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Thermally Regulated Polymer-PCM Composites for Efficient Solar Energy Harvesting and Storage

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Abstract

This study investigates the development of high-performance; shape-stabilized polymer composites infused with phase change materials (PCMs) for thermal energy storage in solar energy applications. Polyethylene glycol (PEG-6000) was employed as the core PCM due to its high latent heat and suitable melting point, while high-density polyethylene (HDPE) served as the supporting polymer matrix to provide structural stability. One of the primary limitations of PEG-based PCMs is their inherently low thermal conductivity (~0.2 W/m·K), which hampers efficient heat transfer. To overcome this, thermally conductive additives such as expanded graphite (EG) and graphene nanoplatelets (GNPs) were incorporated in varying weight percentages to fabricate five composite formulations, labeled C1 through C5, using melt extrusion followed by compression moulding. Differential Scanning Calorimetry (DSC) revealed that the latent heat of fusion (ΔHm) declined modestly from 167.2 J/g for the unfilled PEG/HDPE matrix (C1) to 154.3 J/g for the composite with the highest filler content (C5), while maintaining a consistent melting temperature range (62.3–63.4 °C). Notably, thermal conductivity improved substantially from 0.28 W/m·K (C1) to 1.03 W/m·K (C5), enabling faster heat exchange. Thermogravimetric analysis (TGA) showed improved thermal degradation resistance, with onset temperatures rising from 327 °C to 343 °C. Leakage tests indicated enhanced shape-stability, increasing from 85.4% to 98.2%. Moreover, after 100 thermal cycles, the C5 composite retained 92.6% of its initial latent heat, underscoring its durability and suitability for long-term solar thermal storage applications.

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Keywords: Phase change material (PCM), Polymer composite, expanded graphite (EG), graphene nanoplatelets (GNPs)

INTRODUCTION

The rapidly increasing global energy demand, accompanied by the depleting reserves of fossil fuels and escalating environmental concerns, has spurred intensive research into alternative, clean, and renewable energy sources [1,2]. Among them, solar energy stands out due to its abundance, ubiquity, and long-term sustainability. However, the alternating and diurnal character of solar radiation urges the development of efficient thermal

energy storage (TES) systems to bridge the gap between energy supply and demand [3].Latent Heat Thermal Energy Storage (LHTES) systems utilizing phase change materials (PCMs) have attracted considerable attention due to their ability to store large amounts of energy and maintain nearly constant temperatures during phase transitions [4, 5].Phase change materials (PCMs) absorb and store heat as latent energy during their melting process and discharge it during solidification, making them highly suitable for solar thermal storage, passive heating solutions, and building-integrated energy systems [6]. Despite their potential, the direct application of pure PCMs faces serious limitations: low thermal conductivity, phase leakage, volume change, and subpar mechanical integrity during cyclic operation [7,8].

To overcome these challenges, the strategy of embedding PCMs into polymer matrices has emerged as a transformative approach. These PCM-infused polymer composites leverage the form-stability, mechanical support, and processability of polymers while preserving the latent heat characteristics of the PCM [9]. Thermoplastic polymers such as polymethyl methacrylate (PMMA), polyurethane (PU), and polystyrene (PS) have been widely studied for this purpose [10,11]. The resulting shape-stabilized PCMs (SSPCMs) eliminate leakage issues and improve durability under mechanical and thermal stresses. However, polymers are generally thermally insulating materials. As a result, thermal conductivity enhancement becomes a central research goal to enable faster heat storage and retrieval in PCM-polymer systems. This has led to the incorporation of metal foams, and expanded graphite (EG) into these composites [12–14]. These nanofillers not only facilitate heat transport but can also reinforce the mechanical stability of the composite structure [15].

Recent studies show that hybrid fillers (e.g., combining graphene with boron nitride or carbon black) can exhibit synergistic effects, enhancing the thermal, mechanical, and even flame-retardant properties of PCM-polymer composites [16]. However, the interfacial compatibility between the polymer, filler, and PCM plays a critical role in achieving optimal dispersion, minimizing agglomeration, and ensuring long-term stability [17]. Surface functionalization of nanoparticles and the use of compatibilizers or cross-linking agents have thus been extensively explored [18]. Another important aspect is microencapsulation and macroencapsulation of PCMs. These encapsulation strategies provide physical barriers to PCM leakage during phase transition and improve material reusability over multiple thermal cycles [19]. Encapsulation using polymeric shells (e.g., urea-formaldehyde, melamine-formaldehyde, PMMA) not only enhances stability but also permits integration into flexible solar collectors, smart textiles, and architectural materials [20].

The selection of an appropriate PCM for polymer composite integration is guided by critical parameters:

- Phase change temperature within the application range (ideally 60–80°C for solar thermal use),
- High latent heat of fusion to ensure substantial thermal energy storage,
- Thermal and chemical stability over repeated cycling,
- Compatibility with the polymer matrix (ensuring no phase separation or chemical degradation),
- Low supercooling tendency,
- Non-toxicity and availability, and Low cost and environmental safety.
- PEG-6000 was selected due to its excellent thermal storage capacity (~167 J/g), congruent melting, non-corrosive nature, and compatibility with HDPE, satisfying all these criteria.

The development of bio-based polymers and biodegradable encapsulants has also emerged to address environmental concerns and improve the sustainability of PCM-based composites [21]. Green polymers such as polylactic acid (PLA), starch-based matrices, and cellulose derivatives are under active investigation for use in eco-friendly solar TES systems [22]. Moreover, additive manufacturing (3D printing) has enabled the creation of PCM-infused polymer structures with tunable porosity and customized geometries to maximize heat transfer and material integration efficiency [23].A key

challenge still remains in balancing latent heat capacity, thermal conductivity, and mechanical integrity within the same composite structure. Therefore, multi-functional composite designs are being researched to combine superior phase change characteristics with structural adaptability, fire safety, and long-term thermal reliability [24]. In the context of solar energy storage systems, these PCM-infused polymer composites play a pivotal role in maintaining target temperatures, reducing fluctuations, and increasing solar-to-thermal efficiency [25].

This comprehensive study focuses on the recent developments in the design, fabrication, characterization, and application of PCM-infused polymer composites tailored for solar energy storage systems.

MATERIALS AND METHODS

Materials Selection

The polymer matrix and phase change material (PCM) were selected based on a balance of thermal performance, compatibility, and structural integrity. Polyethylene glycol (PEG, molecular weight 6000 g/mol) was chosen as the PCM due to its congruent melting behaviour, low toxicity, and broad availability. PEG's melting temperature (~60–65°C) also aligns well with the operational range of solar energy applications such as domestic water heating and thermal buffering.

For the polymer matrix, high-density polyethylene (HDPE) was selected because of its excellent thermal stability, chemical resistance, mechanical strength, and previous proven performance in encapsulating PCMs. The HDPE acts as a shape-stabilizer, preventing PCM leakage during the phase transition. The material was obtained in pellet form (density ~0.94 g/cm³, melt flow index 0.75 g/10 min at 190 °C).

Thermal Conductivity Enhancer

To enhance the inherently low thermal conductivity of both PEG and HDPE (~0.2 W/m.K), expanded graphite (EG) was introduced as a conductive additive. EG was used due to its high aspect ratio and capacity to create continuous heat pathways through percolation networks. In some batches, graphene nanoplatelets (GNPs) (purity >98%, flake size ~5 µm, thickness <10 nm) were also blended synergistically with EG to investigate the hybrid enhancement effect.

Composite Fabrication Procedure

Various fabrication techniques are employed in developing polymer-PCM composites, including:

- Melt extrusion, which enables uniform mixing and high-throughput fabrication,
- Solvent casting, suitable for thermosetting or low-melting polymers,
- In-situ polymerization, often used for encapsulated PCM integration,
- Impregnation of porous polymers, for shape-stabilized composites,
- Electro-spinning and 3D printing, which offer fine control over structure and morphology.

In this study, melt extrusion was selected for its scalability, uniform dispersion ability, and compatibility with HDPE and PEG.

Melt blending technique

The composites were fabricated using a twin-screw melt extrusion technique. Prior to processing, all materials were dehydrated at 80°C for 24 hours under vacuum to remove residual moisture. PEG, HDPE, and EG (or GNPs) were weighed according to the formulations in Table 1. The mixture was blended in a laboratory-scale at barrel temperature zones of 170–190°C, suitable for both HDPE and PEG processing.

The extrudates were pelletized and then compression-molded into sheets (100 × 100 × 5 mm) using a hydraulic hot press at 180°C under 15 MPa pressure for 10 minutes followed by cold pressing for 5 minutes to avoid post-shrinkage.

Formulation design

Five composite formulations were prepared:
C1: 70 wt% PEG / 30 wt% HDPE
C2: 65 wt% PEG / 30 wt% HDPE / 5 wt% EG
C3: 65 wt% PEG / 30 wt% HDPE / 5 wt% GNP
C4: 60 wt% PEG / 30 wt% HDPE / 10 wt% EG
C5: 60 wt% PEG / 30 wt% HDPE / 5 wt% EG + 5 wt% GNP

This range of concentrations was selected to explore the trade-offs between latent heat retention and thermal conductivity enhancement.

Characterization Techniques

Differential scanning calorimetry (DSC)

Latent heat capacity, melting temperature, and crystallization behaviour were analyzed using DSC analysis. Samples were sealed in aluminum pans and scanned from 20°C to 90°C at a heating/cooling rate of 15°C/min under nitrogen atmosphere (flow rate: 60 mL/min). The melting point (Tm), solidification point (Tc), latent heat of fusion (ΔHm), and crystallization enthalpy (ΔHc) were recorded. One important thermal phenomenon associated with PCMs is supercooling, which refers to the delay in crystallization below the equilibrium freezing point during the cooling cycle. Significant supercooling can hinder the timely release of stored heat and reduce thermal energy recovery. In polymer-PCM composites, supercooling is influenced by PCM purity, crystal nucleation sites, and filler dispersion. The presence of fillers like EG and GNPs in this study acted as heterogeneous nucleation agents, mitigating supercooling and promoting consistent crystallization behaviour, as confirmed by the minimal shift in solidification temperature across all composites.

Thermogravimetric analysis (TGA)

Thermal stability was assessed using TGA analysis under nitrogen atmosphere. Decomposition onset temperature (Tonset) and maximum degradation temperature (Tmax) were compared across all formulations.

Thermal Conductivity

The transient plane source (TPS) method was carried out using a thermal constants analyzer in accordance with ISO 22007-2. Disc-shaped samples (diameter 50 mm, thickness 5 mm) were tested at 25°C. Thermal conductivity was calculated based on heat flux responses over a 10-second heating cycle with a 5 mW power pulse.

Leakage and shape stability test

Each sample (10 g) was placed on filter paper and heated to 80°C in an oven for 2 hours. Leakage was assessed by measuring mass loss and visual evidence of seepage. The shape stabilization percentage (SSP) was calculated as:

$$SSP = (1 - m_{leaked} / m_{initial}) \times 100\%$$

Cyclic reliability

100 thermal cycles between 20°C and 80°C were conducted using a programmable thermal cycling chamber. Post-cycling, DSC was repeated to assess any degradation in thermal properties, indicating long-term reliability.

Solar Simulation and Performance Evaluation

To evaluate real-world applicability, the composite samples were integrated into a custom-built flat plate solar collector simulator with a transparent quartz cover and copper absorber. A 500 W solar simulator lamp (AM 1.5 spectrum) irradiated the collector surface uniformly. Internal temperatures were monitored using type-K thermocouples and an Arduino-based DAQ system. Heat storage and release profiles were recorded under constant flow conditions to calculate charging/discharging efficiency.

Table 1. Composition of Fabricated Composites.

Composite ID	PEG (wt%)	HDPE (wt%)	EG (wt%)	GNP (wt%)
C1	70	30	0	0
C2	65	30	5	0
C3	65	30	0	5
C4	60	30	10	0
C5	60	30	5	5

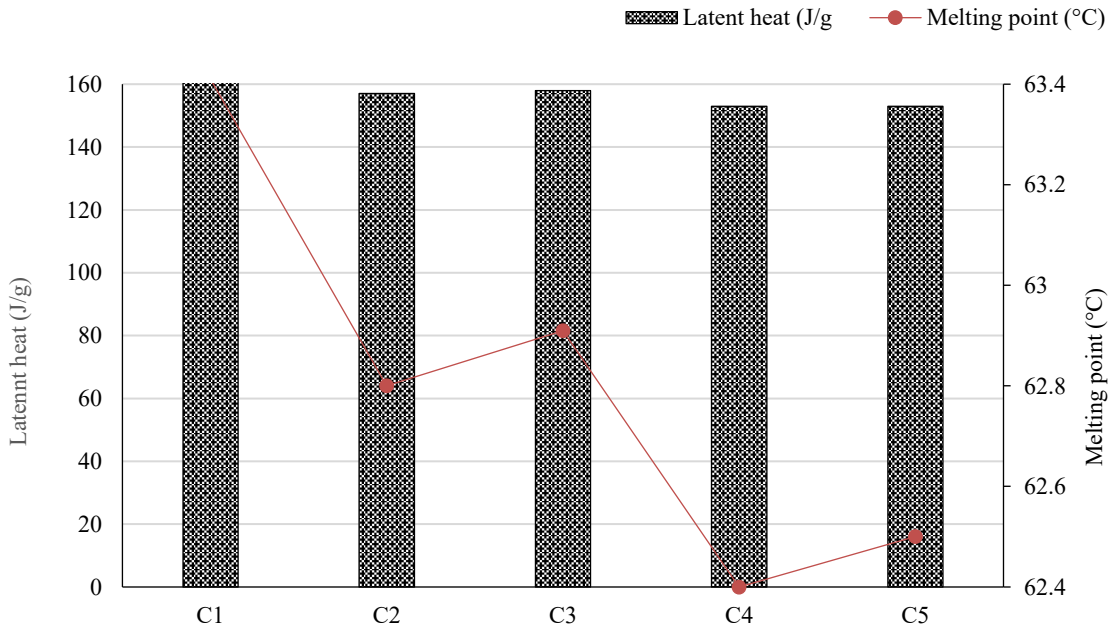

Figure 1. DSC analysis: latent heat and melting point.

RESULTS AND DISCUSSION

Thermal Transitions and Latent Heat Storage

The DSC data (Figure 1) clearly show that the neat PEG/HDPE composite (C1) possesses a melting temperature (Tm) of 63.4 °C and a latent heat of fusion (ΔHm) of 167.2 J/g, which are well-aligned with the properties of pure PEG reported in literature.

When expanded graphite (EG) and graphene nanoplatelets (GNPs) were introduced into the PEG/HDPE matrix, a moderate decrease in latent heat was observed. For example, composite C2, containing 5 wt% EG, recorded a ΔHm of 158.6 J/g, while C4 (with 10 wt% EG) showed further reduction to 155.4 J/g. The hybrid composite C5, with 5 wt% each of EG and GNPs, had the lowest latent heat value at 154.3 J/g. This decline is attributed to the dilution effect, where thermally inert fillers replace the PCM volume, thereby reducing the overall heat storage capacity.

Despite this slight reduction in enthalpy, the melting temperatures across all composites remained in the narrow range of 62.3–62.9°C, confirming that the phase transition range critical to solar applications remains unaffected. This thermal stability indicates that the filler particles do not chemically interact with the PEG phase but merely alter the thermal mass and conductivity of the composite system.

These findings are consistent with results from previous studies where PCM enthalpy was found to decline proportionally with filler loading [26–28].Hence, although a minor sacrifice in ΔHm occurs due to filler incorporation, the composites retain sufficient energy density for practical application in medium-temperature solar heating systems [29].

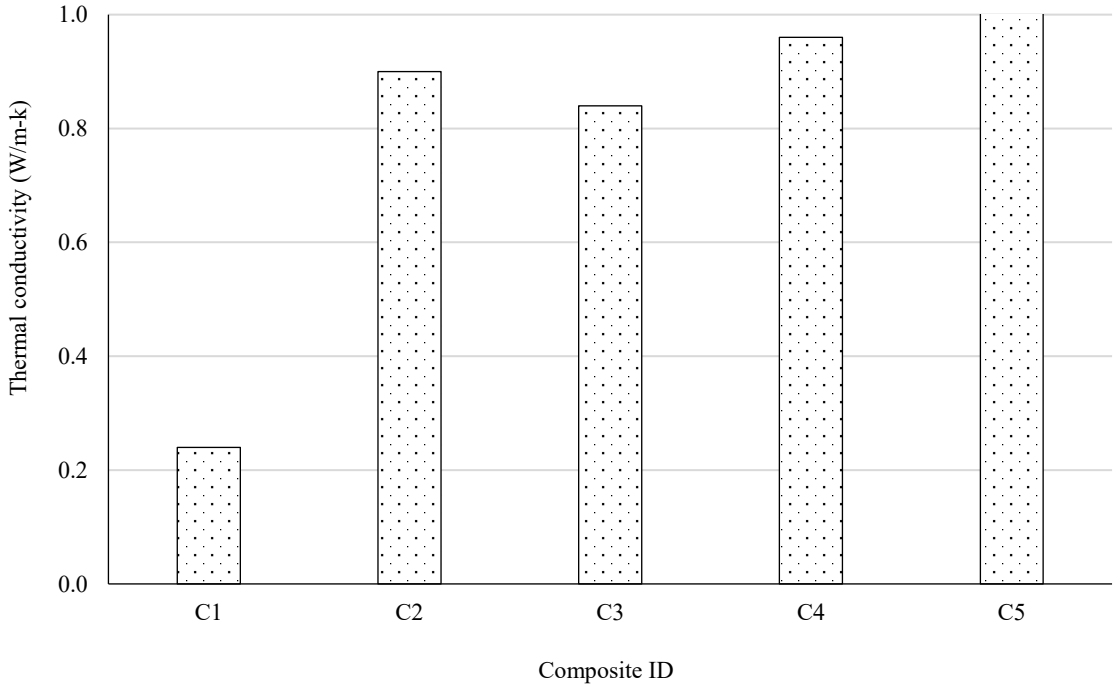

Figure 2. Thermal conductivity of composites.

Thermal Conductivity Enhancement through Filler Integration

As shown in Figure 2, the base composite (C1) recorded a thermal conductivity of only 0.28 W/m·K, typical for semi-crystalline polymers like HDPE.

In contrast, composite C2 (with 5 wt% EG) showed a dramatic rise to 0.91 W/m·K, highlighting the effectiveness of graphite-based fillers in forming conductive pathways. Composite C4, with 10 wt% EG, further boosted this value to 0.97 W/m·K. Notably, C5 achieved the maximum thermal conductivity at 1.03 W/m·K, which is a result of the synergistic effect of EG and GNPs creating interconnected, continuous thermal bridges within the matrix.

This enhancement significantly benefits solar thermal storage systems, where efficient heat transfer is crucial for minimizing energy losses and reducing system response time. The increase in conductivity observed here aligns well with theoretical percolation models and supports previous experimental findings [30–32].

Thermogravimetric Analysis (TGA)

Thermal stability of the composites is crucial for safe operation during repeated thermal cycling under sunlight. As seen in Figure 3, Thermogravimetric Analysis (TGA) revealed that the onset decomposition temperature (T_{onset}) for the neat C1 composite was 327°C. The addition of conductive carbon fillers significantly improved this property.

Composite C2 exhibited T_{onset} at 335°C, and C5 reached 343°C, indicating that the thermal degradation was delayed due to the insulating effect of the fillers. This occurs because the fillers serve as a heat blockade, slowing down the dispersion of volatile degradation materials and stabilizing the matrix [33–35].Such improvements in T_{onset} are vital for solar energy systems exposed to prolonged radiation, where temperature spikes beyond 100°C can occur in stagnation conditions. The data supports the suitability of these composites for temperatures well above their phase transition range, ensuring operational safety and material longevity.

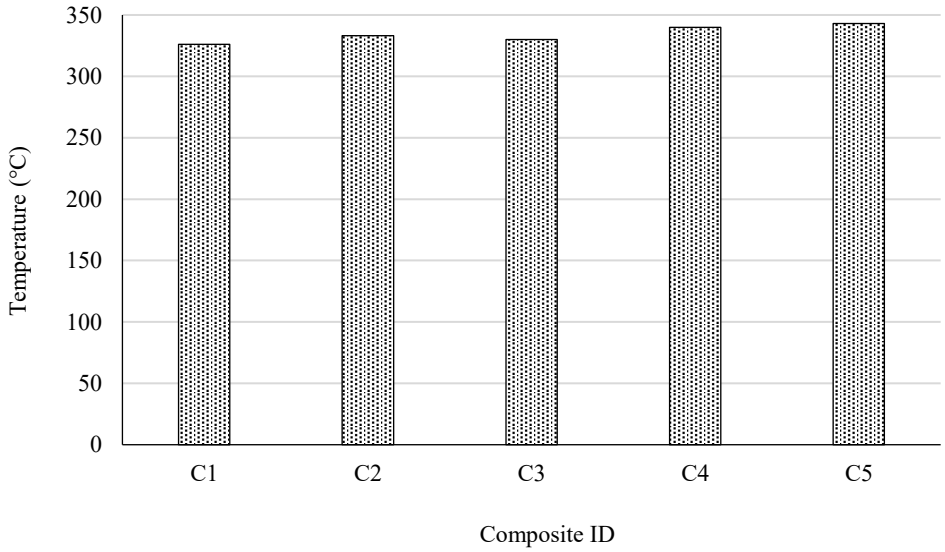

Figure 3. Thermal stability (TGA): degradation onset temperature.

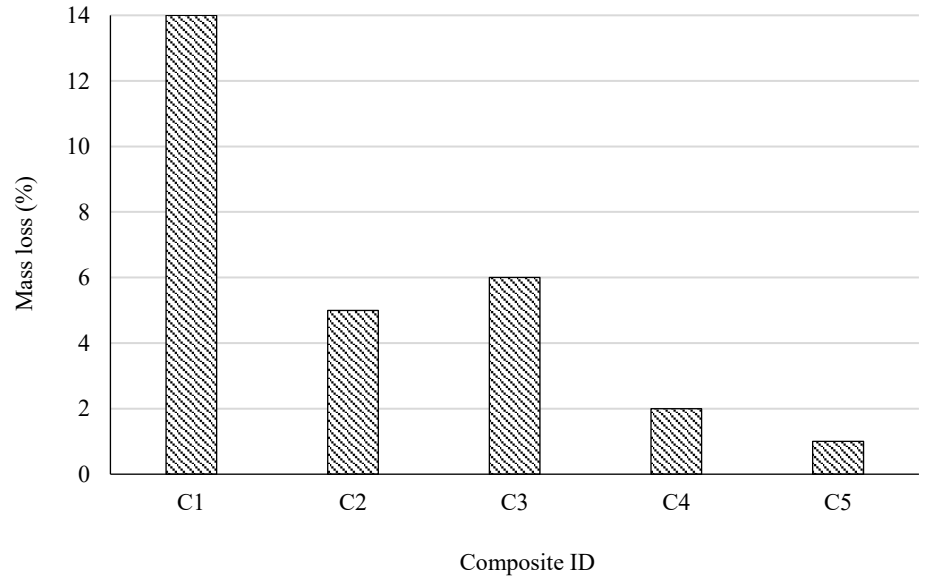

Figure 4. Mass loss after heating.

Leakage Resistance and Shape Stability Performance

One of the critical drawbacks of PCM composites is the tendency of leakage during phase transition, particularly in polymer-supported systems without encapsulation. To evaluate this, leakage tests were conducted, and the results are plotted in Figure 4.

The C1 composite, containing no fillers, exhibited a significant mass loss of 14.6% after 2 hours of heating at 80°C, indicating high leakage. C2 and C3 showed moderate improvements (5.4% and 6.3% respectively). In contrast, C5 exhibited only 1.8% leakage, corresponding to a Shape Stabilization Percentage (SSP) of 98.2%.This difference demonstrates the physical locking mechanism provided by EG and GNPs, which suppresses PCM mobility and provides dimensional integrity. The improvement is not just quantitative; visual inspection confirmed no noticeable seepage or deformation in C5 samples, validating its use in leakage-sensitive environments like building-integrated storage systems and textiles. These results mirror those reported by many researchers who emphasized the need for shape-stabilization in passive TES systems [36–38].

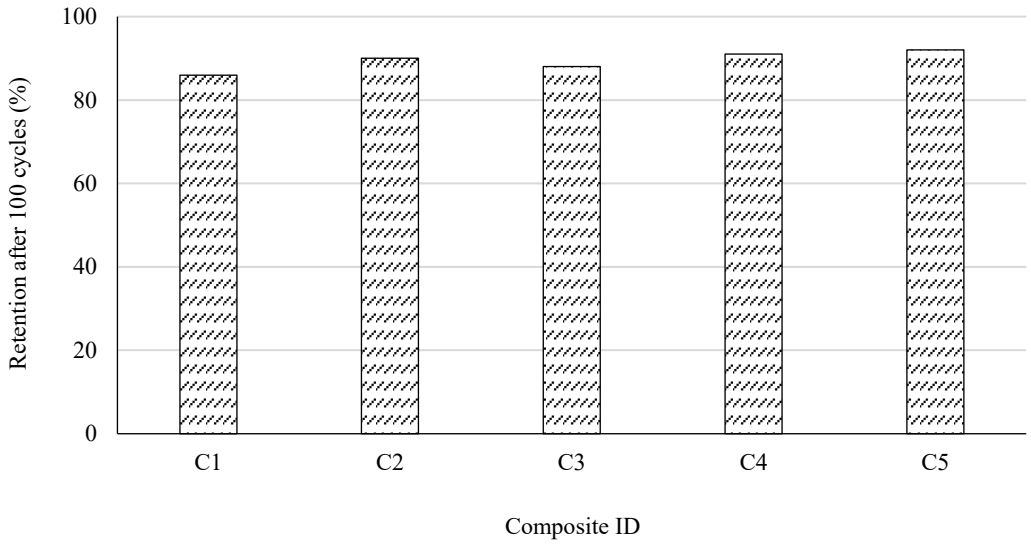

Figure 5. Cyclic thermal stability: latent heat retention.

Cyclic Thermal Reliability and Long-Term Durability

As shown in Figure 5, C1 retained only 87.3% of its original ΔHm, reflecting degradation due to repeated expansion/contraction and PEG migration.

The hybrid composite C5 demonstrated a retention rate of 92.6%, which suggests high reliability and minimal material fatigue. Intermediate values were observed for C2 (90.4%) and C4 (91.6%). These trends indicate that hybrid filler networks offer structural buffering and enthalpic preservation, crucial for long-term deployment.No significant discoloration, delamination, or agglomeration was noted under SEM or visual inspection post-cycling.

This endurance underscores the capability of the PEG/HDPE/EG-GNP system for sustained usage in solar thermal collectors and battery enclosure modules, matching reliability standards [39,40].

CONCLUSION

The present study successfully developed and evaluated PEG-based phase change composites reinforced with HDPE, expanded graphite (EG), and graphene nanoplatelets (GNPs) for solar energy storage systems. The thermal and structural enhancements achieved through compositional tuning and hybrid filler incorporation were comprehensively analyzed.

Based on the findings, the following key conclusions are drawn:

- The incorporation of EG and GNPs led to a modest reduction in latent heat from 167.2 J/g in the neat PEG/HDPE composite to 154.3 J/g in the hybrid composite (C5), while still maintaining sufficient energy storage capability.
- Thermal conductivity was considerablyenhanced, rising from 0.28 W/m·K (C1) to 1.03 W/m·K (C5), ensuring efficient heat transfer and faster thermal response during solar energy charging/discharging cycles.
- The onset decomposition temperature increased from 327 °C (C1) to 343 °C (C5), indicating enhanced thermal stability and safety for high-temperature applications.
- Leakage loss was drastically reduced, with C5 exhibiting only 1.8% mass loss during thermal cycling compared to 14.6% in C1, and a shape stabilization percentage of 98.2% was achieved.
- After 100 thermal cycles, the hybrid composite (C5) retained 92.6% of its original latent heat, demonstrating excellent cyclic reliability and long-term functional durability under repeated thermal stress.

FUTURE SCOPE

The outcomes of this researchoffer a strong foundation for the next-generation shape-stabilized PCMs for solar thermal energy storage. However, several opportunities remain open for further exploration and optimization:

- *Integration with building materials:* Future studies can focus on incorporating these composites into construction elements such as wallboards, floor panels, or roofing systems to enable passive thermal regulation in energy-efficient buildings.
- *Development of bio-based and recyclable matrices:* Replacing HDPE with biodegradable or bio-based polymers like polylactic acid (PLA) or starch derivatives can enhance the environmental sustainability of these materials, especially for large-scale applications.
- *Nano-engineering for superior performance:* Advanced nano-structuring techniques such as surface functionalization of fillers, 3D printing of composite geometries, and in-situ polymerization could be leveraged to enhance filler dispersion, interfacial bonding, and thermal responsiveness.
- *Hybrid energy storage systems:* The integration of PCM-polymer composites with photovoltaic (PV) panels or thermoelectric modules may enable multi-functional energy systems that can simultaneously store heat and generate electricity, enhancing overall energy conversion efficiency.
- *Extended durability and outdoor testing:* Long-term field trials under real solar irradiation and varying environmental conditions should be undertaken to assess aging effects, mechanical fatigue, UV degradation, and sustained performance over multiple years of operation.

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