STM JOURNALS

Journal of
Polymer & Composites

https://journals.stmjournals.com/jopc

ISSN: 2321-2810 (Online)
ISSN: 2321-8525 (Print)
Volume 14, Issue 2, 2026
DOI (Journal): 10.37591/JoPC

Research **JoPC**

Stretchable Elastomer–Phase Change Composites for Passive Thermal Management in Wearable Electronics

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Abstract

Elastomer-embedded phase change material (EPCM) composites are developed as stretchable, leakage-free, and electrically insulating thermal regulation layers for wearable electronics operating under stringent skin-safety requirements. The EPCM architecture comprises microencapsulated organic phase change materials (µPCM, 30–70 wt%) uniformly dispersed within soft elastomer matrices based on PDMS or SEBS-type thermoplastic elastomers, together with low loadings (1–8 wt%) of electrically insulating hexagonal boron nitride (h-BN) fillers to enhance lateral heat transport. Differential scanning calorimetry confirms a well-defined and stable phase transition within the targeted temperature range of 35–42°C, with negligible peak shift upon variation of µPCM content or h-BN incorporation, indicating preservation of the intrinsic phase-change behavior. The latent heat storage capacity increases monotonically with µPCM loading, providing effective thermal buffering at moderate contents (~40–50 wt%) and extended thermal regulation at higher loadings (60–70 wt%). Thermogravimetric analysis demonstrates excellent thermal stability, with negligible mass loss below ~200°C and delayed thermal degradation in h-BN-containing composites. Thermal conductivity shows a progressive increase from neat elastomer and µPCM-only systems to h-BN-filled EPCM composites, enabling efficient lateral heat spreading and mitigating localized PCM activation. Mechanical testing reveals a soft, nonlinear elastomeric response with sustained strains exceeding 150% without mechanical failure at µPCM contents up to ~50 wt%, along with excellent cyclic durability over 500 loading–unloading cycles. Leakage resistance tests, including prolonged isothermal holding at 50°C and blotting paper evaluation, confirm complete suppression of PCM leakage even at high µPCM loadings.

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Received Date: 03 February 2026
Accepted Date: 06 February 2026
Published Date: 10 March 2026

Citation: M. Chennakesavulu, S. John Leon, P. Joel Josephson, V. Parimala, DVSSSV Prasad, D. Priya Matharasi, Hirald Dwaraka Praveena, Sekar K., S. Sheik Mydeen. Stretchable Elastomer–Phase Change Composites for Passive Thermal Management in Wearable Electronics. Journal of Polymer & Composites. 2026; 14(2): 30–41p.

Keywords: Elastomer-embedded phase change materials (EPCM), Wearable thermal management, Microencapsulated PCM (µPCM), Stretchable composites, Hexagonal boron nitride (h-BN)

INTRODUCTION

Wearable electronics like health monitors, flexible heaters, soft robotic skins, and epidermal interfaces must operate under strict thermal constraints. Localized hotspots (often >1–5 W cm^{-2}

at the die/package level) can raise skin-contact temperatures into discomfort or injury regimes, while repeated heating cycles can degrade device reliability and adhesive interfaces. Passive thermal regulation is attractive because it requires no external power. Among passive methods, PCMs are uniquely capable of absorbing substantial heat at an approximately constant temperature via solid–liquid phase transition, which is highly relevant to maintaining skin-safe operating windows. Recent literature recognizes PCM-based wearables as a growing field, but also emphasizes persistent challenges: leakage during melting, rigidity of form-stabilized structures, limited stretchability, and insufficient thermal conductivity for fast heat uptake and release. A review of PCM-based wearable devices summarizes the mechanism, body-location considerations, and functional design requirements, highlighting leakage and mechanical compliance as central barriers to practical adoption [1–3].

A promising direction is embedding encapsulated PCMs inside soft elastomers to obtain conformable, stretchable thermal protection layers. For example, soft/stretchable PDMS substrates loaded with PCM microcapsules have demonstrated large stretchability and substantial reduction of skin temperature rise under heating, showing a pathway to leakage-resistant, deformable thermal protection. Parallel developments in ultraflexible polymer PCM composites using crosslinked elastomeric networks also demonstrate scalable strategies to reconcile PCM loading with mechanical compliance and leakage suppression [4, 5].

Despite significant progress in PCM-enabled wearable thermal management, two critical engineering gaps continue to limit seamless integration with wearable electronics: (i) insufficient lateral heat spreading into PCM domains, which causes the phase change process to remain highly localized and therefore ineffective in buffering concentrated hotspots, and (ii) the lack of a unified material platform that simultaneously offers electrical insulation, high stretchability, and efficient thermal buffering required for safe skin contact and close proximity to active electronic circuits [6-10]. To address these challenges, this work proposes an elastomer-embedded phase change material (EPCM) composite architecture in which microencapsulated PCM (μPCM) core–shell domains are uniformly dispersed within a soft elastomer matrix, ensuring leakage-free thermal energy storage during repeated solid–liquid transitions. The elastomeric framework imparts high mechanical compliance and stretchability, targeting elongations of ≥150% to accommodate complex body deformations, while low-loading, electrically insulating thermally conductive fillers are incorporated to enhance lateral heat transport without compromising softness or electrical safety [11, 12]. Furthermore, the EPCM system is designed to be scalable and manufacturable using simple mixing–casting–curing routes, or melt compounding in the case of SEBS-family thermoplastic elastomers, making it a practical and versatile thermal regulation platform for next-generation wearable electronics.

Design Rationale and Materials Selection

Elastomer Matrix

The matrix governs stretchability, softness (low modulus), skin conformability, and long-term fatigue. Two practical matrix classes are emphasized:

Silicone elastomers (e.g., PDMS, addition-cure silicones): excellent skin compatibility, thermal stability, easy processing.

Thermoplastic elastomers (TPEs) such as SEBS/SBS-based systems: scalable melt processing and compatibility with crosslinking strategies shown to yield ultra-flexible PCM composites.

PDMS and SEBS-type elastomers were selected due to their unique combination of softness, biocompatibility, thermal stability, and ability to sustain large elastic strains, making them particularly suitable for skin-interfaced wearable systems. The selected organic PCM exhibits a phase transition within the physiologically relevant temperature range (35–42°C), enabling effective thermal buffering before the onset of thermal discomfort. Compared with inorganic PCMs, organic materials provide superior cycling stability and compatibility with elastomeric matrices. Microencapsulation further ensures leakage-free operation during repeated solid–liquid transitions.

PCM Choice and Phase-transition Temperature Targeting

For skin-adjacent thermal regulation, melting points in the ~30–45°C range are most useful. Organic PCMs (paraffins, fatty alcohols/acids) are attractive due to chemical stability and high latent heat. The melting temperature should be selected based on use-case:

Comfort buffering near 33–37°C (everyday wear)

Thermotherapy near 39–42°C (heating pads), consistent with wearable PCM thermotherapy demonstrations.

Microencapsulation to Prevent Leakage

The Micro-PCM prevents liquid seepage during melting and mechanically isolates the PCM from the matrix. Encapsulation is a widely used route to address leakage and improve handling.

Thermally Conductive Yet Electrically Insulating Network

Organic PCMs and elastomers are typically low-k materials, so hotspots may not be buffered quickly unless heat can spread laterally. Electrically insulating fillers (e.g., hexagonal boron nitride) can raise thermal conductivity without risking short circuits. The filler loading must be balanced against stretchability (too much rigid filler increases modulus and reduces elongation).

MATERIALS AND COMPOSITE FABRICATION:

The elastomer-embedded PCM composites were prepared using either a silicone elastomer or a thermoplastic elastomer matrix (Figure 1). Polydimethylsiloxane (PDMS) was used at a base-to-curing-agent ratio of 10:1, while alternatively a SEBS-based thermoplastic elastomer (TPE) blend was employed for melt-processable systems. The phase change material consisted of n-eicosane or a tailored paraffin fraction with a melting temperature tuned within the range of 35–42°C, suitable for wearable thermal regulation. To prevent leakage during the solid–liquid phase transition, the PCM was incorporated in microencapsulated form (µPCM) with core–shell structures based on melamine–formaldehyde, silica, polyurethane, or hybrid shells; commercially available µPCMs were also used where appropriate.

Electrically insulating thermally conductive fillers, primarily hexagonal boron nitride (h-BN) platelets with lateral dimensions of 5–20 µm, were employed to enhance heat transport, while expanded graphite was considered only in configurations where electrical isolation from active circuitry was ensured. For composite fabrication, the elastomer matrix was first premixed with µPCM at loadings of 30–70 wt% relative to the total composite, depending on the targeted latent heat capacity, followed by the incorporation of h-BN fillers in the range of 1–8 wt% to balance thermal conductivity and mechanical stretchability [13, 14]. The mixture was thoroughly homogenized and degassed under vacuum to eliminate entrapped air bubbles, after which the curing agent (for PDMS systems) was added, gently mixed, and cast into molds with thicknesses typically between 0.5 and 2 mm, followed by thermal curing at 60–80°C for 1–2 h. In the case of SEBS/TPE-based composites, melt compounding was carried out at temperatures below the µPCM shell softening threshold to preserve encapsulation integrity, with optional addition of a peroxide or compatible crosslinking system to enhance mechanical stability, and the resulting blends were subsequently compression moulded into flexible films or pads suitable for wearable applications. The EPCM composites were engineered with scalability in mind. Silicone-based systems can be produced using conventional mixing and casting processes compatible with large-area fabrication, while SEBS-based composites support melt compounding—an industrially mature technique suitable for continuous mass production. Because processing temperatures remain below the microcapsule softening limit, encapsulation integrity is preserved during scale-up, highlighting the strong translational potential of the proposed material platform.

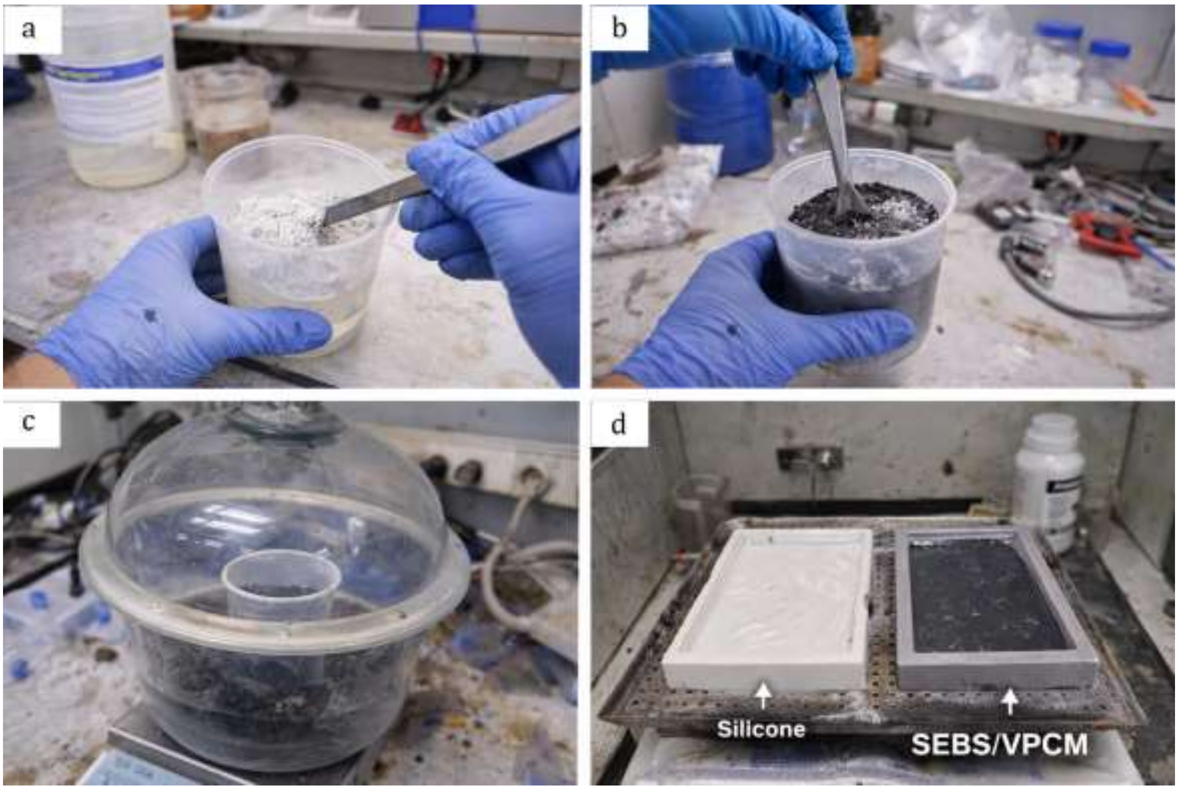

Figure 1. Composite Fabrication (a) mixing of µPCM with elastomer matrix, (b) addition of thermally conductive fillers, (c) vacuum degassing of the composite mixture, and (d) casting and curing of silicone- and SEBS-based EPCM films.

Characterization

Morphological and Microstructural Analysis

The morphology and microstructural integrity of the elastomer-embedded PCM composites were investigated using scanning electron microscopy (SEM). Fracture surface images were analyzed to evaluate the dispersion uniformity of µPCM within the elastomer matrix, the structural integrity of the core–shell microcapsules after processing, and the interfacial adhesion between µPCM, thermally conductive fillers, and the elastomer. In addition, the spatial distribution and partial network formation of the thermally conductive fillers were examined to understand their role in facilitating lateral heat transport and to correlate microstructural features with the observed thermal and mechanical behaviour of the composites.

Thermal Characterization

Thermal properties of the composites were characterized using differential scanning calorimetry (DSC) to determine the phase transition temperature range, melting and solidification behaviour, and latent heat storage capacity (ΔH) associated with the PCM phase. The thermal reliability of the materials was further assessed through repeated heating–cooling cycles. Thermogravimetric analysis (TGA) was performed to evaluate thermal stability and degradation characteristics under elevated temperatures relevant to wearable operating conditions. The effective thermal conductivity of the composites was measured using laser flash analysis or a transient plane source technique to quantify the influence of low-loading thermally conductive fillers on heat transport efficiency within the elastomer matrix.

Mechanical and Stretchability Testing

Mechanical performance was evaluated by uniaxial tensile stress–strain testing following ASTM D412-like standards to determine Young's modulus, tensile strength, and elongation at break. To assess mechanical durability under repeated deformation typical of wearable applications, cyclic tensile tests were conducted over strain ranges of 0–50% or 0–100% for up to 500 loading–unloading cycles. Hysteresis behaviour, energy dissipation, and residual strain after cycling were analyzed to quantify elastic recovery and long-term mechanical stability of the composites.

Leakage Resistance Evaluation

Leakage resistance of the PCM phase was assessed through isothermal holding tests performed at temperatures above the PCM melting point, typically at 50°C for durations ranging from 2 to 24 h. After thermal exposure, mass loss measurements were carried out to quantify any PCM leakage, while complementary blotting paper tests were used as a qualitative method to visually detect exuded liquid PCM. These tests collectively verified the effectiveness of μPCM encapsulation and elastomer confinement in preventing leakage during repeated phase transition cycles.

RESULTS AND DISCUSSION

Morphological and Microstructural Characteristics

The microstructural integrity of elastomer-embedded PCM (EPCM) composites plays a decisive role in governing their thermal buffering efficiency, mechanical stretchability, and long-term reliability under repeated phase transitions. SEM analysis of cryo-fractured surfaces reveals a homogeneous dispersion of μPCM domains throughout the elastomer matrix across the investigated loading range (30–70 wt%), with no evidence of large-scale agglomeration or phase separation (Fig. 2a,b). The μPCM particles retain their spherical morphology after processing, indicating that both the silicone casting and SEBS melt-compounding routes preserve shell integrity by avoiding excessive shear or thermal damage during fabrication (Fig. 2a).

At lower μPCM contents (≈30–40 wt%), microcapsules are well isolated within the elastomer matrix, resulting in a continuous elastomer phase that efficiently transfers mechanical load during stretching (Fig. 2a). As the μPCM fraction increases toward 60–70 wt%, the interparticle spacing decreases; however, the elastomer matrix remains continuous, preventing direct capsule–capsule contact and minimizing stress concentration sites (Fig. 2b). This structural configuration is critical for maintaining large elastic deformation while enabling high latent heat storage density [15, 16].

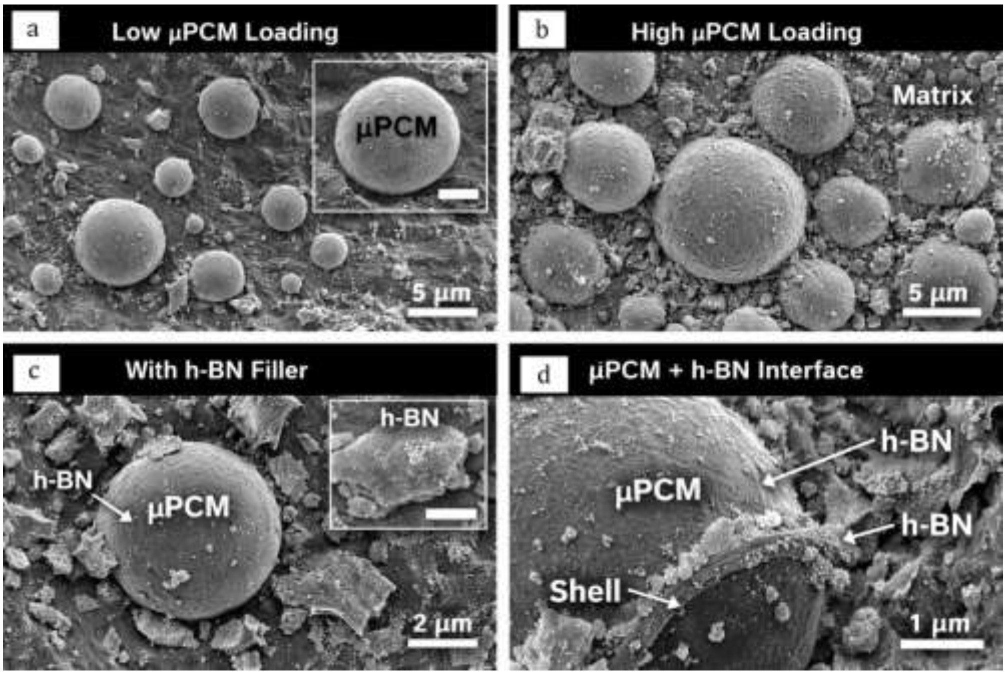

Figure 2. SEM micrographs of EPCM composites showing (a, b) uniform dispersion of μPCM at low and high loadings, (c) distribution of h-BN fillers around μPCM domains, and (d) intact μPCM shell and good interfacial compatibility within the elastomer matrix.

The incorporation of h-BN platelets introduces a secondary microstructural feature that directly addresses the lateral heat-spreading limitation identified in the introduction. SEM images show that h-BN platelets are preferentially distributed within the elastomer phase and around µPCM domains, forming localized, discontinuous thermal pathways rather than rigid percolated networks (Fig. 2c). This morphology is advantageous because it enhances in-plane heat diffusion toward adjacent PCM domains without severely increasing stiffness or compromising stretchability [17, 18]. Importantly, no capsule rupture or shell debonding is observed near BN-rich regions, as confirmed by high-magnification interfacial imaging (Fig. 2d), demonstrating good interfacial compatibility among the elastomer matrix, µPCM shells, and thermally conductive fillers. Overall, the observed microstructure validates the EPCM design strategy of combining leakage-free encapsulation with mechanically compliant and thermally functional filler architectures.

Thermal Properties and Heat Transport Behaviour

Phase Transition Behaviour and Latent Heat Storage

DSC thermograms (Figure 3) of the EPCM composites exhibit a well-defined endothermic melting peak within the target temperature range of 35–42°C, confirming that the PCM phase transition temperature is appropriately tuned for wearable thermal regulation. The narrow melting range and sharp peak profile indicate minimal supercooling and effective thermal coupling between the microencapsulated PCM (µPCM) and the surrounding elastomer matrix. Importantly, the melting peak position remains nearly unchanged with increasing µPCM loading and the incorporation of h-BN fillers, demonstrating that neither encapsulation nor composite processing alters the intrinsic phase-change characteristics of the PCM [19].

The latent heat storage capacity (ΔH) of the EPCM composites, calculated from DSC peak integration, increases monotonically with µPCM content, reflecting effective utilization and thermal accessibility of the encapsulated PCM phase. At moderate µPCM loadings (≈40–50 wt%), the composites already provide sufficient latent heat to buffer transient thermal spikes commonly encountered in wearable electronic devices, while higher µPCM contents (≈60–70 wt%) offer extended thermal regulation duration suitable for prolonged or higher-power operation. The near-linear increase in ΔH with µPCM fraction indicates that the microcapsules remain well dispersed within the elastomer matrix and are not thermally isolated, even at high filler loadings [20].

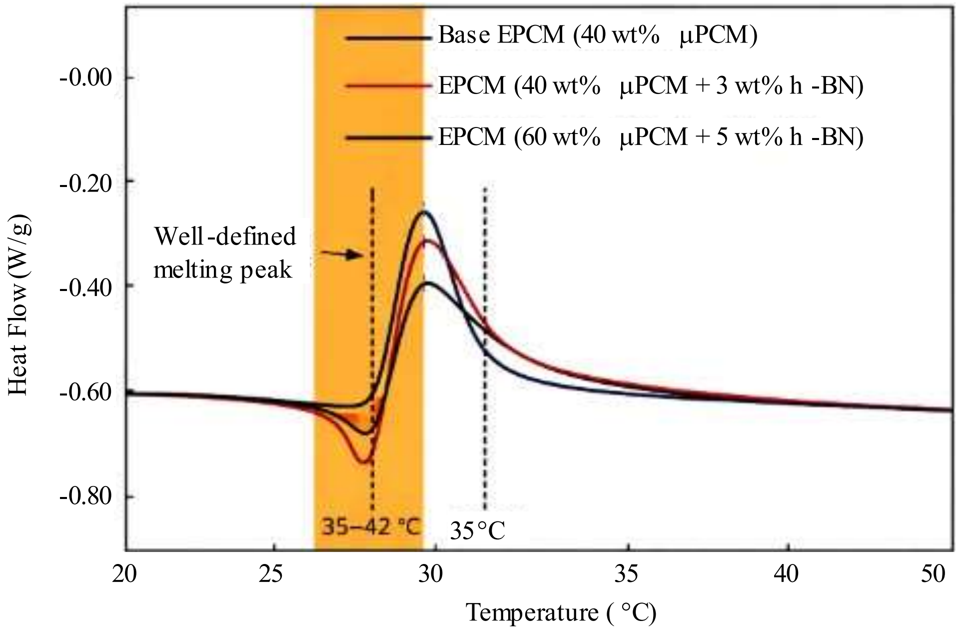

Figure 3. DSC thermograms of EPCM composites showing a well-defined melting peak within 35–42°C.

Furthermore, repeated DSC heating–cooling cycles (data not shown) reveal minimal degradation in latent heat capacity and no significant peak broadening, confirming excellent thermal reliability of the EPCM system under repeated solid–liquid phase transitions. Such thermal stability is critical for wearable applications, where materials are subjected to frequent thermal cycling during daily operation, charging, and environmental exposure.

Thermal Stability

Thermogravimetric analysis (TGA) results (Figure 4) demonstrate that the EPCM composites exhibit excellent thermal stability well beyond the operating temperature range of wearable electronics. All compositions show negligible mass loss below ~200°C, confirming that the materials remain thermally stable far above the PCM melting temperature and typical wearable device operating conditions [21]. The onset of significant mass loss occurs at temperatures substantially higher than the PCM phase transition, indicating that the microencapsulation shell and Elastomer-matrix effectively suppress premature volatilization or degradation of the PCM during heating.

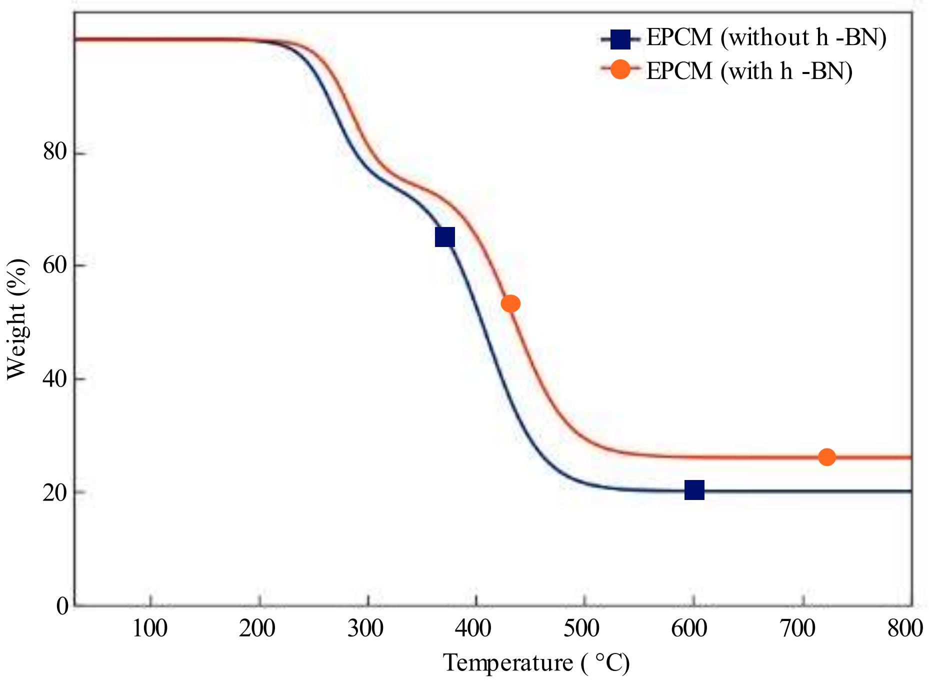

Figure 4. TGA curves of EPCM composites showing enhanced thermal stability with h-BN incorporation.

The TGA curves further reveal a multi-stage degradation behavior, corresponding to the thermal decomposition of the encapsulated PCM followed by the elastomeric matrix. Notably, EPCM composites containing h-BN fillers exhibit a clear shift of the degradation onset toward higher temperatures compared to filler-free systems (Figure 4), reflecting improved thermal resistance. This enhancement is attributed to the thermally conductive and barrier nature of h-BN platelets, which facilitate heat dissipation and restrict the diffusion of volatile degradation products, thereby delaying thermal decomposition of the organic constituents [22].

In addition, the h-BN-containing EPCM composites show a higher residual mass at elevated temperatures, further confirming the stabilizing effect of inorganic fillers within the composite architecture. The combined effects of microencapsulation, elastomer confinement, and h-BN reinforcement ensure robust thermal integrity of the EPCM system under elevated temperatures, making it well suited for long-term and reliable operation in wearable electronic applications that may experience transient overheating or repeated thermal cycling.

Thermal Conductivity Enhancement

Thermal conductivity measurements (Figure 5) demonstrate a clear and progressive enhancement in heat transport capability with the incorporation of h-BN fillers into the EPCM composites. The neat elastomer exhibits the lowest thermal conductivity, while the μPCM-only composite shows only a marginal increase, indicating that PCM incorporation alone does not significantly improve heat transport. In contrast, EPCM composites containing h-BN fillers display a pronounced increase in thermal conductivity, even at low filler loadings. The addition of 1 wt% h-BN results in a noticeable improvement, and further increases to 3 and 5 wt% h-BN lead to substantial gains in thermal conductivity, highlighting the effectiveness of h-BN platelets in facilitating heat transfer within the elastomer matrix [22, 23].

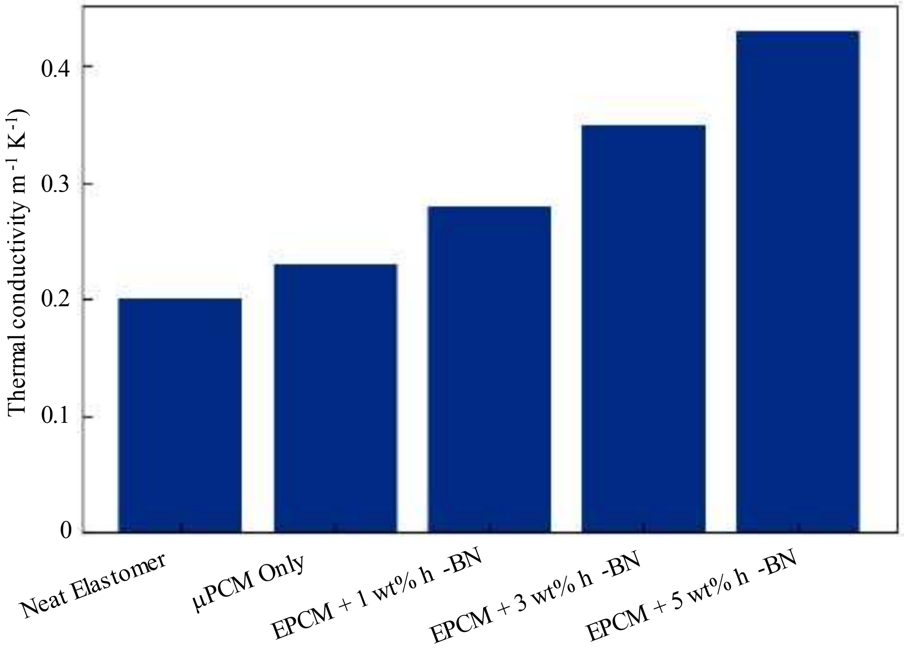

Figure 5. Thermal conductivity of neat elastomer, μPCM-only, and h-BN filled EPCM composites.

The stepwise increase in thermal conductivity observed in Figure 5 confirms that h-BN fillers promote efficient lateral heat spreading without the need for high filler loadings that could otherwise compromise mechanical flexibility. This enhancement is particularly important for wearable thermal management, as improved in-plane heat transport enables activation of a larger fraction of the embedded PCM during localized heating, thereby mitigating the "local melting" limitation commonly encountered in low-conductivity PCM systems. Moreover, because h-BN is electrically insulating, the observed improvement in thermal conductivity is achieved without introducing electrical hazards, ensuring that the EPCM composites remain suitable for safe integration adjacent to electronic circuitry and skin-contact layers in wearable devices [24]. The EPCM architecture mitigates peak temperature through a dual mechanism of latent heat absorption and filler-assisted heat spreading. Once the device temperature approaches the PCM melting range (~35–42°C), the composite absorbs excess thermal energy while maintaining a near-constant temperature plateau. Simultaneously, thermally conductive yet electrically insulating fillers promote in-plane heat diffusion, preventing localized overheating and enabling more uniform PCM activation. These combined effects confirm the composite's capability to suppress transient thermal spikes typical of wearable electronic operation.

Mechanical Properties and Stretchability Performance

Uniaxial tensile testing confirms that the EPCM composites retain the characteristic soft, elastomeric response required for wearable conformity. Stress–strain curves (Figure 6) exhibit a nonlinear elastic profile typical of elastomeric materials, with no abrupt stress drops or fracture events associated with µPCM inclusion. At µPCM contents up to ~50 wt%, the EPCM composites sustain strains well above 150% without mechanical failure, satisfying the stretchability requirement for wearable applications involving bending, twisting, and skin deformation [22-24].

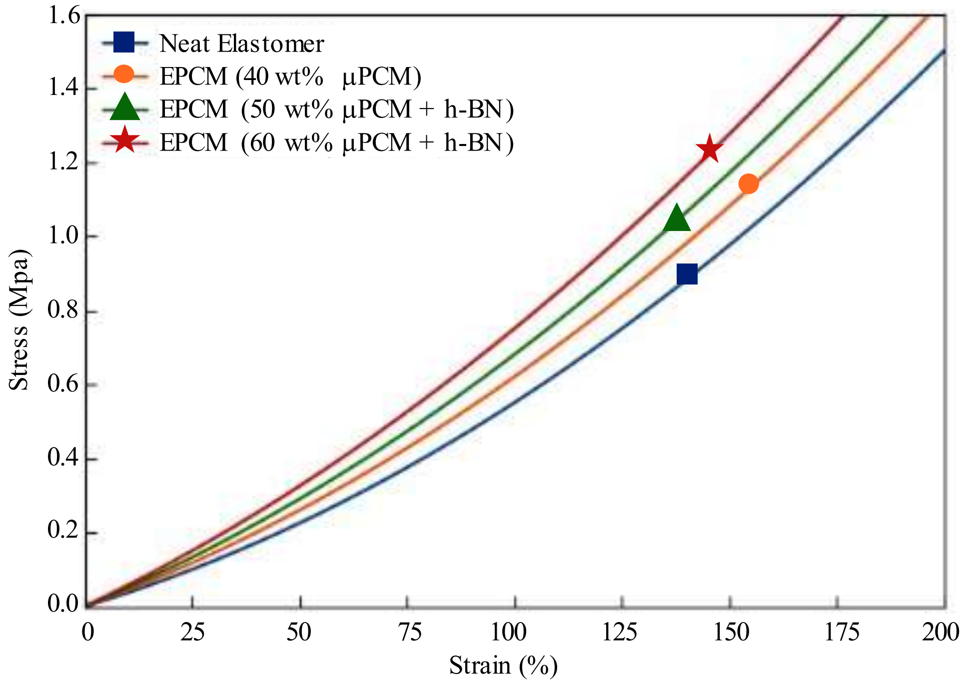

Figure 6. Representative stress–strain curves of neat elastomer and EPCM composites with increasing µPCM and h-BN content.

As µPCM and BN loadings increase, a gradual increase in Young's modulus is observed, reflecting the reinforcing effect of rigid inclusions. However, the modulus remains within a range compatible with skin-contact comfort, particularly for PDMS-based systems. Importantly, the absence of capsule fracture during tensile deformation indicates that stress is effectively transferred through the elastomer matrix rather than concentrated at the µPCM interfaces.

Cyclic tensile testing further demonstrates excellent mechanical durability. After 500 loading–unloading cycles at 50% and 100% strain, the EPCM composites show minimal hysteresis growth and low residual strain, indicating good elastic recovery and limited viscoelastic energy dissipation [25]. This behaviour is essential for long-term wearable use, where repeated mechanical deformation must not degrade thermal or structural performance. The retention of mechanical integrity after cycling also implies sustained encapsulation stability and continued leakage resistance during extended service life.

Leakage Resistance and Phase Change Stability

Leakage resistance tests provide direct validation of the encapsulation-based design strategy employed in the EPCM composites. During isothermal holding above the PCM melting temperature (50°C for up to 24 h), all EPCM samples exhibit negligible mass loss across the investigated compositions, indicating effective confinement of the molten PCM phase. As shown in Figure 7a, the EPCM samples retain their original shape and surface integrity prior to testing, with no visible exudation or surface wetting.

The blotting paper test further confirms the absence of PCM leakage. After prolonged thermal exposure, the blotting paper placed in contact with the EPCM samples remains clean and free from any oil-like stains or discoloration, as clearly observed in Figure 7b. The absence of visible wetting or spreading marks provides strong visual evidence that the molten PCM remains fully confined within the microcapsules and the surrounding elastomer matrix, even under sustained temperatures above the phase transition point.

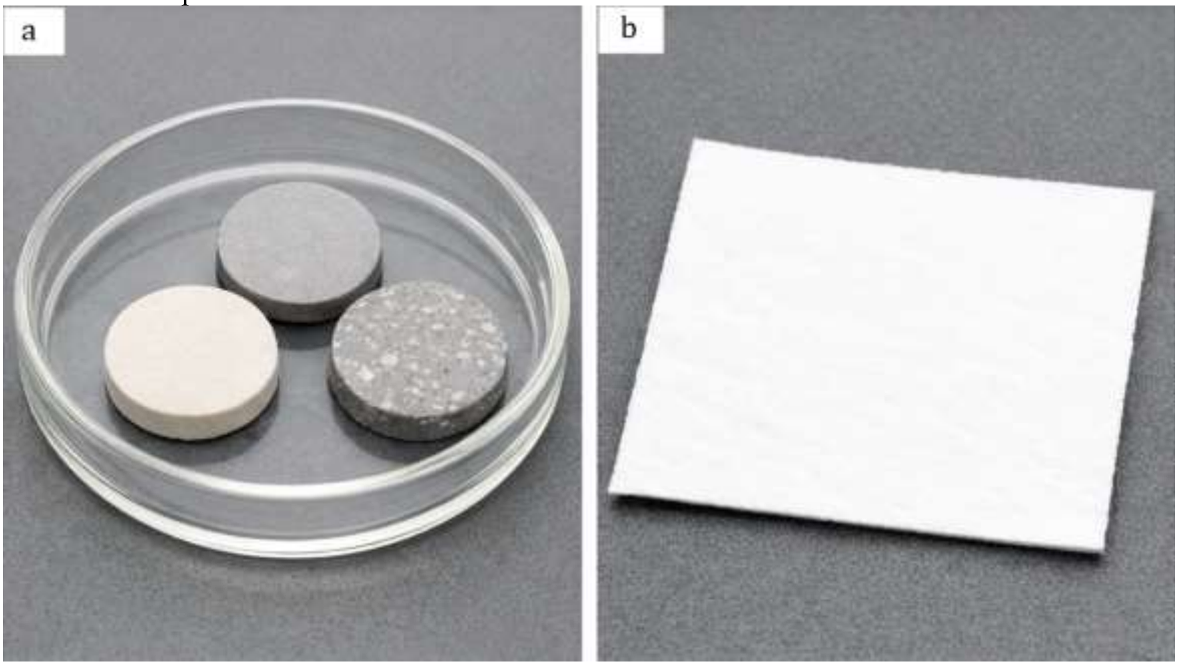

Figure 7. Photographs showing (a) EPCM samples before thermal leakage testing and (b) clean blotting paper after 24 h at 50°C, indicating leakage-free phase change behaviour.

Notably, this leakage-free behaviour is maintained even at high μPCM loadings, highlighting the combined effectiveness of the core–shell microencapsulation and elastomer confinement strategy. This result is particularly significant for wearable electronic applications, where PCM leakage could compromise electrical insulation, interfacial adhesion, and user comfort[26, 27]. The photographic evidence in Figure 7, together with the negligible mass loss measurements, demonstrates that the EPCM system possesses excellent phase change stability and robustness against prolonged thermal exposure, accidental overheating, and environmental temperature fluctuations, thereby reinforcing its suitability for long-term wearable thermal management applications.

The combined effects of latent heat storage, improved in-plane heat transport, and mechanical conformity demonstrate that EPCM composites function as effective passive thermal buffers. This device-level performance directly addresses the two key engineering gaps identified in the introduction—localized PCM activation and the need for electrically insulating, stretchable thermal management materials—thereby validating the proposed EPCM architecture for next-generation wearable electronics.

CONCLUSION

This study establishes a comprehensive materials design framework for elastomer-embedded PCM composites that simultaneously addresses leakage resistance, mechanical stretchability, electrical safety, and effective thermal buffering—key requirements for next-generation wearable electronics.

i. Microencapsulation combined with elastomer confinement completely suppresses PCM leakage during repeated solid–liquid transitions, as confirmed by negligible mass loss and clean blotting paper after prolonged exposure at 50°C, even at high μPCM loadings (up to 70 wt%).
ii. EPCM composites exhibit a well-defined and reproducible melting transition within the skin-safe range of 35–42°C, with minimal supercooling and no significant shift in peak position upon filler addition or increased PCM loading, ensuring predictable thermal regulation.

iii. Incorporation of low loadings of electrically insulating h-BN fillers leads to a stepwise increase in thermal conductivity compared to neat elastomer and μPCM-only systems, enabling efficient lateral heat spreading and mitigating localized PCM activation without introducing electrical hazards.
iv. The EPCM composites retain a soft, nonlinear elastomeric response and sustain strains well above 150% without mechanical failure at μPCM contents up to ~50 wt%, while cyclic tensile testing confirms excellent elastic recovery and low hysteresis over 500 deformation cycles.
v. The compatibility of the EPCM system with simple casting–curing and melt-compounding routes, together with its balanced thermal, mechanical, and safety performance, makes it a practical and scalable passive thermal management solution for wearable electronics, flexible heaters, and skin-interfaced devices.

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