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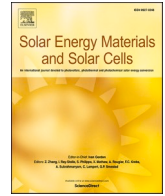
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Coffee and turmeric bio-based shape-stabilized composite PCMs for thermal and solar energy storage applications

Amit Kumar Mishra^a, Matteo Morciano^a, Elena Campagnoli^a, Valter Giaretto^a,
Matteo Fasano^a, Eliodoro Chiavazzo^{a,b,*}

^a Department of Energy, Politecnico di Torino, Corso Duca Degli Abruzzi 24, 10129, Torino, Italy

^b Istituto Nazionale di Ricerca Metrologica, Strada Delle Cacce 91, 10135, Torino, Italy

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ABSTRACT

Bio-based shape-stabilized composite phase change materials (ss-PCMs) are emerging as sustainable solutions for thermal and solar energy harvesting. However, typical shape-stability issues, poor thermal/optical properties, and complex synthesis methods hinder the use of such materials at large scale. This study reports the design of nanofiller-loaded bio-based composite PCMs with enhanced thermo-optical and mechanical properties (shape-stability). Incorporating a 25 wt% biomass-derived porous matrix (coffee/turmeric powder) significantly enhanced the material's structural integrity by effectively controlling PCM leakage while achieving a high latent thermal energy storage (TES) capacity of ~ 130 J/g. Graphene-loaded ss-composite PCMs demonstrated a significant photothermal conversion efficiency enhancement (106 %) and effective thermal management (TM) potential (superheat degree reduced by ~ 10 °C) compared to pristine PCM, due to the improved photo-thermal properties and power density. Thermal cycling (up to 500 cycles) and load-bearing capacity tests ($\sim 212,566$ and $\sim 31,242$ N/m² across the phase transition zone) confirm the high reliability of the proposed novel ss-composite PCMs in terms of thermal and shape stability. These results highlight their strong potential for long-term TES applications, with stable performance even under adverse environmental conditions such as humidity and wetting. This research contributes to the design of bio-compatible (possibly edible) substance-based strategies for creating cost-effective composite PCMs with enhanced thermo-optical and shape stability characteristics, offering significant advancement for latent TES systems and TM technologies.

Nomenclature/Description/Unit

Symbol/abbreviation	Description	Units
PCMs	Phase change materials	–
LA	Lauric acid	–
CBNP	Carbon black nanoparticles	–
GNP	Graphene nanoplatelets	–
SS	Shape stabilized	–
OCT	Optical coherence tomography	–
DSC	Differential scanning calorimetry	–
CSCPCM	Coffee-based ss-CBNP-loaded composite PCM	–
CSGPCM	Coffee-based ss-GNP-loaded composite PCM	–
TSCPCM	Turmeric-based ss-CBNP-loaded composite PCM	–
TSGPCM	Turmeric-based ss-GNP-loaded composite PCM	–
ΔH_s	Latent heat of the sample	J/g
c_p	specific heat capacity	J/(g K)

(continued on next column)

(continued)

k_{PCM}	Thermal conductivity of pristine PCM	W/(m-K)
k_c	Thermal conductivity of nanocomposite PCM	W/(m-K)
k_{sc}	Thermal conductivity of ss-composite PCM	W/(m-K)
TC	Thermal conductivity	W/(m-K)
T_s	Interface temperature	°C
m_s	Mass of the sample	g
t	Time	sec.
A	Area	cm ²
I	Sunlight irradiance	mW/cm ²
TES	Thermal energy storage	–
TM	Thermal management	–
q''	Heat load	W/cm ²
PTC	Photo-thermal conversion	–
η	PTC efficiency	%

* Corresponding author. Department of Energy, Politecnico di Torino, Corso Duca degli Abruzzi 24, 10129, Torino, Italy.

E-mail address: eliodoro.chiavazzo@polito.it (E. Chiavazzo).

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1. Introduction

The rapid growth of human industrialization leads to a massive use of fossil fuels, releasing carbon dioxide, strengthening the greenhouse effect, and causing global climate issues [1,2]. As the global climate crisis grows more severe, scientists are increasingly focusing on sustainable and environmentally friendly energy materials [3]. However, the current advanced green energy technologies, such as solar cells/fuel cells technology, use toxic materials in their device's manufacturing and recycling processes can have long-term effects on the ecosystem [4,5]. Therefore, rising energy consumption and environmental concerns emphasize the development of sustainable energy storage and conversion technologies. Utilizing clean energy from the sun and waste heat has substantial potential to address environmental issues and energy shortages. Among the various approaches, thermal energy storage (TES) technologies stand out as an effective solution for balancing energy supply and demand while lowering carbon emissions [6–9].

In this aspect, photothermal conversion (PTC) energy storage strategies have attracted significant attention due to their cost-effectiveness and high efficiency [10–12]. On the other hand, the growing integration of electronic devices requires efficient thermal management (TM)/cooling technologies to prevent heat accumulation and ensure their safe/reliable function for long-term executions [13–16]. The application of thermal interface materials (TIMs) plays an essential role in enhancing heat dissipation efficiency and TM performance [17,18]. For heat storage/management and solar-thermal conversion, latent heat TES (LHTES) technology using phase change materials (PCMs) is an attractive choice due to their high energy density (nearly twice than sensible heat storage systems), low cost, and capability to store/release thermal energy isothermally during phase transition processes [19–22]. The TM of electronic devices is primarily achieved by active and passive cooling methods and currently, researchers are focused on designing smart passive cooling systems by employing shape-stabilized PCMs with integrated heat sink techniques, surpassing conventional PCM-based approaches [23,24].

For LHTES applications, lauric acid (LA) is a widely adopted organic PCM due to its solid-liquid phase change at relatively low temperature ($\sim 45^\circ\text{C}$), non-corrosiveness, high TES capacity, no-phase-separation, no-supercooling, and good chemical/thermal stability [25,26]. These characteristics make LA an excellent candidate for various low-temperature PCM applications, including solar heat storage, buildings, waste heat recycling, electronics cooling, thermoelectric generators, and automotive fields [9,16,27,28]. Although LA shows outstanding performance in TES, the following major issues limit its practical suitability: (i) low heat conduction ability leading to slow charging/discharging rates, (ii) poor sunlight absorption thus low PTC efficiency, and (iii) risk of PCM leakage during melting phase transition [20,23,29–31]. These crucial drawbacks make conventional PCMs sub-optimal. Hence, shape-stabilized composite PCMs (also known as ss-composite PCMs or form-stable PCMs) are being developed to overcome the above obstacles. Generally, the inclusion of nanofillers with high thermal conductivity (TC) and shape-stabilization techniques such as microencapsulation, polymer matrix, and porous support material adsorption are employed to fabricate ss-composite PCMs with efficient thermophysical characteristics, compared to conventional PCMs [19, 32]. Carbon-based nanofillers are particularly preferred due to their excellent optical and thermo-physical features, such as strong light absorption capacity, high heat conductivity, stable chemical activity, and low-density [33–36].

However, the preparation of ss-composite PCMs using the microencapsulation and polymer matrix techniques is time-consuming and costly due to complex synthesis steps like wet-chemical methods, polymerization, polycondensation, vacuum impregnation, participation of harmful/toxic precursors, limited TC, stability issues, etc., which greatly hinder their large-scale industrial production [37–40]. On the other hand, the porous support material adsorption technique has the

advantages of its easy execution process, high efficiency, and low cost compared to the other ones [41,42]. The porous support, with its enhanced surface area and rich pore volume, generates capillary forces and surface tension that enable the retention of melted PCM within the porous skeleton, effectively preventing overflow [37,43].

Currently, the development of bio-based ss-composite PCMs with a porous structure from biomass is important for creating sustainable and efficient thermal/solar energy storage systems [29,44–46]. These support matrices derived from renewable/biomass resources have a prospect of being environment-friendly, minimizing toxicity, reducing carbon footprint, and achieving cost-effectiveness. To target all these characteristics including enhanced thermo-optical and structural properties, in the past many bio-based ss-composite PCMs were explored [17, 40,41,45,47]. To achieve sustainable energy-saving solutions, it is beneficial to develop green ss-composite PCMs by integrating bio-based porous matrix additives that exhibit beneficial characteristics, including light absorption, high porosity, environmental sustainability, biodegradability, cost-effectiveness, chemical inertness, and antibacterial properties [48–52].

As found, the research work on bio-derived nano-enhanced PCM is still limited and faces scalability issues [53,54]. Previous studies have mainly focused on polymer-based or chemically synthesized ss-composite PCMs, which often require complex and time-consuming preparation processes and involve toxic or non-renewable precursors. Moreover, although several biomass-derived porous supports have been explored, the development of bio-based ss-composite PCMs that simultaneously ensure shape stability, enhanced thermo-optical performance, and simple synthesis remains a major challenge [55,56].

In this context, the present study reports the design of coffee- and turmeric-based nano-enhanced ss-composite PCMs incorporating CBNP and GNP fillers. These materials were prepared without the implication of any time-consuming or complex chemical processes, effectively overcoming the limitations of complex fabrication methods. The synergistic effect of the nanofillers and the bio-based porous matrices significantly improved heat transfer, light absorption, mechanical strength, solar-thermal conversion efficiency, and thermal stability compared to conventional PCMs.

This work therefore addresses the need for sustainable, scalable, and high-performance bio-based ss-composite PCMs for latent thermal energy storage and thermal management applications. The proposed materials exhibit high latent heat capacity ($\sim 130\text{ J/g}$), enhanced photothermal conversion efficiency ($\sim 106\%$), and excellent shape stability even under long-term cycling and load-bearing conditions, demonstrating their strong potential for large-scale, environmentally friendly TES and TM systems.

2. Materials and methods

2.1. Materials and bio-based ss-composite PCMs preparation

Lauric acid, used as base PCM, was purchased from Sigma-Aldrich (99 % purity), and the carbon black nanoparticles (CBNP) and graphene nanoplatelets (GNP) were purchased from PowderNano and Ossila Ltd., respectively, for improvement of heat conduction and photothermal conversion characteristics of pristine PCM. Turmeric powder (*Curcuma*) and coffee powder were used to prepare bio-based ss-composite PCMs. The above constituents received from the suppliers were used without any further purification and chemical treatments.

The CBNP and GNP-based nanocomposite PCMs were prepared by dispersing 4.0 wt% of respective nanofillers into the liquid LA, with the sample temperature maintained above the melting point (bath temperature kept at 50°C). The mixture was then subjected to sonication in a water bath at $\sim 53\text{ kHz}$ and full power for approximately 45 min. After incorporating 25 wt % coffee or turmeric powder into the 4 wt % CBNP-based colloidal solution, the resulting suspension was sonicated and stirred for 45 min. The resulting mixture was cooled at room

temperature and coffee-based ss-CBNP-loaded composite PCM (CSCPCM) or turmeric-based ss-CBNP-loaded composite PCM (TSCPCM) obtained. A similar sample preparation process was also used to prepare coffee-based ss-GNP-loaded composite PCM (CSGPCM) and turmeric-based ss-GNP-loaded composite PCM (TSGPCM), respectively. For thermophysical characterization, the investigated composite samples were prepared (diam. ~ 5.8 cm, height ~ 1.1 cm). Fig. 1 (a) shows the TEM image of the CBNP with discernible particle size of ~ 20 nm and Fig. 1(b)–(c) show TEM images of GNP indicating the planar structures and size of GNP flakes of ~ 1.5 μm . The nanofiller-loaded ss-composite PCMs preparation process steps are illustrated in Fig. 2. Tables 1 and 2 present materials' features and uncertainty-related information, respectively.

2.2. Characterization and performance evaluation of samples

The phase transition temperatures and latent heat values of the pristine LA and CBNP or GNP-loaded shape-stabilized composite PCMs were investigated by differential scanning calorimetry (DSC) under a nitrogen atmosphere with a 3 $^{\circ}\text{C}/\text{min}$ heating rate, using DSC 214 Polyma. The phase change characteristics and latent heat of pristine PCM and PCM-based ss-composites were obtained with an accuracy of ± 0.2 %. The investigation of heat conduction characteristics (thermal conductivity/thermal diffusivity) of the PCM-based samples was determined using a Hot Disk Thermal Constants Analyzer (Hot Disk, TPS 500) with a nominal accuracy of ± 5 % and ± 10 %, respectively. The hot disk sensor comprises an electrically conducting pattern in the shape of a double spiral, which has been etched out of a thin metal (Nickel) foil. This sensor acts as a heat source for raising the PCM sample's temperature and concurrently recording the temperature changes with time. During measurements, a plane hot disk sensor is fitted between two pieces of the sample, and a small constant current is applied; the associated working principle/measurement details of the TPS 500 thermal analyzer are available in Ref. [57]. Optical coherence tomography (OCT) was used to study the microscopic features of the designed ss-composite PCMs using Thorlabs Ganymede (Fig. 4); load-bearing tests were carried out applying a calibrated mass of 6.5 kg on the PCM-based samples (dimensions: 30 mm \times 10 mm \times 6 mm, mass 2.8 g) and then assessing their shape-stability after several thermal cycles across the phase transition zone. The investigation of the thermal stability of PCM-based samples was done with thermogravimetric analysis (TGA) by using TG 209 F3 Tarsus (test range: 25 – 350 $^{\circ}\text{C}$, rate: 5 $^{\circ}\text{C}/\text{min}$, N_2).

A solar simulator (infinityPV ISOSun) was used to mimic concentrated solar light (150 mW/cm^2) and assess the PTC performance of ss-composites PCMs: the sample's temperature variation was recorded by k-type thermocouples every second. The thermal management performance of the ss-composite PCMs, compared to pristine PCMs, was evaluated by monitoring the variation in interface temperature (T_s) between the Al-plate (2 mm thick) and the incorporated sample (1 mm thick) under a constant heat flux of 0.28 W/cm^2 . Each measurement was

performed 3 times and the mean value reported along with the standard deviation.

3. Results and discussion

3.1. Phase change characteristics

Fig. 3(a) and (b) show the heat flow curves, during melting and solidification for the pristine PCM and coffee/turmeric-based ss-composite PCMs, respectively. The heat flow curves exhibit a single endothermic peak (energy absorbed) or exothermic peak (energy released), indicating first-order solid-liquid phase transitions. The latent heat values of the respective samples were determined from the areas under the heat flow curves. DSC revealed the peak melting and solidification temperatures of pristine PCM as 45.9 $^{\circ}\text{C}$ and 40.1 $^{\circ}\text{C}$, respectively. The latent heat of pristine PCM during melting/solidification phase transition processes amounted to ~ 172.5 and ~ 170.4 J/g , respectively. The latter values find good agreement with previously reported results in the literature [58,59]. It is evident from Table 3 that the phase transition temperatures of ss-composite PCMs were not altered substantially with the presence of additives (shape-stabilizer agents: coffee/turmeric powder; nanofillers: CBNP/GNP), which illustrates the absence of any effective interaction between the incorporated additives and the PCM base material. DSC analysis (Fig. 3-c) reveals that the latent heat value and specific heat capacity of the designed ss-composite PCMs are lower than pure PCMs, coherently with previous evidence in the literature [60–62]. Despite this, the bio-based ss-composite PCMs achieved a high latent heat value of approximately 130 J/g , with a decrease of about 25 % compared to the latent heat of pure PCM. Nevertheless, they still demonstrate significant potential for TES capacity in industrial applications. In the solid state, the examined c_p value of ~ 2.03 $\text{J}/(\text{g} \cdot \text{K})$ for LA shows good agreement with literature data at room temperature [63].

The lower latent heat and specific heat capacity of ss-composite PCMs with respect to the bare PCM is attributed to the effective reduction of PCM mass content within the composite core volume, since additives like coffee/turmeric powder are inert and do not show phase transition thus not being able to contribute by latent heat storage, and CBNP/GNP have poor c_p compared to the host matrix [64–67]. An overview of the measured phase change characteristics and specific heat capacity of the tested PCM-based samples is reported in Table 3.

Optical coherence tomography (OCT) is a contactless and non-destructive technique for high-resolution microstructure imaging within scattering media based on depth-resolved optical reflectivity [68, 69]. Fig. 4 (a) and (c) show the OCT images of the prepared ss-PCMs (TSPCM and CSPCM), while Fig. 4(b)–(d) show their front reflection images, respectively. These OCT images illustrate the internal microstructure of ss-PCMs, identifying distinct components with brighter and darker streaks, which correspond to the pristine PCM and supporting matrix (turmeric/coffee) uniformly distributed within the composite. These OCT microstructural features helped to analyse the homogenous

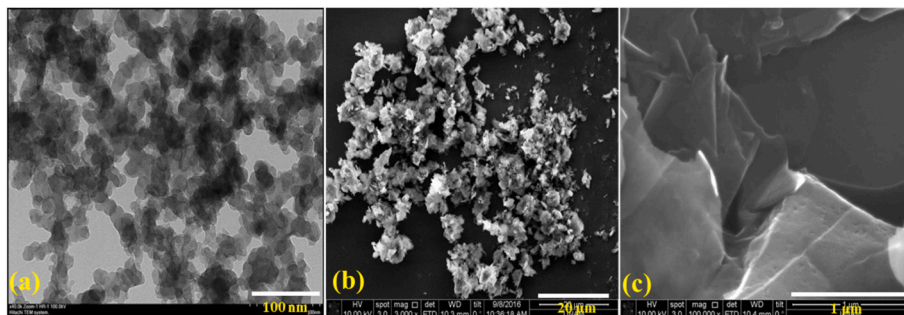


Fig. 1. TEM images of (a) carbon black nanoparticles (CBNP), (b)–(c) graphene nanoplatelets (GNP), which are provided by PowderNano and Ossila Ltd., respectively.

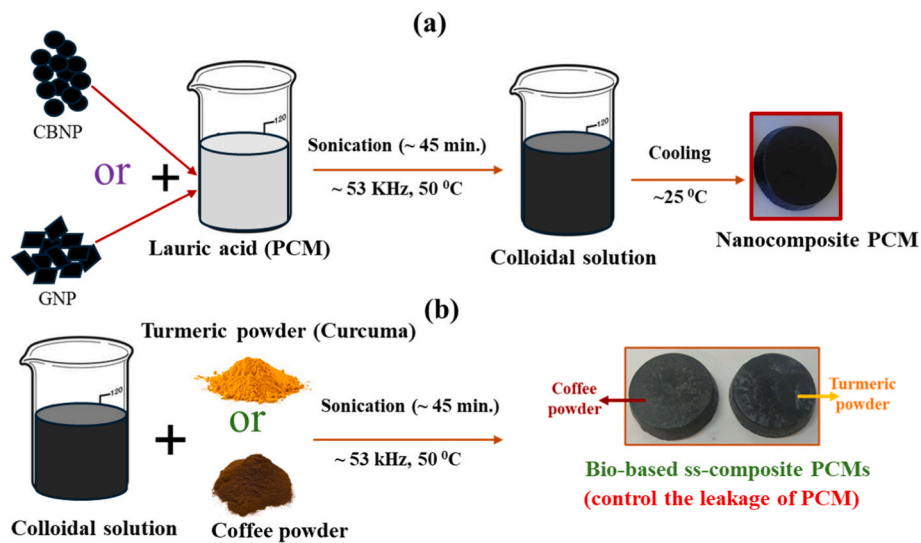


Fig. 2. Illustration of (a) CBNP and GNP-based nanocomposite PCMs; (b) coffee/turmeric-based ss-composite PCMs preparation steps.

Table 1

Description of materials' suppliers and features.

Material	Source	Purity (%)	Dimensions	Specific surface area (m ² /g)	Bulk thermal conductivity (W/m·K)
Lauric acid (LA)	Sigma-Aldrich	~99	–	–	~0.20
Carbon black nanoparticles (CBNP)	Powder-Nano	~98	Size ~20 nm	~195	~0.2–0.4
Graphene nanoplatelets (GNP)	Ossila Ltd.	~99.9	Size ~1.5 μm, Thickness ~3 nm	~800	~3000
Coffee powder	Nescafe Classic	~100	–	–	~0.11–0.13
Turmeric powder	TRS Asia's Finest Foods	~100	–	–	~0.07

Table 2

Uncertainty of instruments/tools.

Instruments/tools	Model	Property/quantity	Uncertainty
Hot Disk Thermal	TPS500	Thermal conductivity	~ ± 5 %
Differential scanning calorimetry (DSC)	DSC 214 Polyma	Phase change temperatures and latent heat values	~ ± 0.2 %
Solar simulator	infinityPV ISOSun	Solar irradiance	~ ± 2 %
Thermocouple	K-type	Temperature measurement	~ ± 1.5 °C
Heat pad	RS PRO silicon heater	Thermal load	~ ± 5 %

Table 3

DSC analysis of ss-composite PCMs.

Sample	Melting Process		Solidification Process		c _p (J/g K) Solid-state, 25 °C
	Melting point (°C) (±0.1 °C)	Latent heat (J/g) (±0.4 J/g)	Freezing point (°C) (±0.1 °C)	Latent heat (J/g) (±0.4 J/g)	
PCM (LA)	45.9	172.5	40.1	170.4	2.03
CSCPCM	46.2	133.7	39.2	130.5	1.94
CSGPCM	46.7	126.0	39.9	122.8	1.86
TSCPCM	46.3	134.1	39.5	127.4	1.85
TSGPCM	46.5	142.0	39.6	133.2	1.72

distribution of the PCM within the supporting matrix and reveal that their present porous activity characteristics contribute to improving the structural integrity of the ss-composite PCMs.

4. Enhanced heat conduction performance

Fig. 5(a) shows the enhancement of TC of lauric acid-based nanocomposite PCMs compared to pristine PCMs. Thermal measurement results show that the TC of pure PCM is effectively enhanced with the inclusion of these carbon-based nanofillers. The TC enhancement (%) was calculated using the expression $\left[\left(\frac{k_{c/sc}}{k_{pcm}} - 1 \right) * 100 \right]$, where k_c , k_{sc} , and k_{pcm} indicate the effective TC of the nanocomposite PCM, ss-composite PCM, and pristine PCM in the solid state, respectively. The TC of solid pristine PCM was enhanced by ~50 % and ~75 % after 4 wt % CBNP and GNP loading of nanoparticles, respectively. This is attributed to the generation of internal stress fields during solidification [70, 71]; the dispersed nanoparticles in the solution are therefore pushed towards the grain boundaries and build efficient heat conduction pathways (percolation networks) [72–74]. The superior TC enhancement of GNP-loaded nanocomposite PCMs could be attributed to its inherent higher bulk TC (~3000 W/(m · K)) and to the 2D structure of nanoparticles that ease plane-to-plane contacts, which comparatively help to create efficient percolation pathways with lower thermal resistance [75,76]. On the other hand, CBNP (expected bulk TC ~ 0.25–4.0 W/(m · K)) is substantially poorer than GNP also work to effectively enhanced the TC of pristine PCM is attributed due to their high compressibility, and volume-filling characteristics, leading to form quasi-2D percolation networks in the solid state [19,77]. Fig. 5 (b) illustrates schematically the nanofiller percolation networks created

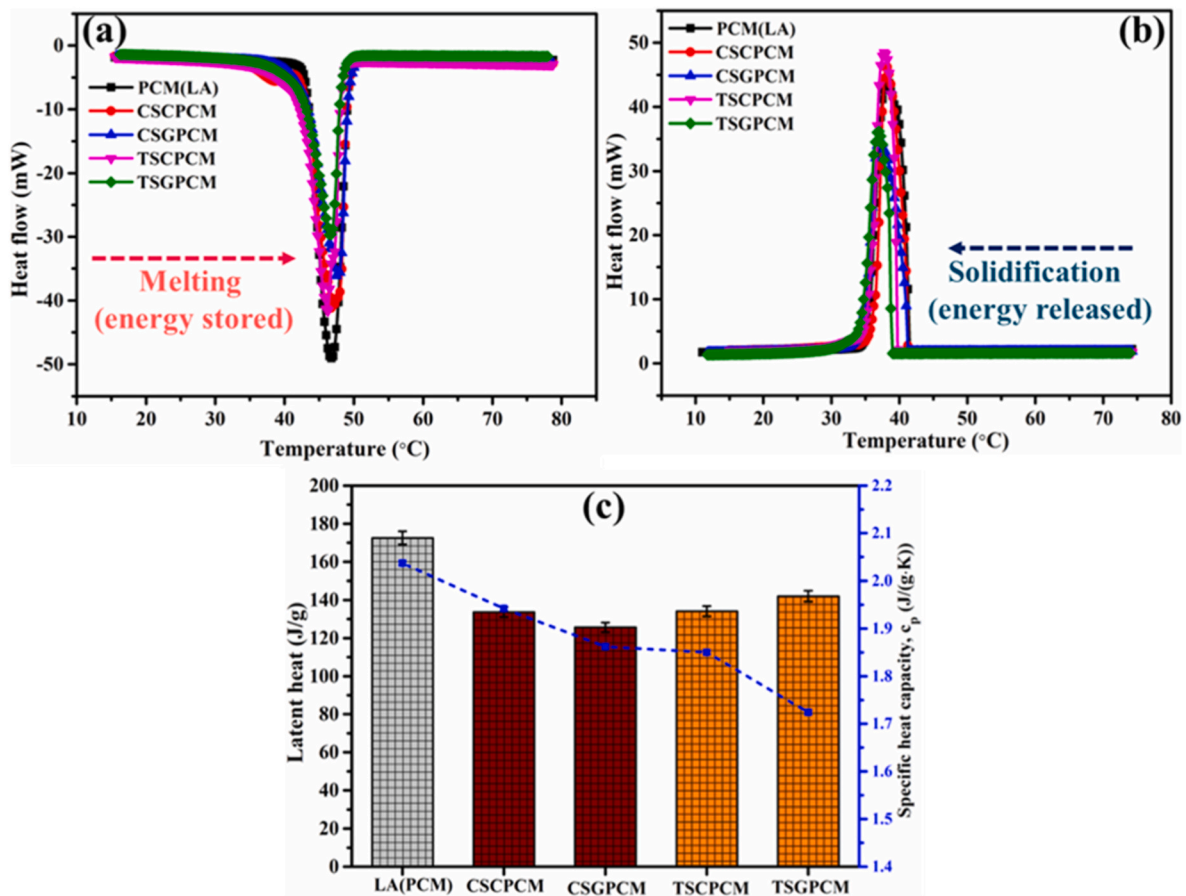


Fig. 3. (a)–(b) shows DSC curves during the melting and solidification processes of various coffee and turmeric-based ss-composite PCMs. (c) Variation of latent heat values during melting, and specific heat capacity of respective ss-composite PCMs at 25 °C. Phase change characteristics of pure PCM are also shown for comparison.

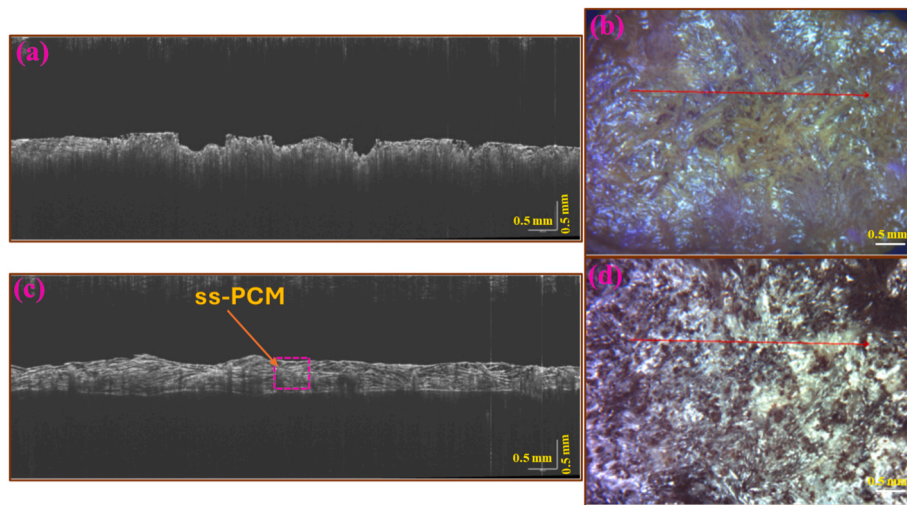


Fig. 4. (a) and (c) show OCT images of turmeric and coffee-based shape-stabilized PCMs. (b) and (d) show their corresponding front reflection images, respectively. The scale bars for lateral/axial dimensions refer to 0.5 mm.

during the solidification of the host matrix, which improves phonon-mediated heat transfer in the solid state.

In Fig. 5 (c), coffee and turmeric-based ss-composite PCMs show enhanced TC with respect to pristine PCM, however, the achieved TC is lower than their respective CBNP and GNP nanocomposite PCMs (without shape-stabilization). This is because the considered supporting matrixes (coffee/turmeric powder) have poor heat conduction

characteristics (lower than bare PCM) [67,78,79]. On the other hand, ss-composite PCMs achieved better heat transfer efficiency than pristine PCMs due to the contribution of the nanofiller's thermophysical properties and well-stabilized network pathways (facilitating the phonon propagation within the composite matrix) [23,80]. The investigated thermal properties of ss-composite materials are summarized in Table 4.

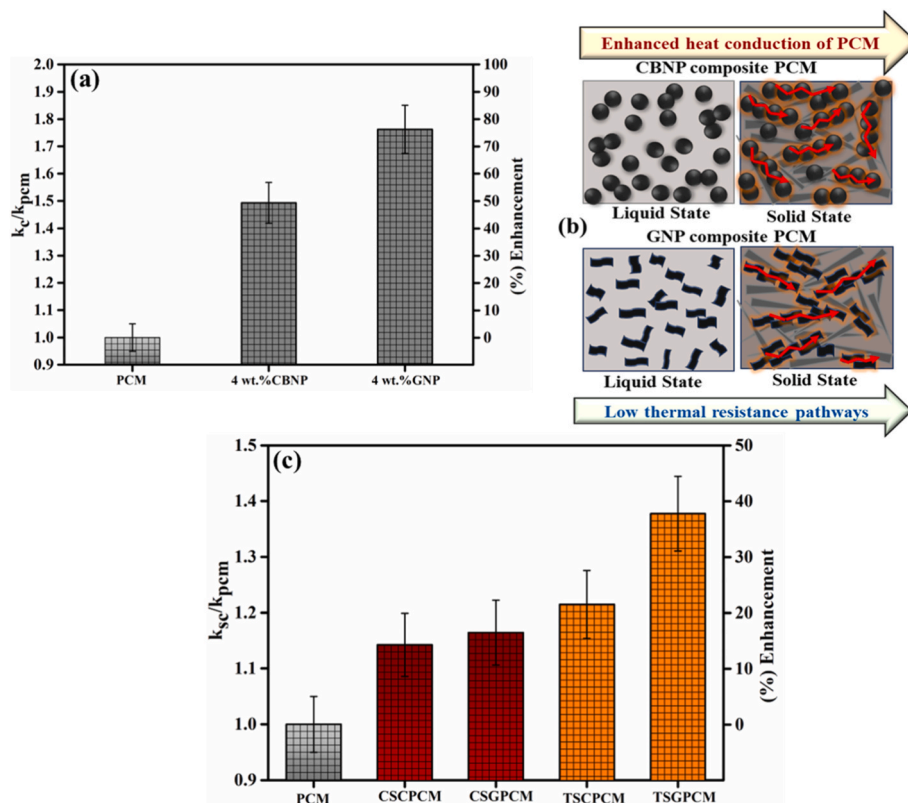


Fig. 5. (a) Variation of k_c/k_{pcm} and related % enhancement for PCM at 4 wt% CBNP and GNP loadings. (b) Schematic representation of percolating network pathways enhancing heat transfer in composite PCMs. (c) Variation of k_{sc}/k_{pcm} and related % enhancement for coffee and turmeric-based ss-composite PCMs. k_c , k_{sc} and k_{pcm} indicate the effective TC of the nanocomposite PCM, ss-composite PCM, and pristine PCM in the solid state, respectively.

Table 4

Thermal properties of all investigated samples.

Samples	Thermal conductivity (TC) (W/(m·K)) (± 5 %)	Thermal diffusivity (mm^2/s) (± 10 %)	TC (%) enhancement/respect to pure PCM
LA (PCM)	0.219	0.102	0
4 wt% CBNP	0.327	0.222	49.3
4 wt% GNP	0.385	0.255	75.8
CSCPCM	0.250	0.146	14.3
CSGPCM	0.255	0.189	16.4
TSCPCM	0.265	0.165	21.0
TSGPCM	0.301	0.205	37.4

4.1. Improved shape-stability of PCM

The shape-stability assessment of pristine PCM and its corresponding coffee/turmeric-based ss-PCMs was evaluated as a function of time under constant load-bearing pressure stress ($\sim 212.6 \text{ kN/m}^2$). As illustrated in Fig. 6 (a), the bare PCM fails to maintain its structural integrity within a brief period (20 min). Instead, the coffee/turmeric-based ss-PCMs demonstrated significant structural integrity performance for over 20 h at room temperature. This indicates that the proposed coffee/turmeric-based ss-PCMs have enough mechanical strength to avoid liquid PCM leakage/cracking in engineering applications. Furthermore, the material leakage test was also carried out at phase transition ($\sim 45^\circ\text{C}$). The pristine and ss-composite PCMs were placed on a hot plate maintained at $\sim 50^\circ\text{C}$: Fig. 6 (b) (I-VI) shows pure PCM and ss-composite PCMs (CSCPCM and TSGPCM) during heating at time $t = 0$ to up to 300 s across the phase transition zone. It can be seen from Fig. 6 (b) (IV) that significant leakage is observed in the case of pristine PCM, whereas

material leakage is effectively controlled for the ss-composite PCMs cases (V-VI). Furthermore, the shape-stability/leakage test of CSCPCM was checked against applied compressed force through a weight of 1000 g ($\sim 31.2 \text{ kN/m}^2$) (Fig. 6 (c)). It was observed that such bio-based supporting matrices effectively maintained structural integrity, even under mechanical stress at temperatures exceeding the melting point (Table 5). The above results reveal that coffee and turmeric support have substantial potential to improve shape-stability and mechanical strength. Their distinct material characteristics related to the improvement of light-harvesting and microstructural properties have been previously explored [48,81–83], however, the sustainability of such cost-effective composite PCMs along with their synergistic contribution with nanostructure characteristics in terms of thermo-optical and thermal management, still needs further exploration. The superior-weight load (stress) bearing characteristics across the phase transition zone reveal their high practical suitability for TES applications.

4.2. Photothermal conversion performance

Fig. 7(a) shows a schematic diagram of the PTC performance investigation. The PTC performance of ss-composite PCM in comparison to pristine PCM was evaluated under simulated sunlight irradiation at an intensity of 150 mW/cm^2 , therefore mimicking concentrated solar energy. Fig. 7 (b) shows a schematic presentation of the basic mechanism of the PTC process: the incorporated nanofillers are rich in π -conjugated electrons, which absorb radiant energy and are then excited from π bonding to π^* antibonding molecular orbitals. After that, excited electrons return to the lower state through lattice relaxation, and a portion of stored radiant energy is converted into heat [87]. Fig. 7(c) displays the temperature variation of different ss-composite PCMs irradiated by such a light source, which were recorded by DAQ system. Intense solar irradiation causes the temperature of ss-composite PCMs to rise rapidly,

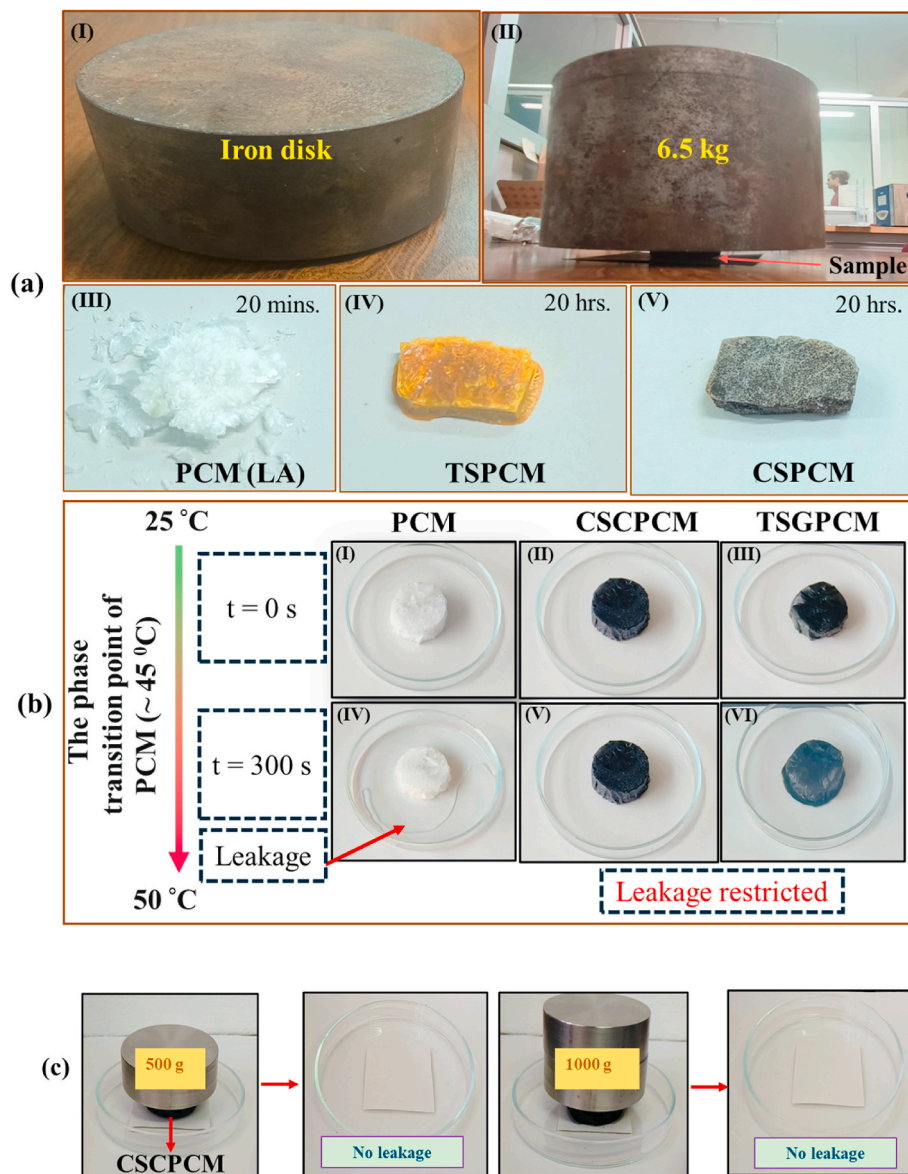


Fig. 6. (a) Investigation of shape-stability test between pristine PCM and coffee/turmeric ss-PCMs (dimensions: 30 mm × 10 mm × 6 mm) under a pressure exerted by 6.5 kg weight (at 25 °C). (b) Typical photographs of the pristine PCM, coffee/turmeric-based ss-composite PCMs across their phase transition temperature. (c) The shape-stability was investigated via leakage test for CSCPCM (radius: 10 mm; height: 10 mm) against compressed loading weights of 500 g or 1000 g at 50 °C.

Table 5

Comparison of the load compression test of the designed ss-composite PCMs with the previous studies.

Sample	Load bear capacity (Compression)	Area (cm ²)	Ref.
PEG-CNC	50 g	–	[84]
LA@CHNP	180 g	7.06	[85]
PCLS-4	200 g	4.00	[86]
CSCPCM	1000 g	3.14	Present study

unlike pure PCMs. Additionally, in the phase transition region, the temperature stabilizes on a plateau value, namely the melting temperature. When the power was turned off, the temperature dropped dramatically and led to the solidification phase at constant temperature during phase change. The experimental findings show that ss-composite PCMs have better PTC performance than PCMs. This could be attributed to the presence of carbon nanofillers (CBNP and GNP), which act as strong light absorbers and converters into heat. This behaviour is coherent with previous studies, which demonstrated that carbon-based

nanofillers like CNTs, GNPs, and EG (expanded graphite) have superior optical absorption in the solar spectrum compared to conventional PCMs. This is due to the resonant π -plasmon excitation/band-to-band transitions under solar radiation [35,88], which leads to an increased extinction coefficient up to $\sim 0.6 \text{ cm}^{-1}$ [19,34,89]. Moreover, the incorporated coffee/turmeric shape-stabilizers also has light absorption characteristics due to melanoidin's pigment and curcumin structure, which synergically contribute with nanofillers to boost the PTC conversion performance of their respective ss-composite PCMs [90–94].

The PTC efficiency (η) is defined as the ratio of the TES during the solid-liquid phase change process to the incident light energy, evaluated as:

$$\eta = \frac{m_s \Delta H_s}{IA(t_e - t_s)}, \quad (1)$$

where ΔH_s , m_s , I , A , and $(t_e - t_s)$ indicate latent heat, mass of the sample, sunlight irradiance, sample area, and phase transition interval (t_e and t_s indicating transition ends/starts time), which are determined by the

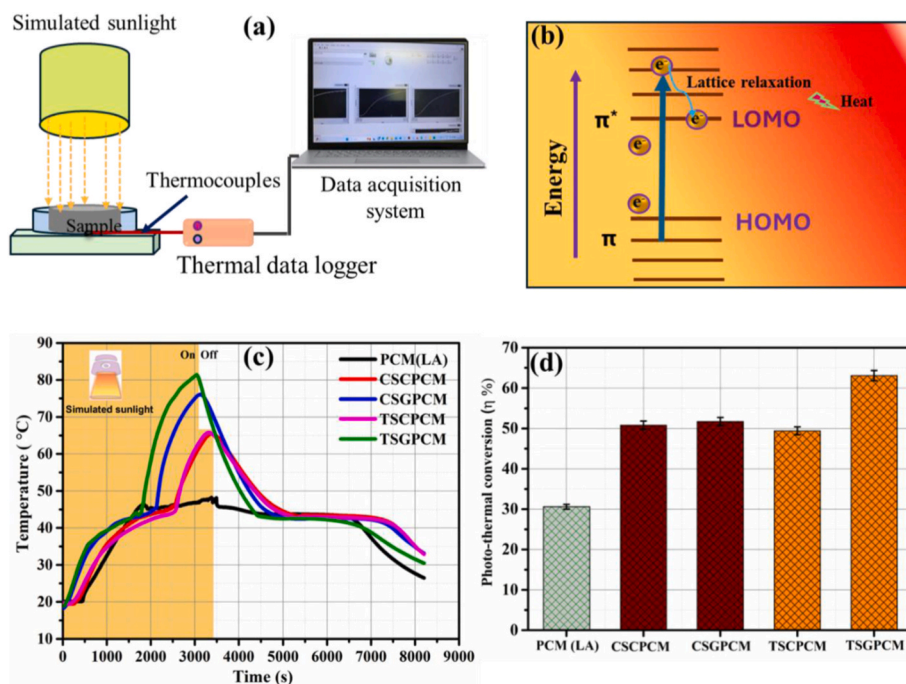


Fig. 7. (a)–(b) Schematic representation of photo-thermal measurement setup and PTC and storage mechanism. (c) Photo-thermal conversion for pristine PCM and coffee/turmeric-based ss-composite PCMs. (d) Investigated PTC efficiency of samples.

tangent method at the phase transition zone of the temperature change curves (intersection of pre and post transition tangent with phase transition (plateau) region [95], respectively). It can be seen from Fig. 7 (d) that the PTC efficiency of ss-composite PCMs was evaluated in comparison to pristine LA, resulting in 30.6 %, 50.8 %, 51.7 %, 49.4 %, and 63.1 % for PCM, CSCPCM, CSGPCM, and TSCPCM, respectively. The ss-composite PCMs show high PTC performance on account of their enhanced light absorption capacity in the UV–Vis–NIR frequencies and effective heat diffusion performance within the host matrix. The TSGPCM shows better PTC performance compared to other PCM-based ss-composites (Fig. 7 (d)): this could be attributed to better heat conduction (Fig. 5 (c)), which comparatively accelerates the LHTES charging process. The parameters related to the calculation of PTC efficiency are listed in Table 6. We also compared the effective enhancement of the photo-thermal efficiency of the proposed bio-composite PCM relative to its host matrix (PCM) with previously reported bio-based and conventional composite PCM enhancements (see Table 7). In summary, the designed composite PCMs have high TES capacity (latent heat) and solar-thermal conversion ability (photo-thermal efficiency).

4.3. Stability and durability assessment

The thermal stability of PCM-based samples is a crucial parameter for their application in real-world applications: to assess it, TGA experiments were carried out. The TGA temperature increment range was set between 25 and 350 °C at a heating rate of 5 °C/min. Fig. 8 (a) shows the weight loss (W, in %) as a function of sample temperature (T) for the

pristine LA and its coffee and turmeric-based ss-composite PCMs (CSCPCM and TSGPCM). All materials show a constant and smooth trend up to 110 °C, thereafter decomposition has started. Up to ~100 % and ~75 % decomposition (weight loss) occurred for the pristine and ss-composite PCMs cases, respectively, just below ~ 240 °C. Fig. 8 (b) shows the variation of differential weight (dW/dT, differential thermal analysis – DTA) as a function of sample temperature for the pristine LA and respective ss-composite PCMs. Single peaks dW/dT curves indicate one-step thermal degradation at ~235, ~227, and ~225 °C for the pristine PCM, CSCPCM, and TSGPCM, respectively. Beyond, 240 °C, the bare PCM was completely decomposed (i.e. evaporated) and the residual weights (25 wt%) corresponded to the residual coffee/turmeric matrices. The TGA and DTA findings reveal that LA and designed ss-composite PCMs exhibit similar thermal characteristics and have excellent thermal stability within the operating range for low-temperature TES applications, namely below 110 °C.

The assessment of thermal stability performance (after several melting/solidification cycles) of ss-composite PCMs is essential for practical TES applications in the long term. To probe the thermal stability of the ss-PCMs in comparison to pristine PCMs, DSC studies were conducted over 500 successive melting and cooling thermal cycles for each sample. Fig. 9(a–b) shows the DSC heat flow curves during repeated (heating/cooling) cycles of PCM-based samples. Even after (500 cycles), the phase transition temperatures and latent heat values of samples remain almost unchanged. Again, the shape-stability (leakage-test) of shape-stabilized PCMs was investigated by performing 100 melting/solidification thermal cycles across a temperature range (25–50 °C) using (forced ventilation oven), and mass loss occurred only

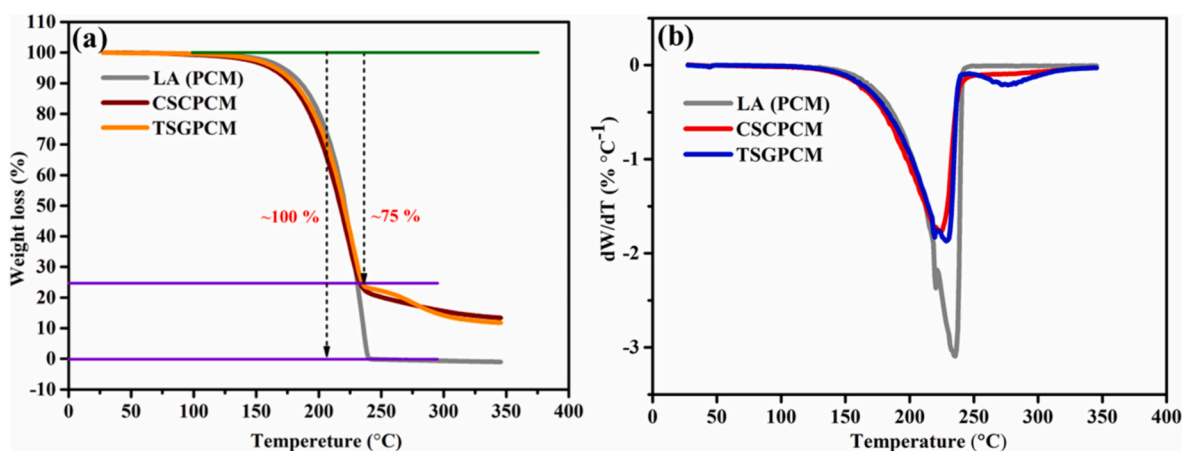
Table 6
Photothermal conversion-related parameters of the samples.

Sample	m_s (g)	t_s (sec)	t_e (sec)	$(t_s - t_e)$ (sec)	ΔH_s (J/g)	A (cm ²)	I (mW/cm ²)	η (%)
PCM(LA)	32	1775	3465	1690	172.5	71	150	30.6
CSCPCM	32	1650	2440	790	133.7	71	150	50.8
CSGPCM	32	1368	2100	730	126.0	71	150	51.7
TSCPCM	32	1721	2535	814	134.1	71	150	49.4
TSGPCM	32	1112	1762	650	137.6	71	150	63.1

Table 7

Comparison of PTC enhancement of bio-based composite PCMs with other reported bio-derived and conventional composite PCMs in related research.

Sample	ΔH_m (J/g)	η (%) enhancement of composite PCM in comparison to host PCM matrix	Light Intensity (mW/cm ²)	Preparation of sample process (complexity)	Ref.
LA/EG-5 %	154.5	34.78	100	complex	[96]
CSC/PEG/GF10	123.6	162.24	150	complex	[97]
CF/LA-MA/ BN35 %	95.05	100	150	complex	[98]
PEG/BPC@M	154.9	47.22	120	complex	[99]
GO3/PEG	84.27	87.3	100	simple	[100]
70 % EG/OP70	126.7	68.20	–	simple	[101]
CW-3/OP44E	206.3	50	100	complex	[102]
PCS600	100.2	54.49	100	complex	[103]
CSGPCM	126.0,	68.95,	150	Very economical and simple	Present study
TSGPCM	142.0	106.20	150		

**Fig. 8.** (a) TGA and (b) DTA curves of pristine PCM (LA) and CSCPCM/TSGPCM ss-composite PCMs, respectively.

1.78 and 5.14 wt%, respectively, reveal their practical potential for TES coherently with earlier studies [45,48]. The typical photographic images and minimal mass loss reveal these bio-based ss-PCMs have superior leakage control activity (Fig. 9c–d). Furthermore, the durability of the heat conduction performance of their ss-composite PCMs was also analysed after prolonged 100 phase change cycles. These findings reveal the thermophysical characteristics of designed ss-composite PCMs sustained superior reliability, which shows their potential for sustainable TES and TM applications (Fig. 9-e).

Furthermore, for real-world practical applications (the effect of moisture/humidity on ss-composite PCMs' thermal characteristics is interesting to explore) the thermal stability of designed ss-composite PCMs also demonstrated underwater by carrying out successive melting/cooling cycles across its phase transition temperature with the help of a temperature management furnace. It can be noticed from (Fig. 10-c) that after going through multiple phase change cycles (100 times) the DSC thermal characteristics of these ss-composite PCMs remain unchanged (latent heat ~ 125 J/g; closed to original cases), which reveals their practical suitability against moisture and high potential to sustain and perform in different thermal environments.

4.4. Thermal management performance

Executing effective thermal regulation and management for electronic devices utilizing PCM-based passive techniques remains a current area of interest [16,104,105]. Fig. 11(a) shows a schematic illustration of the experimental setup for investigating the thermal management performance of ss-composite PCMs compared to pristine PCMs. To reveal their TM ability, the variation of interface temperature (T_s) characteristics of pristine PCM and different ss-composite PCMs were recorded during the melting process. A constant uniform heat load (flux)

($q'' = 0.28$ W/cm²) was supplied by a heater pad (RS pro silicon heater) connecting a DC power controller. The temperature variation characteristics at the interface between the Al plate (thickness: 2 mm) and PCM-based sample (thickness: 1 mm) were recorded using (k-type) thermocouples. Experimental findings from Fig. 11(b) show the variation in interface temperature (T_s) for Al plate - pristine PCM and Al plate - ss-composite PCMs cases, which underwent distinct behavior compared to bare Al plate - air (without PCM). This is attributed to PCM having high latent TES ability across its phase transition region and being helpful in effective heat storage/thermal management. Furthermore, findings reveal that TM performance is significantly enhanced due to the employment of ss-composite PCMs, since a more stable temperature control and superheat degree minimized by approximately 10 °C (indicated by purple arrow, Fig. 11-b). The thermal cycling performance reveals that the TM activity (blue region: effective heat storage/TM control) of the designed ss-composite PCMs is quite durable (across phase transition zone) and suitable for long-term TM applications (Fig. 11-c). The TM characteristics of our designed bio-based ss-composite PCMs highlight their efficacy in TM fields as resuming the earlier demonstrated ss-composite PCM's TM performance in BTMS applications [23,106–109]. This is attributed to the designed nano-enhanced ss-composite PCMs, which have high heat conduction and power density, significantly boosting the TM capacity, especially for applications involving electronic components and batteries.

5. Conclusion

In the present work, coffee/turmeric bio-based ss-composite PCMs are designed to the demand for effective and sustainable solutions for TES and management. The synergic contribution of nanofillers (CBNP/GNP) with supporting matrix (coffee/turmeric) characteristics boost the

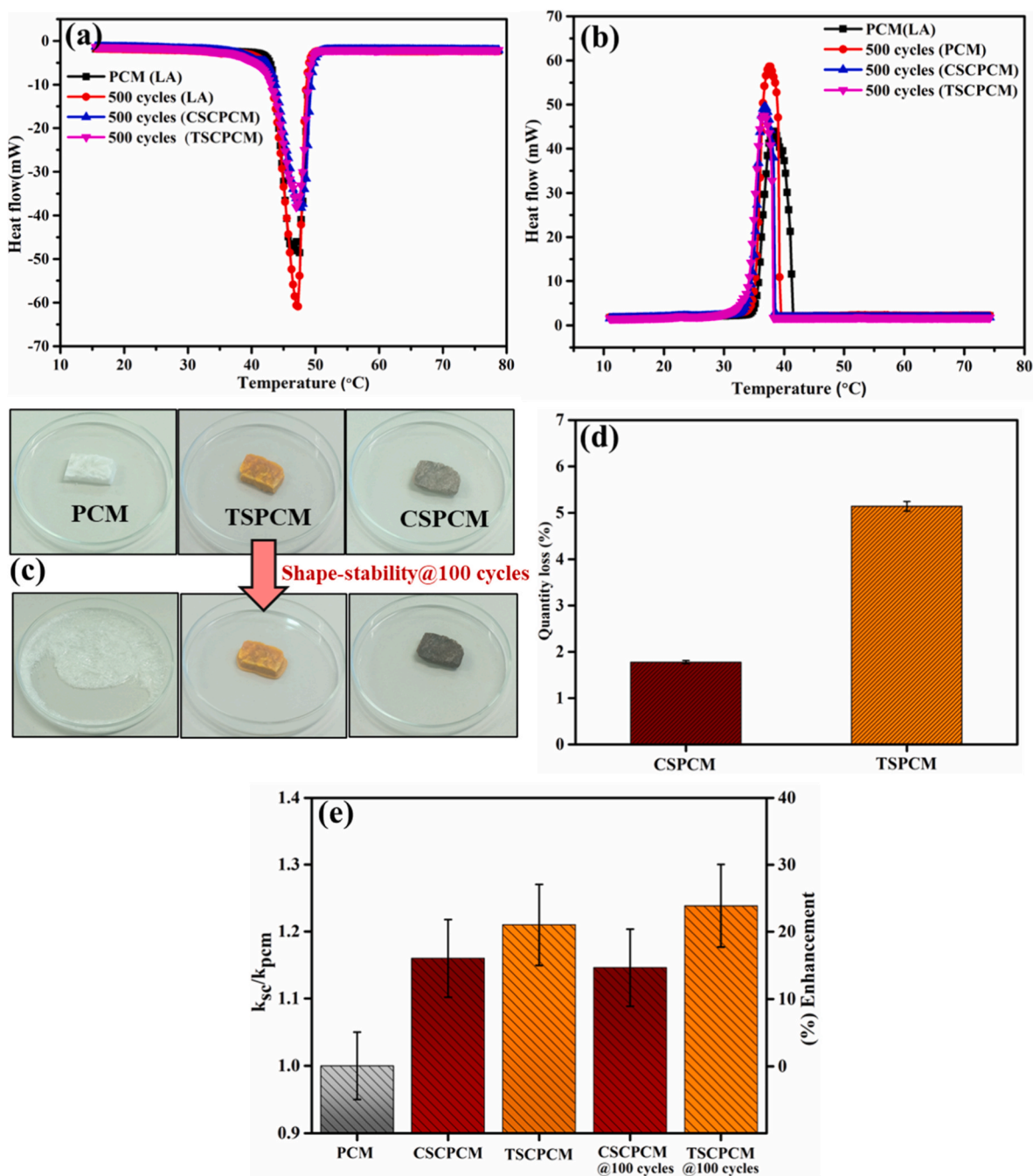


Fig. 9. (a)–(b) Heat flow curves during melting/cooling of pristine PCM and coffee/turmeric-based ss-composite PCMs. (c) Shape-stability performance of ss-composite PCMs by repeated thermal cycles across their phase transition temperature. (d) Evaluated sample’s mass loss (%) after 100 cycles. (e) Durability of thermal conduction characteristics of ss-composite PCMs after 100 cycles.

thermo-optical, and mechanical properties of pristine PCM. The experimental findings demonstrate that nano-enhanced ss-composite achieved high PTC efficiency enhancement (~106.28 %) and structural integrity by preventing PCM leakage across the phase transition zone. This is attributed to GNP and CBNP nanofillers exhibiting strong light absorption characteristics and improved heat dissipation performance within composite through their conductive pathways. The porous characteristics of coffee and turmeric power effectively minimized the mass loss (<5 %) after ~100 thermal cycles, demonstrating their excellent shape stability, this stability is attributed to capillary forces and surface tension activity between the ss-porous matrix and PCM. The designed ss-composites show a high TES density (latent heat value) of approximately 130 J/g across the phase transition zone. The thermal

cycles testing reveals the designed ss-composite PCMs have high thermal stability and LHTES performance. The ss-composite PCMs show superior TM activity (minimizing the superheat degree by ~ 10 °C) and their durable thermal features encourage their execution for long-term applications. The designed low-cost and biodegradable nano-enhanced ss-composite PCMs exhibit high thermo-optical characteristics and shape stability, making them suitable for sustainable energy storage and conversion. As cost-effective bio-composite materials, they have the potential to play a significant role in low-temperature energy storage (LHTES) applications, including thermal regulation of buildings, electronics cooling, solar-thermal systems, textiles/wearables, and battery thermal management. Despite such advancement of bio-based composite PCMs for effective TES and solar-thermal conversion. The future

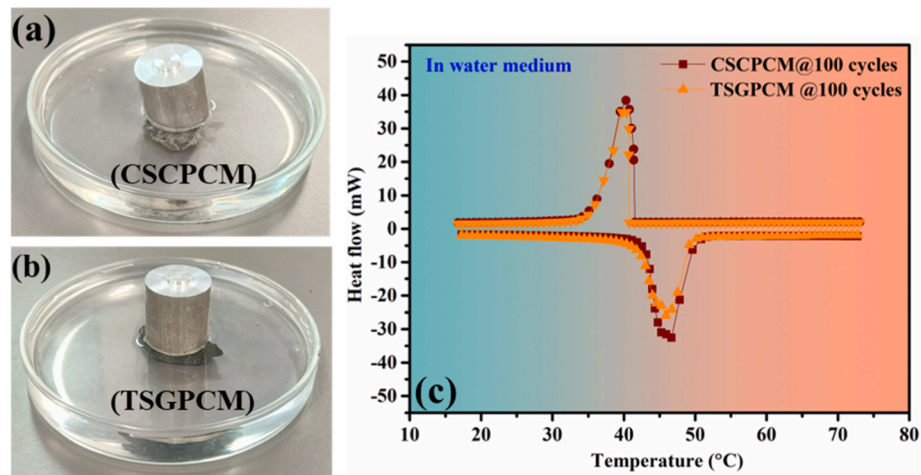


Fig. 10. (a)–(b) show the typical photographic images of CSCPCM and TSGPCM ss-composite PCMs that went through 100 thermal cycles (melting/solidification) underwater; (c) DSC curves of respective ss-composite PCMs after completion of 100 thermal cycles.

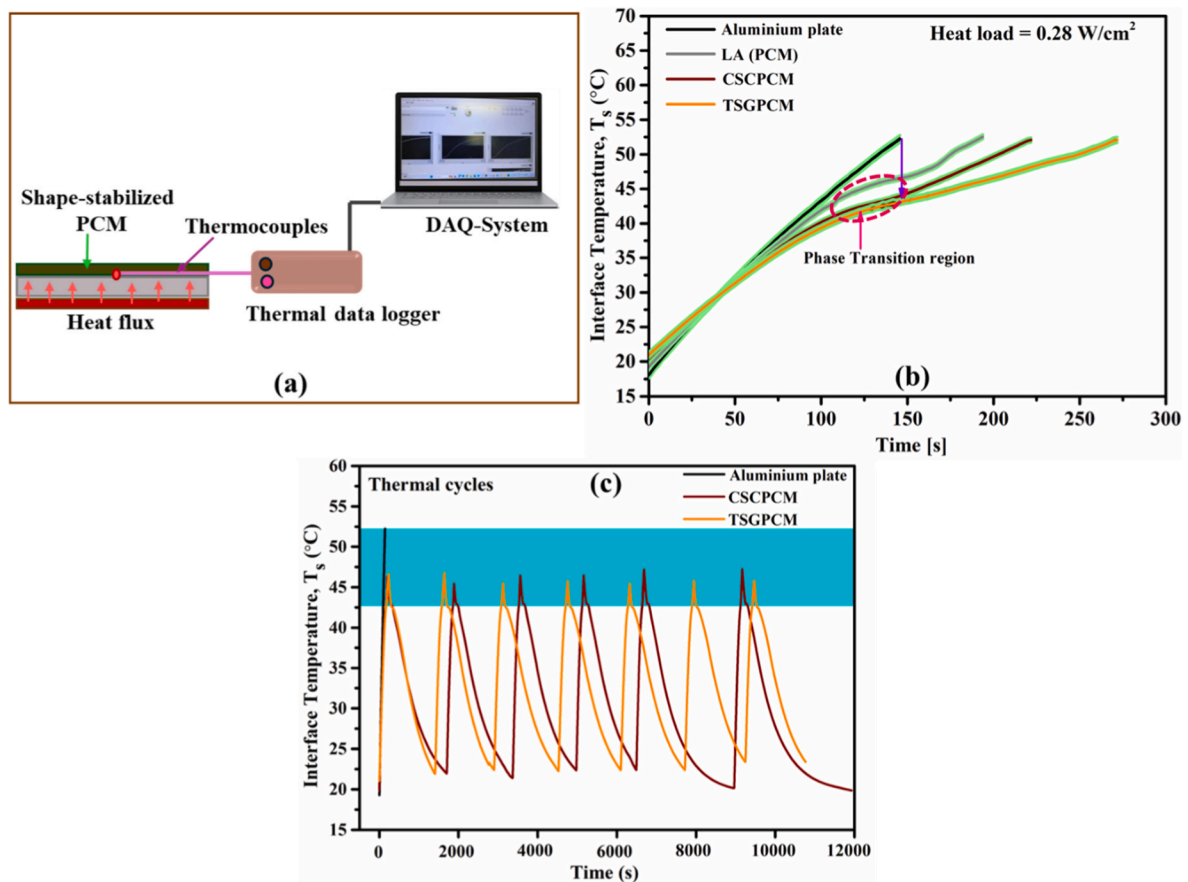


Fig. 11. (a) Schematic of thermal measurement performance setup; (b) Temperature evolution of pristine PCM and Coffee/Turmeric-based ss-composite PCMs; (c) Investigated the TM activity of ss-composite PCMs by performing thermal cycles. The error band (green trace) represents the possible measurement uncertainty. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

research should focus on optimization of additive concentration, long-term thermal/chemical stability, and testing their TES performances under field operation.

CRedit authorship contribution statement

Amit Kumar Mishra: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation,

Conceptualization. **Matteo Morciano:** Writing – review & editing, Supervision, Formal analysis, Data curation. **Elena Campagnoli:** Writing – review & editing, Resources, Investigation. **Valter Giaretto:** Writing – review & editing, Resources, Investigation. **Matteo Fasano:** Writing – review & editing, Supervision, Formal analysis, Data curation. **Eliodoro Chiavazzo:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Eliodoro Chiavazzo reports financial support was provided by Ministero dell'Ambiente e della Sicurezza Energetica (MASE). If there are other authors, they 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

Data will be made available on request.

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