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Modeling the thermal behavior of multifunctional syntactic foams containing phase change materials for heat management applications

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Abstract

Syntactic foams are lightweight porous composites obtained by the addition of hollow glass microspheres (HGMs) to a polymer matrix. This work experimentally and numerically investigates the thermal behavior of epoxy-based syntactic foams with enhanced multifunctionality produced by incorporating microencapsulated phase change materials (PCMs) as functional fillers, providing thermal energy storage and temperature stabilization due to its large latent heat of melting. Foams are fabricated using varying ratios of HGMs (0–40 vol%) and PCM (0–40 vol%) to achieve tuned densities and thermal properties with a total filler content of up to 40 vol%. A one-dimensional numerical heat transfer model based on the enthalpy method is presented to simulate the thermal behavior of these materials. The model accurately incorporates the specific heat and thermal conductivity of the foam components, as well as the latent heat absorption during PCM melting (up to 68 J/g). The model is experimentally validated by demonstrating good agreement with the measurements of transient temperature profiles during heating tests on the foam samples. The validated tool is then leveraged to model the thermal response of the foams under a sinusoidally varying heat source, similar to the possible real applicative scenarios. These results can help optimize foam compositions balancing energy storage density, heat transfer properties, and structural support for lightweight energy storage systems. Potential applications include high-efficiency thermal management in battery packs, electronic cooling, solar energy storage, and sustainable temperature-controlled buildings.

Highlights

- Foams with 0–40 vol% PCM and 0–40 vol% HGM show up to 68 J/g latent heat.

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- 1D numerical model accurately captures thermal conductivity and PCM phase change.
- Validated model simulates thermal response under sinusoidal heat, optimizing foam composition.
- Foams act as thermal reservoirs, maintaining up to 5°C lower surface temperatures than epoxy.

KEYWORDS

epoxy resin, hollow glass microspheres, multifunctional composites, phase change materials, syntactic foams, thermal energy storage

1 | INTRODUCTION

Thermal energy storage (TES) and management using phase change materials (PCM) is an area of increasing interest.^{1,2} Organic PCMs such as paraffins, poly(ethylene glycol)s, and fatty acids and alcohols can store and release heat at a nearly constant temperature, bridging the gap between energy supply and demand.³ This enables the efficient use of solar energy and other renewable resources,⁴ as well as heat management and temperature control in buildings,⁵ textiles,⁶ electronics,⁷ batteries,^{8–13} hydrogen tank cooling,¹⁴ domestic refrigeration,¹⁵ and more.^{16,17} Although they have high latent heat (up to 250 J/g), tunable melt point, low density, and low cost, their main drawback is their leakage above the melting temperature.¹⁸ This issue is generally solved via micro- and nano-encapsulation,¹⁹ which can confine the material, prevent interactions with the surrounding environment, accommodate volume changes during the phase transition, and enhance heat transfer.^{20,21}

Typically, heat management is handled by separate supplementary components, such as fans or finned structures, which can be bulky and add weight and volume.^{22,23} A more efficient alternative would be to build this function into structural elements using multifunctional composites, wherein PCMs can be incorporated as a functional filler along with reinforcing agents to obtain a composite with balanced structural and heat storage properties.^{24,25} Although the concept of a composite combining structural and heat management functions has been studied extensively in the buildings field, the evaluation has been limited to embedding PCMs in concrete and gypsum materials, which show considerable potential.^{26–29} Conversely, little research has been conducted on polymer composites.^{30,31} Our group has explored this concept by developing polymer/PCM/reinforcement systems using paraffinic PCMs within thermoplastic or thermoset matrices and continuous or discontinuous reinforcing fibers. This allows thermal management to be integrated into the

material, rather than requiring separate, dedicated components.^{32–40}

Our group has recently incorporated paraffin microcapsules into another type of structural composite that is, epoxy/hollow glass microsphere syntactic foams (SFs). Polymeric SFs are lightweight, closed-cell porous materials composed of a polymer matrix and rigid hollow particles,^{41,42} whose microstructure provides mechanical properties superior to those of bulk materials or traditional foams on a per-mass basis.^{43–45} This feature, combined with their thermal, electrical, and acoustic insulation and fire resistance, makes them attractive for weight-critical aerospace, transportation, and marine applications.^{46–49} Epoxy foams with hollow glass microspheres (HGMs) have been widely studied for their tunable density and thermomechanical properties, which depend on HGM size, volume fraction, and shell thickness. Generally, a high HGM content and thin shells reduce density but limit strength and impact resistance, despite specific properties often increasing with the HGM fraction.^{50,51}

Incorporating paraffin microcapsules into epoxy/HGM SFs imparts TES capability, thereby further expanding the property set. Our group's recent work on epoxy/HGM/PCM multifunctional syntactic foams^{52,53} demonstrated versatile tuning of the thermal and mechanical properties of these materials for the first time. Incorporating HGM significantly reduced the density compared to that of neat epoxy, thereby considerably increasing the specific tensile and compressive moduli, although the impact strength decreased owing to brittleness. The PCM inclusion provided a thermal energy storage capacity of up to 68 J/g while retaining specific tensile properties and restraining HGM-induced embrittlement. Modifying the HGM and PCM contents allowed balancing of the mechanical performance, thermal conductivity, and heat storage ability. Foams with 20–30 vol % PCM showed robust specific moduli, reasonable impact resistance, thermal conductivities around 0.16–0.17 W/

(m K), and heat storage of 45–68 J/g. Improvements in the interfacial adhesion could further enhance the mechanical performance.

Our previous work highlighted the effectiveness of this concept and showed the relationship between the composition and final thermomechanical properties of these materials experimentally. However, proper modeling of the heat transfer in these multiphase materials is still lacking. More specifically, the study and validation of a model that captures the effectiveness of the introduction of PCMs in epoxy/HGM syntactic foams to increase the thermal buffering capacity of these materials is yet to be developed. This would enable a broad comprehension of the thermal behavior of these materials and thus their widespread industrial application.⁵²

Theoretical and numerical modeling provide a powerful complement to the experimental characterization of the heat transfer properties of epoxy/HGM/PCM multifunctional syntactic foams. Physics-based simulations can predict the thermal performance of foams under various conditions, saving significant time and resources compared with physical testing alone. Models allow exploring a wider range of material compositions and microstructural parameters to optimize the heat storage capacity, thermal conductivity, and temperature profiles for different applications. In this specific case, an accurate model capturing the interplay between the different constituents (epoxy matrix, HGM, and PCM microcapsules) and the phase transition dynamics is crucial for predicting the overall thermal behavior. Moreover, numerical modeling can predict the thermal behavior of materials characterized in different settings from those used for testing. This allows for checking the material suitability for specific applications without costly prototyping, for example, in conducting feasibility studies.

Therefore, this work aims to study heat transfer in epoxy/HGM/PCM syntactic foams both experimentally and theoretically. While previous studies have experimentally characterized the thermal and mechanical properties of such epoxy/HGM/PCM syntactic foams, this work presents, for the first time, the development and validation of a numerical heat transfer model to simulate the complex thermal behavior of multifunctional syntactic foams with enhanced thermal buffer capability. The model incorporates phase change phenomena to enable design optimization and performance prediction under various conditions without extensive physical testing.

The preparation of epoxy/HGM/PCM syntactic foams of various compositions was initially performed, followed by a comprehensive characterization of their thermal properties. A model for the heat transfer within these foams was subsequently developed by considering a constant heat source. To validate the accuracy and reliability

of this model, an experiment tailored for the validation of heat transfer was conducted. Once validated, the model was utilized to simulate the thermal response of syntactic foams when subjected to a dynamic (i.e., oscillatory) heat source. This simulation aimed to showcase the potential of these foams in providing superior thermal insulation and acting as a thermal reservoir compared to syntactic foams that do not contain PCM.

2 | MATERIALS AND METHODS

2.1 | Materials

The epoxy system was a bicomponent mixture, EC157/W342, kindly provided by Elantas Europe Srl (Collecchio, Italy). The epoxy base EC157 had a density of 1.15 g/cm³ and a viscosity of 600–800 mPa s at 25°C, while the hardener W342 had a density of 0.95 g/cm³ and a viscosity of 30–70 mPa·s at 25°C, as declared by the producer's data-sheet. The employed microencapsulated PCM MPCM43D, provided by Microtek Laboratories Inc. (Dayton, USA), was composed of a paraffinic core and a melamine-formaldehyde shell (85:15 wt:wt) with a mean size of 17–22 µm, and a density of 0.9 g/cm³. The declared core's melting temperature was 43°C and the total melting enthalpy was 190–200 J/g. Hollow glass microspheres (HGMs) K15 were provided by 3 M Italia Srl (Piolto, Italy) (density = 0.15 g/cm³, mean particle size (D_{50}) = 60 µm, crush strength (90% survival) = 2.07 MPa; thermal conductivity = 0.055 W/(m K)). All materials were used as received.

2.2 | Sample preparation

The epoxy/HGM/PCM syntactic foams were prepared as described in previous works of our group.^{52,53} Briefly, the epoxy base, PCM, and HGMs were mixed in a beaker for 5 min at 100 rpm with a mechanical mixer Dispermat F1 (VMA-Getzmann GmbH, Germany) and then degassed through a vacuum pump for 5 min. The hardener was then added to the mixture, which was mixed and degassed again, cast in silicone molds, cured for 24 h at room temperature, and post-cured for 6 h at 80°C. This step was repeated for all four compositions relevant to this study, which comprised neat epoxy resin (EPG-0.0) and three samples with a total filler volume fraction of 40 vol%, and namely EPG-0.40, containing 40 vol% of HGM, EPG-40.0, containing 40 vol% of PCM, and EPG-20.20, containing 20 vol% of HGM and 20 vol% of PCM. The prepared samples and their nominal weight and volume composition are listed in Table 1. A more detailed

TABLE 1 List of the prepared samples with nominal weight and volume compositions.

Sample	Epoxy (vol%/wt%)	PCM (vol%/wt%)	HGM (vol%/wt%)
EPG-0.0	100.0/100.0	0.0/0.0	0.0/0.0
EPG-0.40	60.0/91.6	0.0/0.0	40.0/8.4
EPG-40.0	60.0/64.6	40.0/35.4	0.0/0.0
EPG-20.20	60.0/75.8	20.0/20.7	20.0/3.5

description of the sample preparation is reported in our previous work.^{52,53}

2.3 | Experimental characterization

The microstructure of the prepared samples was investigated by observing the fracture surface (obtained as described in our previous works^{52,53}) using a Zeiss Supra 40 (Carl Zeiss AG, Oberkochen, Germany) field-emission scanning electron microscope (SEM). The samples were made conductive via Pt-Pd sputtering.

Differential scanning calorimetry (DSC) of the neat PCM was performed using a Mettler DSC30 calorimeter (Mettler Toledo LLC, Columbus, USA). Specimens with a total mass of approximately 10 mg were sealed in aluminum crucibles and subjected to a thermal ramp between 10 and 60°C at a rate of 1°C/min under a N₂ flow of 100 mL/min. These tests led to the measurement of the melting temperature and enthalpy of the PCM (T_m , ΔH_m). On the other hand, the complete characterization of the prepared composites was reported in a previous work of our group,⁵² which also describes in detail the preparation of the specimens for the thermal analysis.

The thermal diffusivity and conductivity of all prepared syntactic foams were determined using an LFA 467 light flash analyzer (Netzsch Holding, Selb, Germany). Tests were performed at 30°C on square specimens ($12.7 \times 12.7 \times 4$ mm³) cut from cast plates and coated with graphitic spray. Three pulses were applied to each specimen. These tests allowed direct measurement of the thermal diffusivity (α). Then, the thermal conductivity (λ) was calculated through Equation (1), as

$$\lambda = \alpha \cdot \rho \cdot c_p, \quad (1)$$

where ρ is the pycnometer density (at 23°C) and c_p is the specific heat calculated via DSC, as reported previously.⁵²

A lab-scale experiment was performed to analyze the operating principles of this material. The experiment was performed on cylindrical cast specimens (diameter:

27 mm; height: 38 mm), as depicted in Figure 1. Blind holes with a diameter of 2 mm and depth of 13 mm were machined along the height of the cylinders to allow the insertion of a type-K thermocouple connected to a data logger. Thermocouples were placed to measure the temperature along the cylinder axis at the distances of 3, 13, 23, and 33 mm from the bottom base. These specimens were placed in an insulating mantle made of polystyrene foam and then placed ($t = 0$ s) onto a hot plate ($T = 86^\circ\text{C}$), while the top base was left exposed to the surrounding air ($T = 26^\circ\text{C}$). The temperature was recorded at one point per second. Thus, it was possible to determine the time required for each thermocouple to reach a stationary temperature during heating.

2.4 | Theoretical modeling

It is widely recognized that problems involving moving boundaries where phase changes occur, known as Stefan problems, can be effectively solved using the so-called enthalpy method.^{54–56} This is particularly true for one-dimensional domains, such as that presented in this paper, where heat conduction occurs in only one direction (Figure 1). The problem can then be simulated using the one-dimensional equation of heat conduction, written using specific enthalpy H explicitly, as reported in Equation (2):

$$\rho \frac{\partial H}{\partial t} = \frac{\partial}{\partial x} \left(\lambda \frac{\partial T}{\partial x} \right), \quad (2)$$

in which T is temperature, t is time, x is the spatial coordinate, ρ is density and λ is thermal conductivity. Equation (2) must be solved coupled with one initial condition and two boundary conditions, which depend on the specific problem, as described hereafter.

This study addressed two problems. In the first one (Problem 1), the experimental measurements described in Section 2.3 were reproduced to validate the model. For such a case, Equation (2) was subjected to the initial and boundary conditions represented by Equation (3) and Equations (4)–(5), respectively,

$$T = T_0 \quad t = 0; \quad 0 < x \leq L, \quad (3)$$

$$T = T_1 \quad x = 0; \quad t > 0, \quad (4)$$

$$-\lambda \frac{\partial T}{\partial x} = h(T - T_0) \quad x = L; \quad t \geq 0 \quad (5)$$

in which $T_0 = 26^\circ\text{C}$ is the initial temperature, equal to room temperature, $T_1 = 86^\circ\text{C}$ is the heat source

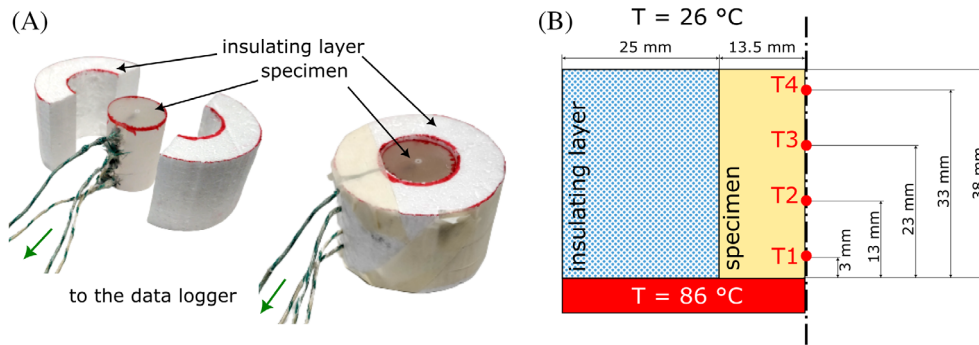


FIGURE 1 Experimental setup to test one-dimensional heat transfer. (A) Cylindrical specimen with thermocouples and open (left) and closed (right) insulating layer. (B) Schematic representation of the experiment, with the bottom base of the cylinder in contact with a hot plate at 86°C and the top base exposed to the external environment at 26°C. T1, T2, T3, and T4 represent the thermocouple positions.

temperature, $L = 38$ mm is the total length of the specimen, and h is the film coefficient. For this case, a film coefficient of $15 \text{ W/m}^2/\text{°K}$ was used to simulate heat transfer by natural convection in calm air.

In the second case (Problem 2), after validation, the model was used to predict the thermal conduction through syntactic foams resulting from a sinusoidally varying heat source, as a case of potential practical interest. Such a problem can be interesting for gaining insight into the applications of such foams as thermal buffering materials. The simulations were conducted on a 10-mm thick specimen with a sinusoidal heat source varying between 12 and 72°C with a period of 15 min; thus, four cycles were completed in 1 h. This case is intended as an example of what can be achieved using these materials, but the same procedure can be repeated for different periods and specimen thicknesses.

In the case of Problem 2, Equations (3)–(5) take the form described in Equations (6)–(8).

$$-\lambda \frac{\partial T}{\partial x} = h(T - T_0) \quad x = L; \quad t \geq 0. \quad (8)$$

To solve these two problems, it is necessary to provide the specific enthalpy as a function of temperature, along with the parameters describing the thermal behavior of the materials. For materials that do not undergo phase change in the considered temperature interval, the specific enthalpy can be simply given by Equation (9), as

$$H = c_p(T - T_0). \quad (9)$$

If, on the other hand, a phase change is present, enthalpy will have a more complex temperature dependence. For instance, if melting occurs at $T = T_m$, a suitable form for enthalpy is pictured in Figure 2, which, despite being quite simple, still describes the physical phenomenon accurately. Using data from the DSC scans, this enthalpy behavior can be schematized using Equation (10):

$$H = \begin{cases} c_p(T - T_0) & T_0 \leq T \leq T_m - \frac{\Delta T_m}{2} \\ c_p \left(T_m - T_0 - \frac{\Delta T_m}{2} \right) + \frac{\Delta H_m}{\Delta T_m} (T - T_m) + \frac{\Delta H_m}{2} & T_m - \frac{\Delta T_m}{2} \leq T \leq T_m + \frac{\Delta T_m}{2} \\ c_p(T - T_0 - \Delta T_m) + \Delta H_m & T \geq T_m + \frac{\Delta T_m}{2} \end{cases} \quad (10)$$

$$T = T_0 \quad t = 0; \quad 0 \leq x \leq L, \quad (6)$$

$$T = T_0 + \frac{T_1 - T_0}{2} (1 - \cos \omega t) \quad x = 0; \quad t \geq 0, \quad (7)$$

where ΔH_m is the enthalpy of fusion and ΔT_m is the temperature interval at which phase change takes place. Both quantities can be estimated from the DSC thermogram. In writing Equation (10), it was assumed that the specific heat was the same both above and below the melting

temperature. The so-called mushy region corresponds to the temperature range $T_m - \frac{\Delta T_m}{2} \leq T \leq T_m + \frac{\Delta T_m}{2}$, in which phase change is taking place.

To generalize the problem, Equation (2) and the conditions reported in (3)–(8) were non-dimensionalized using the units reported in Table 2, leading to the initial boundary value problem reported in Equation (11)–(14) that is,

$$\frac{\partial \eta}{\partial \tau} = \text{St} \frac{\partial^2 \vartheta}{\partial \xi^2}, \quad (11)$$

$$\vartheta = 0 \quad \tau = 0; \quad 0 < \xi \leq 1, \quad (12)$$

$$\vartheta = 1 \quad \xi = 0; \quad \tau \geq 0, \quad (13)$$

$$-\frac{\partial \vartheta}{\partial \xi} = \text{Bi} \vartheta \quad \xi = 1; \quad \tau \geq 0, \quad (14)$$

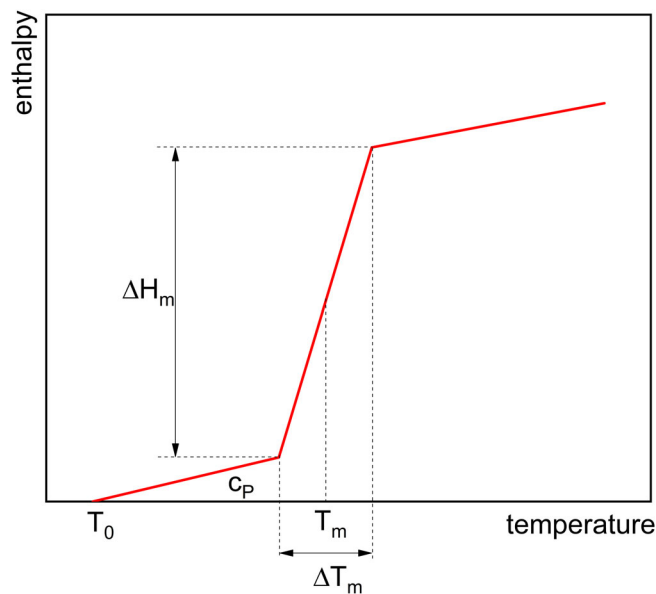


FIGURE 2 Typical specific enthalpy versus temperature curve of a phase change material. Specific heat was assumed to remain constant above and below the melting temperature.

TABLE 2 Experimental data of the prepared syntactic foams used for the model calibration.

Property	Test method	EPG-0.0	EPG-0.40	EPG-40.0	EPG-20.20
ΔH_m (J/g)	DSC	n.a.	n.a.	67.6	44.5
ρ (g/cm ³)	He pycnometry	1.137 ± 0.004	0.735 ± 0.001	1.009 ± 0.001	0.872 ± 0.001
α (mm ² /s)	LFA	0.131 ± 0.004	0.123 ± 0.006	0.127 ± 0.006	0.134 ± 0.004
λ (W/(m K))	LFA	0.199 ± 0.006	0.120 ± 0.006	0.193 ± 0.009	0.168 ± 0.005

Abbreviation: n.a., not applicable.

and in Equations (11) and (15)–(17) for Problem 2, as

$$\vartheta = 0 \quad \tau = 0; \quad 0 \leq \xi \leq 1, \quad (15)$$

$$\vartheta = \frac{1 - \cos \text{Fo} \, t}{2} \quad \xi = 0; \quad \tau \geq 0, \quad (16)$$

$$-\frac{\partial \vartheta}{\partial \xi} = \text{Bi} \vartheta \quad \xi = 1; \quad \tau \geq 0. \quad (17)$$

The above equations use the non-dimensional quantities reported in Equations (18),

$$\vartheta = \frac{T - T_0}{T_1 - T_0}; \quad \eta = \frac{H}{\Delta H}; \quad \xi = \frac{x}{L}; \quad \tau = \frac{\lambda}{\rho c_p L^2} t \quad (18)$$

and the Stefan, Fourier, and Biot numbers defined in Equation (19).

$$\text{St} = \frac{c_p(T_1 - T_0)}{\Delta H}; \quad \text{Fo} = \frac{\omega \rho c_p L^2}{\lambda}; \quad \text{Bi} = \frac{hL}{\lambda}. \quad (19)$$

Also, the non-dimensionalized relationships between enthalpy and temperature that is, Equations (9) and (10), are represented by Equations (20) and (21), as

$$\eta = \text{St} \vartheta, \quad (20)$$

$$\eta = \begin{cases} \text{St} \vartheta & 0 \leq \vartheta \leq \vartheta_m - \frac{\Delta \vartheta}{2} \\ \text{St} \left(\vartheta_m - \frac{\Delta \vartheta}{2} \right) + \frac{1}{\Delta \vartheta} (\vartheta - \vartheta_m) + \frac{1}{2} & \vartheta_m - \frac{\Delta \vartheta}{2} \leq \vartheta \leq \vartheta_m + \frac{\Delta \vartheta}{2} \\ \text{St}(\vartheta - \Delta \vartheta) + 1 & \vartheta \geq \vartheta_m + \frac{\Delta \vartheta}{2} \end{cases} \quad (21)$$

with $\vartheta_m = (T_m - T_0)/(T_1 - T_0)$ and $\Delta \vartheta = \Delta T/(T_1 - T_0)$, respectively.

The relevant initial-boundary-value problems were solved numerically with the finite difference method using 200 linear elements of equal length in the non-dimensionalized domain. Concerning time discretization, the usual Crank-Nicholson method was employed for materials that do not change phase, while in the other cases the approach of Voller and Cross was followed.⁵⁴ Briefly, the equations were integrated using an explicit scheme in time. To reduce the oscillatory disturbances that are typical of explicit schemes, time instants were chosen so to predetermine the location of the midpoint of the mushy region as follows. Using Equation (21), at the midpoint of the mushy region the non-dimensionalized enthalpy is reported in Equation (22), as

$$\eta_m = \text{St} \left(\vartheta_m - \frac{\Delta\vartheta}{2} \right) + \frac{1}{2}. \quad (22)$$

Here, the temperature is clearly equal to ϑ_m . As shown by Voller and Cross,⁵⁴ by equilibrating the thermal fluxes entering and leaving each line element, if this point falls exactly at the midpoint of a certain line element, then the numerical solution is accurate. Then, at every time step, the method employs an initial working run of the explicit scheme to identify the precise time instant when the midpoint of the mushy region coincides with the midpoint of a line element. Once this time instant is known, temperatures at all nodal points at that time instant can be calculated accurately by central finite differences in space. The method is also well described by Date.⁵⁷ Interested readers are strongly encouraged to refer to Figure 2 in the article by Voller and Cross⁵⁴ for a schematic representation of the method.

The authors acknowledge that the thermal properties of materials are generally temperature-dependent. However, the proposed model employs the simplifying assumption of using temperature-independent values. Nonetheless, as will be discussed later (Figure 4), the specific heat of the investigated materials does not change significantly within the considered temperature interval. Moreover, despite density (ρ) and thermal conductivity (λ) change with temperature, their ratio remains mostly unchanged,⁵⁸ thus keeping the thermal diffusivity $\alpha = \lambda / (\rho \cdot c_p)$ approximately constant at all temperatures of interest. Since the transient conductive heat transfer phenomenon primarily depends on α , and α is nearly temperature-independent in the investigated range, this justifies the hypothesis of using a model with temperature-invariable material properties.

3 | RESULTS AND DISCUSSION

SEM micrographs of the fracture surfaces of the prepared syntactic foams are shown in Figure 3A–C. All samples exhibited brittle fracture, as evidenced by the flat fracture surfaces. HGMs (Figure 3A) are homogeneously dispersed within the epoxy matrix with rather poor interfacial adhesion, as indicated by the smooth gaps left by the detached microcapsules and the smooth surface of the remaining ones. However, the SEM micrographs acquired in other zones (not reported, see⁵²) also show broken HGMs, which are empty and have smooth inner sides. This suggests that breakage occurred during the fracture process, not during mixing, thereby confirming that the processing parameters were mild enough to preserve HGM integrity.

Figure 3B shows the microstructure of sample EPG-40.0, which contained 40 vol% PCM. The PCM consists of core-shell spherical microcapsules approximately 20 μm in diameter, whereas the HGMs have a larger diameter ($D_{50} = 60 \mu\text{m}$), agreeing with the producers' datasheets. Both the HGMs and the PCM are well distributed in the samples, and no visible agglomerates are present. The comparison of Figure 3A and Figure 3B shows the size difference between the PCM microcapsules and HGMs. This is also evident in the micrograph of the sample EPG-20.20 (Figure 3C), displaying the relative filler distribution. The smaller PCM capsules were located within the interstices of the larger HGMs, but the size difference does not promote agglomeration or segregation, which ensures the uniformity of the composition and the properties across the whole sample volume.

Moreover, the adhesion between the PCM outer shell and the epoxy matrix is suboptimal, although the fracture appears to propagate across the microcapsules rather than at the interface. Additionally, most PCM capsules retain their rough, irregular paraffinic core, whereas the core of smooth empty capsules has likely remained on the other, complementary fracture surface. The fact that neither the HGMs nor the PCMs contain residues of epoxy resin in their inner surfaces indicates that the fracture of these particles occurred during the microscopy sample preparation and not during processing. This indicates that the PCM integrity is preserved during processing, excluding the possibility of paraffinic leakage from the composite during service. A deeper microstructural characterization of the prepared syntactic foams and a closer look at the filler/matrix interfacial adhesion is reported in our previous work.^{52,53}

DSC was used to measure the melting/crystallization enthalpy and corresponding phase change temperatures of the PCM used to produce the syntactic foams. The

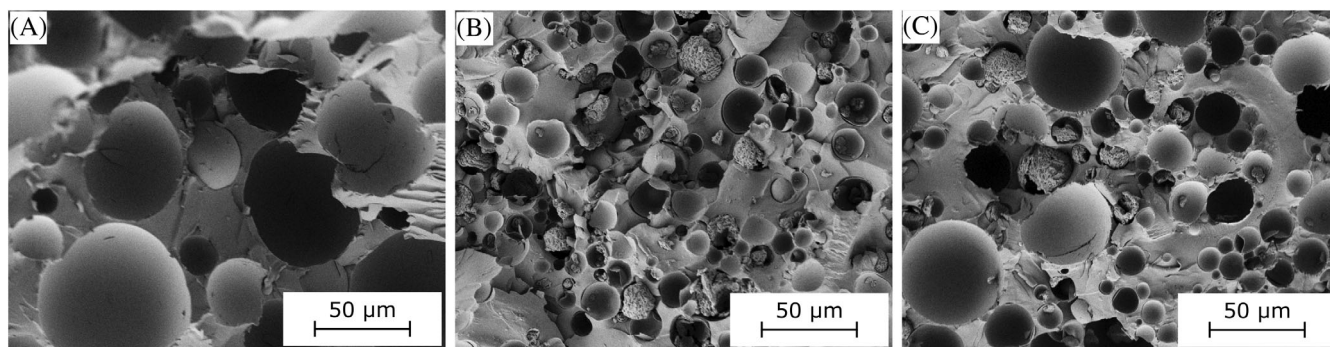


FIGURE 3 Scanning electron microscopy micrographs of the tensile fracture surface of the prepared syntactic foams. (A) EPG-0.40; (B) EPG-40.0; (C) EPG-20.20.

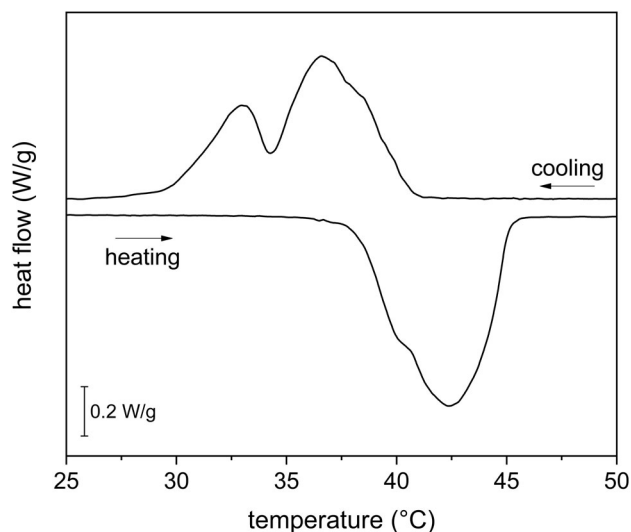


FIGURE 4 Differential scanning calorimetry thermogram (first heating scan and cooling scan) of the neat phase change materials at 1°C/min.

thermogram of the PCM at 1°C/min is reported in Figure 4. The PCM displays a prominent endothermic peak between 35 and 60°C in the first heating scan, associated with the melting of the paraffinic core. The peak melting temperature is 41.2°C and the total melting enthalpy is 213 J/g, in good agreement with the technical datasheet. From this thermogram, the melting temperature of the PCM and its specific heat capacity as a function of temperature were calculated and used for the simulations. The constancy of the measured heat flow before and after the melting peak justifies the hypothesis of unchanging specific heat.

This signal of PCM melting is present also in the DSC thermograms of EPG-40.0 and EPG-20.20 in the same temperature range, as reported in our previous work.⁵² As displayed in Table 2, the total melting enthalpy of these samples is nearly proportional to the nominal weight fraction of PCM and is 67.6 J/g for EPG-40.0 and

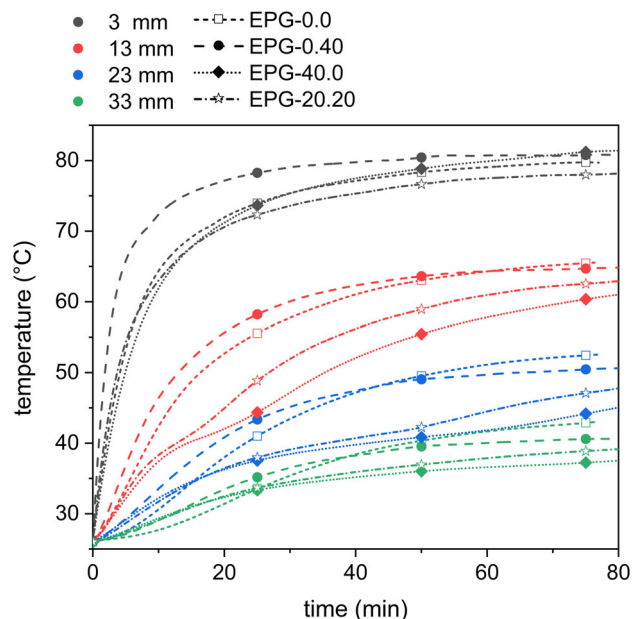


FIGURE 5 Results of thermal transient tests. Temperature as a function of time for the four prepared samples at four different distances from the hot plate (3, 13, 23, and 33 mm).

44.5 J/g for EPG-20.20. These values were also used for the simulations, together with the values of density, thermal diffusivity, and thermal conductivity of these samples, calculated via helium pycnometry and light flash analysis (LFA) as described in our previous work⁵² and also shown in Table 2. Since no compositional or property gradient was found along the cylinder axis or radius, and given the symmetry of the system and the spherical shape of both filler types, the composites were modeled as isotropic.

Figure 5 shows the experimental results of the 1D heat transfer test across the cylindrical specimens and reports the temperature as a function of time measured at the four distances from the hot plate. As expected, the thermocouples at 3 mm from the hot plate are those in

which the temperature rises more quickly, and no evident qualitative or quantitative differences are appreciated as a function of the composition. On the other hand, a significant effect of the PCM starts to be seen from the results of the thermocouples located at 13 and 23 mm from the hot plate, while approaching the PCM melting point. As a result of the latent heat absorbed during the melting event, an evident deflection from a saturated exponential trend can be observed in the samples containing PCM, which delays the heating trend considerably. Moreover, as is evident from the thermocouple located at 33 mm, the maximum temperature reached during the test is lower for all syntactic foams than for

the neat epoxy, as expected from the lower thermal conductivity. As a result, the PCM does not change phase; therefore, no latent heat effect can be observed from the curves. Considering the measurements at 13 mm (red curve), it can be seen that the heating rate typical of the experiment is approximately $1^{\circ}\text{C}/\text{min}$ that is, the same value that was chosen for the DSC test performed on the PCM (Figure 4).

The experimental results shown in Figure 5 must be compared with the numerical results obtained for Problem 1. These are depicted in Figure 6A–D for samples EPG-0.0, EPG-0.40, EPG-40.0, and EPG-20.20, respectively.

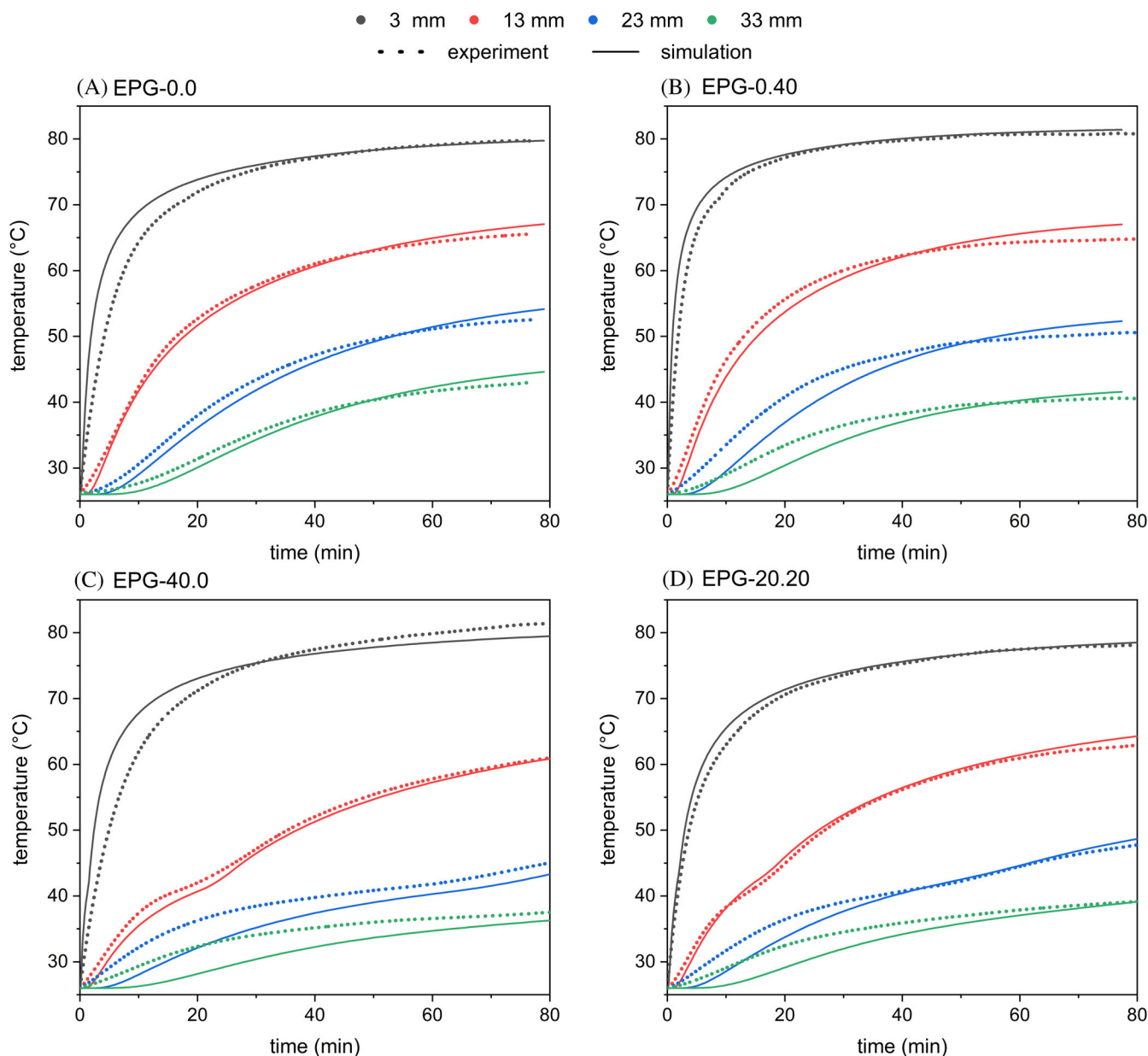


FIGURE 6 Comparison between the model simulation (solid line) and experimental results (dotted line) for (A) EPG-0.0; (B) EPG-0.40; (C) EPG-40.0; EPG-20.20 samples.

The numerical model closely reproduces the experimental findings, particularly for the temperature readings closer to the heat source, specifically at 3 and 13 mm for all materials. All the numerical curves exhibit good qualitative agreement with the experiments, showing a plateau at 35–40°C for the samples undergoing phase change (Figure 6C,D). Moreover, the trends are accurately approximated from a quantitative standpoint, particularly with regard to the long-term behavior after saturation.

The only partial lack of agreement occurs in the case of points placed farther from the thermal source, namely at 23 and 33 mm. Here, for samples EPG-0.40, EPG-40.0, and

EPG-20.20, the experimental data anticipate the numerical model, which displays a certain lag time before the temperature increases. This may be due to inaccuracies in the experimental validation of the model. Conversely, the reported material properties (Table 2) were likely measured correctly because the long-term behavior and the final plateau temperature, dominated by the thermal conductivity, are modeled very closely. Although the precise identification of the root cause of this partial lack of agreement is yet to be identified, the model is still able to predict the most important features of the investigated materials, especially the lag due to phase change, with a good degree of qualitative and quantitative agreement.

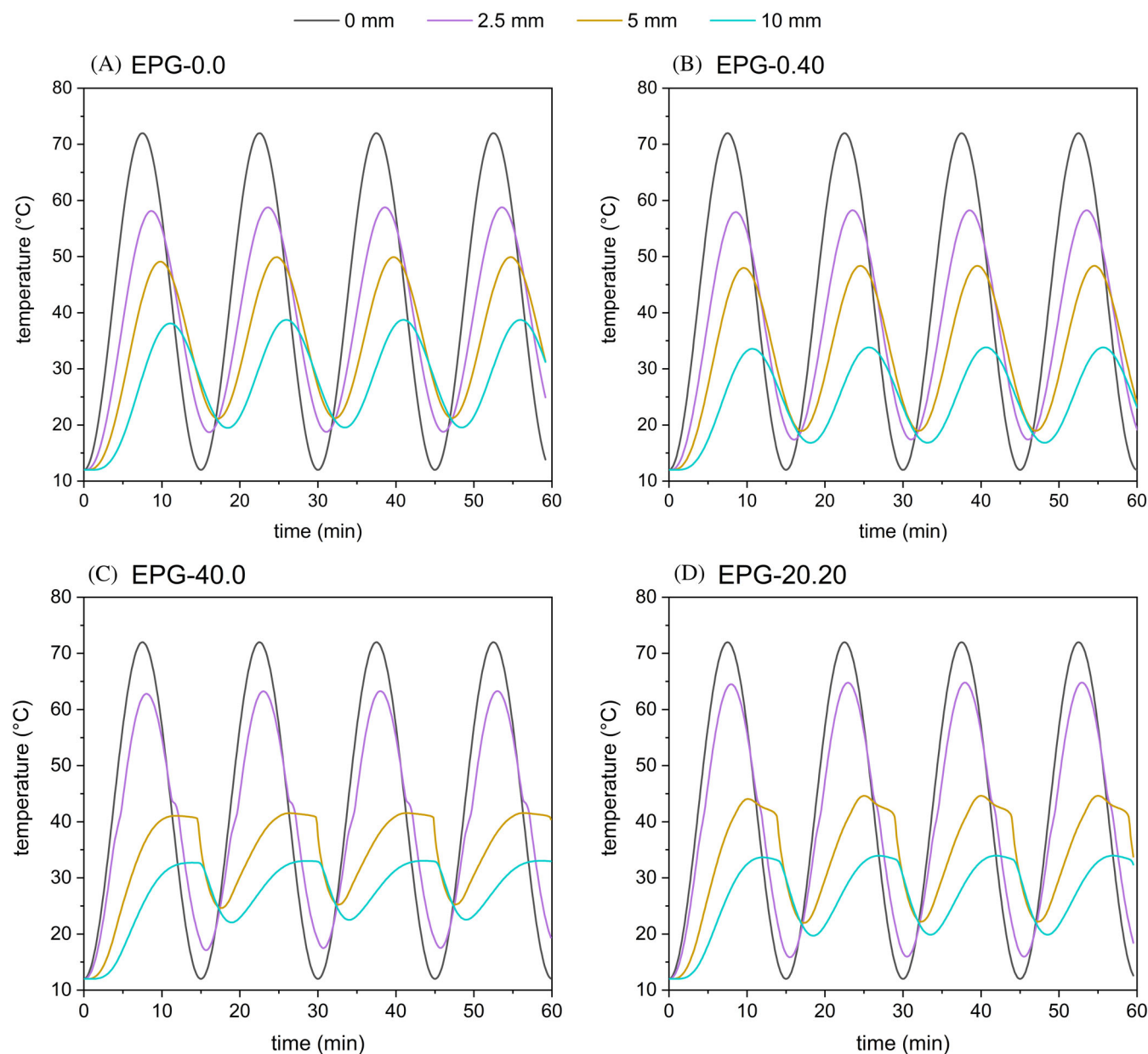


FIGURE 7 Simulation of sinusoidal oscillatory behavior of the heat source for (A) EPG-0.0, (B) EPG-0.40, (C) EPG-40.0, and (D) EPG-20.20.

Based on these observations, it can be concluded that the model was validated. Therefore, it can be used to predict the thermal behavior of these materials in settings that are different from those studied experimentally. As a case of potential practical interest, the model was used to simulate the thermal response resulting from a sinusoidally varying heat source, in what was called Problem 2. The simulations were conducted on a 10-mm thick specimen with a sinusoidal heat source varying between 12 and 72°C with a period of 15 min; thus, four cycles were completed in 1 h. The results are shown in Figure 7A–D.

Figure 7A shows the model predictions for the neat epoxy (EPG-0.0). As can be seen, despite the heat source temperature varying between 12 and 72°C, as superimposed in the simulation, the maximum temperature estimated at the other side of the specimen is much lower that is, approx. 38°C, whereas the minimum temperature at each cycle was approximately 20°C, but the temperature profile as a function of time is perfectly sinusoidal for all considered locations. The same occurs for the simulation of EPG-0.40 (Figure 7B), the biggest difference being the maximum and minimum temperatures of the 10 mm curves that are lower than those of the EPG-0.0 case. This is justified by the lower thermal conductivity of EPG-0.40.

The situation changes considerably for EPG-40.0 and EPG-20.20 (Figure 7C,D). For EPG-20.20, only two temperature profiles are sinusoidal that is, the first at 0 mm, which is obvious because it is the boundary condition, and the second at 2.5 mm from the bottom surface. The other temperature profiles are clearly not sinusoidal, albeit periodical, since they are affected by the phase change occurring at around 42°C. This is particularly evident in the curve corresponding to a position of 5 mm from the bottom surface that is, at half the thickness of the layer. As a result, considering the temperature profile at 1 cm (cyan line in Figure 7), that is, at the top surface, even though the minimum temperature of the EPG-20.20 system is the same as that of neat epoxy, the maximum temperature is at least 5°C lower, thus demonstrating higher thermal inertia. These results are even more evident for EPG-40.0, where, at full thickness, the maximum and minimum temperatures are closer to each other, thus denoting an even higher thermal buffering effect.

In summary, the use of PCM microparticles in epoxy resin increases the thermal inertia of insulating materials significantly, which is beneficial for applications where insulation is needed against rapidly changing external heat sources. Examples of such applications include electronics cooling, building materials, automotive, and refrigerated transportation. For instance, in electronics cooling, PCM-loaded syntactic foams can act as thermal

buffers to prevent overheating of components subjected to intermittent heat loads by absorbing and releasing heat during peak thermal cycles. In building TES systems, these multifunctional foams can store solar heat during the day and release it gradually at night, reducing energy demands for heating and cooling. Similarly, for automotive and aerospace applications, these materials can provide thermal regulation within the vehicle/aircraft structure by mitigating temperature fluctuations from the external environment without adding extra mass, due to the multifunctional nature of these foams. Moreover, the high thermal inertia makes these materials suitable for insulating food packaging, ensuring temperature stability during transportation.

4 | CONCLUSIONS

This work presented for the first time the development and validation of a 1D numerical model for predicting heat transfer in epoxy/HGM/PCM syntactic foams for thermal management applications. The enthalpy-based model accurately captured the complex thermal behavior resulting from the incorporation of microencapsulated paraffins with a solid–liquid phase transition in the 40–50°C range. The temperature-dependent thermophysical properties of all the constituent materials were incorporated, including the large latent heat absorption that occurred during PCM melting.

The accuracy of the model was demonstrated by its good qualitative and especially quantitative agreement with the transient temperature measurements during heating tests on cylindrical foam samples. The simulations reliably replicated the experimental temperature profiles at varying depths in the foam both qualitatively and quantitatively. Specifically, the model accurately captured the temperature plateau induced by the PCM latent heat, which enhanced the thermal regulation in the multifunctional composites and the final temperature plateau value.

After validation, the model was applied to simulate the thermal conduction through a foam wall subjected to a sinusoidally oscillating heat source between 12 and 72°C. The results highlighted the insulating effect of the presence of PCM, which maintained surface temperatures up to 5°C lower than those of plain epoxy samples. This showed the capability of these foams to act as a thermal reservoir, as well as the model's ability to predict performance improvements from PCM incorporation without extensive physical testing.

In summary, the modeling methodology provided an efficient tool for optimizing the design of HGM/PCM syntactic foams to balance density reduction, thermal

conductivity, and heat storage capacity. It can aid in developing tailored foam compositions for lightweight engineering systems that require load-bearing capacity and passive temperature control, such as batteries, electronics, building materials, and aerospace components. The simulation approach enables rapid virtual prototyping to evaluate the implications of various microstructural parameters on overall thermal management performance. Future work will aim to model the relationship between the thermal management performance of these samples and their load-bearing capabilities to optimize their multifunctional behavior. Moreover, additional simulations will be performed to analyze the material behavior with a variable heat source of different periods (e.g., hours, seconds) and shapes (e.g., square waves).

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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