlN were used as fillers in a 1:2 ratio, the thermal conductivity of the composite reached a maximum of 3.99 W/mK, which was improved by a whopping 1800 % compared to pure PEG (0.2 W/mK). On the other hand, the composites with filler ratios of 1:1 (CF:AlN) and 2:1 (CF:AlN) showed lower thermal conductivities of 3.1 and 1.87 W/mK, respectively. These results imply that as the amount of 2D-shaped CFs increases, the alignment in the in-plane direction is strengthened, resulting in lower vertical thermal conductivity, whereas as the amount of particle-shaped AlN increases, the CFs between adjacent AlNs are aligned vertically, resulting in higher thermal conductivity.

Furthermore, the latent heat of the composites, a vital property for thermal energy storage in PCM applications, was measured, and the results are presented in Fig. 7(b) and S2. In the case of latent heat, unlike thermal conductivity, a similar level of latent heat was observed at all filler ratios.

Although compounding fillers into PCMs often leads to a reduction in their latent heat capacity, the hot-pressing technique enabled the composites to effectively retain an impressive latent heat capacity of 88.1 J/g by minimizing structural defects. Fig. 7(c) presents a comparison of the thermal conductivity and latent heat of the composites using various PCM matrix with those reported in recent studies. Achieving an optimal balance between thermal conductivity and latent heat retention is a significant challenge. Notwithstanding, the CP/CF/AlN composites exhibited exceptional through-plane thermal conductivity while maintaining competitive latent heat storage. This unique combination of efficient heat absorption and dissipation positions these materials as strong candidates for next-generation thermal management applications.

Fig. 8 illustrates the heat management capabilities of the CP/CF/AlN composites, which were evaluated using an infrared camera. The performance of these advanced composites was compared with that of standard commercial heat dissipation pad (CHP) materials. In typical heat dissipation systems, materials with high thermal conductivity usually exhibit a rapid temperature rise. During a 400-second exposure to a heat source (LIB), the PCM composite experienced a faster temperature increase than the CHP owing to its superior thermal conductivity (3 W/mK). Despite this, the PCM composite demonstrated superior cooling performance. Specifically, it kept the LIB temperature

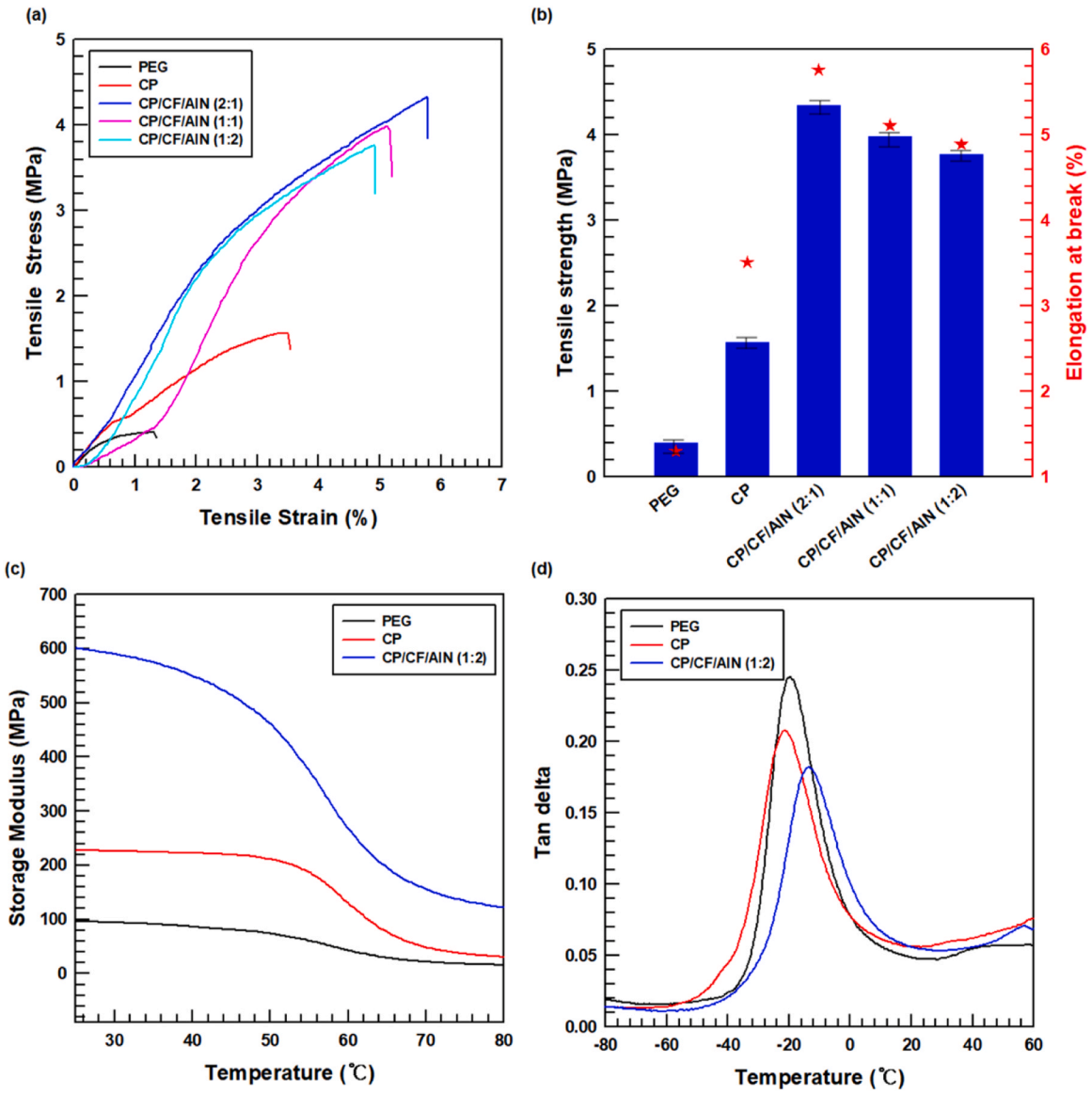


Fig. 9. Mechanical properties of the composites: (a,b) Tensile stress and strain, (c) Storage modulus, (d) Tan δ.

3.4 °C lower than that achieved with the CHP and 4.5 °C lower than the temperature of the untreated LIB. These findings suggest that the thermally conductive PCM-based composites exhibit significant potential for efficient thermal management in LIBs and could outperform traditional heat dissipation materials.

2.4. Mechanical properties of the PCM composites

The mechanical properties of the CP/CF/AlN composites were measured using a universal testing machine, as depicted in Fig. 9. The introduction of fillers into the composite matrix substantially increased the tensile strength from 0.4 MPa to 3.77 MPa and the strain from 1.3 % to 4.89 %. These enhancements are primarily due to the inherent brittleness of CF and AlN fillers. In addition, contrary to the trend of thermal conductivity data, the higher the CF ratio, the better the mechanical properties were exhibited. This is because the more dominant the CF ratio, the more the CF in the form of 2D fibers is arranged in the in-plane direction, forming a more mechanically stable structure. The structural reinforcement of the composite may also be due to significant contributions from the well-integrated in-plane CF network.

Fig. 9(c) and (d) explore how different composite materials respond to temperature and mechanical stress, using dynamic mechanical analysis (DMA) as the tool for investigation. These figures focus on two important indicators: the storage modulus, which measures a material's stiffness, and the glass transition temperature, revealed through the tan δ peak. As expected, pure PEG showed the lowest stiffness at room temperature, with a storage modulus of about 100 MPa. However, when collagen was grafted onto the PEG structure, a noticeable increase in rigidity was observed. This enhancement became even more pronounced with the addition of CF and AlN particles, especially at lower temperatures.

In Fig. 9(d), the thermal behavior of the composites was changed. Composites containing CF and AlN exhibited a rightward shift in the tan δ peak, signaling an increase in glass transition temperature compared to neat PEG. On the other hand, the CP that lacked filler particles showed the opposite trend, with the tan δ peak moving to the left—suggesting a decrease in their glass transition temperature. This contrasting behavior highlights the significant impact of filler materials on the thermal and mechanical stability of polymer composites.

Owing to these superior mechanical properties, the CP/CF/AlN composite stands out as a highly robust material for thermal management applications, offering higher strength and durability than

conventional thermally conductive PCM composites.

3. Conclusions

In conclusion, this study introduces an advanced thermally conductive PCM designed to overcome the shortcomings of conventional PCM composites, including poor thermal conductivity and limited mechanical strength. The composite matrix was synthesized through esterification, and a robust continuous filler network was constructed by combining CF and AlN. The resulting CP/CF/AlN composite demonstrated excellent thermal conductivity along the perpendicular direction, enhanced mechanical durability, and a substantial latent heat capacity. This composite exhibits an endothermic response between 42 °C and 55 °C, enabling it to effectively regulate temperature fluctuations in electronic devices and prevent overheating. This work highlights the tremendous potential of thermally conductive composites as high-performance solutions for efficient thermal management and heat dissipation in advanced electronic applications.

CRediT authorship contribution statement

Park Min: Data curation, Formal analysis, Methodology, Validation, Writing – review & editing. **Kim Jooheon:** Supervision, Project administration, Funding acquisition. **Su Pei-Chen:** Formal analysis, Data curation. **Lee Wondu:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, 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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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.indcrop.2025.121150.

Data availability

The data that has been used is confidential.

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