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Scaling up hybrid insulation: Integration of lignocellulose and phase change materials for sustainable thermal management

Xiang Hu [a,b], Ari Kankkunen [a], Ari Seppälä [a], Maryam R. Yazdani McCord [a,*]

[a] Department of Mechanical Engineering, Aalto University, Finland
[b] Industrial Engineering and Management, Department of Mechanical and Materials Engineering, University of Turku, Finland

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ABSTRACT

This research addresses the need for eco-friendly, thermally protective packaging materials. A scalable process was developed that minimizes greenhouse gas emissions and produces hybrid materials with improved thermal insulation, energy storage, mechanical resilience, and water resistance. By using lignocellulose as a porous carrier and polyethylene glycol (PEG) as a phase change material (PCM), convective drying proved more effective for large-scale production than freeze-drying. The resulting materials are flexible, lightweight (0.03–0.04 g/cm^3), and hydrophobic. They exhibit suitable thermal properties with latent heat capacities within 110–123 J/g and thermal conductivities within 0.037–0.042 W/mK. These hybrids are leak-free during phase transitions with tunable melting points, confirming their practicality. Life Cycle Assessment (LCA) shows that this method uses less energy and produces fewer carbon emissions than freeze-drying. Thus, convective drying is a promising scaling-up method for producing effective, eco-friendly temperature-responsive insulation materials for various applications requiring temperature control.

1. Introduction

Packaging materials play an important role in the daily lives, helping to protect products and facilitate their transportation. Among all materials used for packaging, insulating or protective layers in packaging are crucial because they safeguard the contents from potential damage caused by external factors such as moisture, temperature fluctuations, physical impact, and environmental conditions during storage and transportation, ensuring the integrity and quality of the product upon arrival to the end-users [1]. While packaging with protective layers is commonly used these days, several pressing problems persist. These include complications in recycling due to multilayer packaging and issues related to the design and reuse of plastics [1,2]. Additionally, the thermal insulation properties of current packaging materials are often inadequate, limiting their effectiveness for storage at varying temperatures. Moreover, concerns about the environmental impact of certain packaging materials, such as plastic and Styrofoam, have grown, prompting efforts to reduce their use and find more sustainable alternatives [3]. To mitigate associated shortages and address environmental concerns, industries are looking for new packaging solutions and are willing to adopt practices that promote recycling and environmentally friendly materials.

When considering sustainable alternatives for packaging, cellulose may be first on the list. Cellulose based foams are used in packaging, a type of foam insulation made from recycled paper, typically newspaper, cardboard, or other cellulosic materials [4]. The cellulose fibers are mixed with a foaming agent and water, and then blown into walls, attics or other areas of a building using special equipment [5]. The mixture expands and hardens, creating a layer of insulation that helps prevent heat loss and reduce energy consumption. Cellulose-based foam insulation is considered an environmentally friendly option because it is made from plants or recycled materials, has a low environmental impact with a biodegradable nature [6–8] and is also known for its sound insulation properties [9,10].

In addition to the utilization of cellulose, phase change materials (PCMs) can be considered in the design of sustainable packaging materials [11]. PCMs show the capacity to absorb and release latent heat through their melting and crystallization processes [12,13]. They can effectively store significant quantities of thermal energy during phase transitions at specific temperature ranges [12,14], making them a valuable asset for applications in temperature regulation and insulation [15,16]. As PCMs are excellent materials for thermal regulation and heat

* Corresponding author.
E-mail address: roza.yazdani@aalto.fi (M.R. Yazdani McCord).

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storage, they can be utilized in construction [17], refrigeration [18], packaging [6], electronics [19–21], and solar energy storage [22,23]. In packaging applications, PCMs are used to protect temperature-sensitive products during storage and transport, particularly in industries such as pharmaceuticals, food, and electronics [24–26]. However, the fluidity of PCMs in their melt state leads to leakage issues in practical applications [15,27,28]. Therefore, using PCMs necessitates a confining or coupling strategy to prevent leakage when they are in the melt state. In this context, the confinement of PCMs can be accomplished with various materials, ranging from minerals to polymers [13, 24–29].

Building on the exploration of PCMs in packaging design, another critical aspect in the pursuit of sustainable solutions lies in the field of thermal insulation materials. As the significance of minimizing greenhouse gas emissions gains importance, the environmental implications of novel products become increasingly crucial. Thermal insulation plays a vital role across diverse sectors. In buildings and construction, it enhances energy efficiency and maintains temperature consistency [30]. It is also valued in the packaging sector for thermal protection [31]. However, the conventional manufacturing processes associated with traditional insulation materials contribute significantly to carbon emissions [19]. Recognizing the crucial to address these environmental concerns, there is a pressing need for innovative solutions that not only reduce carbon emissions but also enhance energy efficiency. Such advancements can have a profound influence on the economic and environmental conditions through substantial energy savings. In this context, this research carries out a Life Cycle Assessment (LCA) study, aiming to assess the environmental impact throughout the cradle-to-grave phase of newly developed cellulose insulation materials integrating PCM.

The research employs polyethylene glycol (PEG) as a PCM and utilizes lignocellulose as a leakage-preventive porous carrier for foaming. The lignocellulose-PCM hybrid foam reported here is engineered to possess thermal insulation and energy storage properties while adhering to sustainability principles. PEG possesses advantageous intrinsic properties including biocompatibility, non-toxicity, high latent heat, and an adjustable fusion temperature (by varying molecular mass). For example, PEG with a molecular mass in the 600–8000 g/mol range can achieve a tunable fusion temperature within 20–65 °C. Cellulose has been used for shape stabilization of PCMs, with most research utilizing cellulose in nanofiber form [15, 18, 32–35]. However, cellulose nanofibers are more expensive, and demand extra processing and energy compared to the kraft cellulose fibers (pulp) used in this research. Cellulose fibers serve multiple critical functions in this hybrid foam. Although, primarily, they act as an insulating material, their role extends beyond insulation in this hybrid foam. They also provide structural support and shape stabilization for the PEG. This stabilization is crucial, as PCMs are known for their latent heat storage properties but can suffer from fluidity and leakage issues when used independently. The cellulose fibers help to immobilize the PCM, preventing leakage and ensuring the material retains its shape during phase transitions. As a derivative of cellulose polymer, methylcellulose is used as a binder and foaming agent in the composite. Its primary function is to facilitate the creation of air bubbles in the solution as well as enhancing the bonding between the cellulose fibers after drying, thereby increasing the mechanical strength and durability of the final product. This binding effect contributes to the overall stability and integrity of the composite. The addition of lignin is explored for enhancing the product's hydrophobicity, a critical attribute for its application in packaging, which is new for its use with PCMs in this specific context. The study also investigates the upscaling process, a crucial aspect of the research, exploring various drying methods, namely freeze drying and convective drying, to understand their influence on final products and energy consumption during production. By addressing the challenges of upscaling bio-based and porous composites, the study paves the way for the practical implementation of these materials in commercial packaging. Remarkably, there is a notable shortage of upscaling demonstrations for bio-based and porous composites in the existing literature. This can originate from the limitation associated with freeze drying method commonly used for the preparation of these materials. Finally, the research conducts a LCA to evaluate the environmental impact from cradle to grave, comparing carbon emissions between samples subjected to freeze drying and convective drying. The existing literature notably lacks such comparative analysis, underscoring a noticeable research gap. The LCA attempts to address this limitation by providing insights into sustainability considerations and filling this crucial research void.

The findings not only highlight the potential of the developed product as an excellent addition to the market of packaging materials with heat prevention properties but also introduce a suitable scaling production method aligned with the contemporary aim of reducing carbon footprint. Key product properties under consideration encompass density (aiming for relatively low density), hydrophobicity (indicated by a high contact angle), and various thermal properties, including leakage resistance, thermal protection, thermal conductivity, as well as latent heat capacities of the PCM foams.

2. Materials and methods

2.1. Materials and instruments

PEG (Mn = 4000, melting at 48–55 °C) was selected as the model PCM and was purchased from Sigma-Aldrich, Germany. Methylcellulose was purchased from TCI, Japan. Lignin was supplied by UPM BioPiva, Finland. The cellulose fibers (pulp) used in the study were sourced from UPM, Finland. These fibers were derived from birch wood and were provided in an undried state, containing 15 % solid content.

2.2. Production and scale-up process

2.2.1. Hybrid mixture formation

After dissolving a specific amount of PEG in distilled water, lignocellulosic base materials were mixed in the solution one by one. The temperature of the solution was then increased to 80 °C under continuous mixing. A homogenizer (Package-Set HG - 15A-Set - HT1018) with a blender was used for the stirring process and the stirring speed reached up to 2000 rpm/min. The suspension was mixed for 20 min until a clear viscose solution was obtained. Table 1 shows the composition of the samples. PMC sample contains PEG, methylcellulose, and cellulose fibers, while PMLC sample contains one more component, lignin.

2.2.2. Drying

For freeze drying method, the mixture was subjected to lyophilization under vacuum conditions using a Freeze Dryer (Christ Alpha 2–4), allowing the foams to dry over a period of 48 hours. Freeze drying was limited to small sample sizes due to equipment constraints. Additional information on this is provided in the Supplementary Information. For convective drying, the foamed mixture was discharged onto a plate and placed under an IR lamp (OPAL, 2000 W) with the temperature of 80 °C for 1 hour. It was then left to dry under room temperature for 24 hours. Further illustrations of convective drying are given in Figure S1.

2.2.3. Upscaling

For upscaling process, depicted in Fig. 1, the mixing and drying process was the same in the convective drying process. The foaming

Table 1
The composition of foaming mixture (mass-%). PMC sample contains PEG, methylcellulose, and cellulose fiber, while PMLC sample also contains lignin.

Sample code (%)	PEG (%)	Methylcellulose (%)	Lignin (%)	Cellulose fiber (%)	DI water (%)
PMC	6	1.50	0	1.50	91
PMLC	6	1.25	0.25	1.50	91

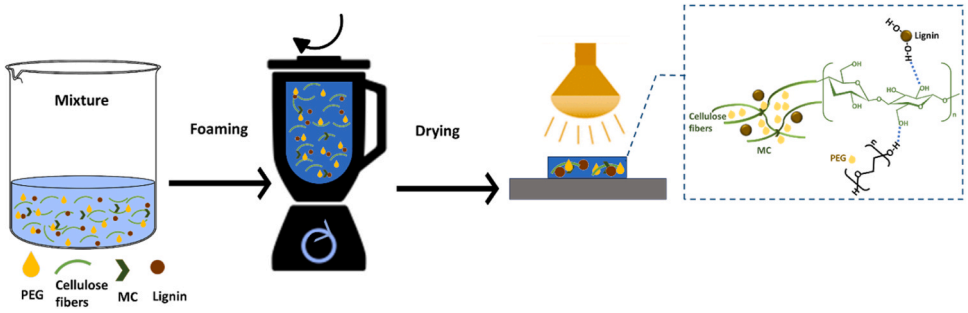

Fig. 1. A Schematic illustration of the scaling up process including mixture preparation, foam forming in a blender and convective drying using an IR lamp together with schematic of the material components and hydrogen bonding between them.

process was as follows: after all components fully mixed in water, the solution was stirred with a blender (SENZ SESB1000 SPORTTI-BLENDERI) for foaming for a few minutes until a homogenous foamed mixture was obtained. The mixture was then discharged on a plate and placed under the IR lamp for drying.

2.3. Morphological characterization

2.3.1. Density measurement

To measure the density of the hybrid foams, the samples were wrapped in a waterproof thin film and suspended in water using a string. The displaced water volume, equivalent to the sample's volume, was measured. The density was then calculated by dividing the sample mass, obtained through standard weighing, by the measured volume.

2.3.2. PCM leakage test

To assess PCM leakage, the hybrid foam samples were placed on clean white paper and heated in an oven at 70 °C, which is above the melting point of the PCM. The samples were exposed to this temperature for 2 hours to observe any potential leakage during the melting process. Photographs were taken before and after the test to document the leakage-free characteristics of the foams.

2.3.3. Contact angle

The water resistance behavior of the hybrid foams was evaluated by measuring water contact angles using a Theta Flex optical tensiometer. A water droplet of approximately 5 μL was deposited on the surface of each sample. The contact angles were recorded immediately upon deposition and again after 400 seconds to assess changes over time.

2.3.4. Scanning electron microscopy

The microstructure of the hybrid foams was examined using scanning electron microscopy (SEM) with a Zeiss Sigma VP instrument operating at 2 kV under vacuum conditions. Prior to SEM analysis, the samples were sputter-coated with a 4 nm layer of gold using a Leica EM ACE600 sputter coater. The samples were mounted on metal stubs using carbon tape to ensure stability during examination.

2.4. Thermal characterization

2.4.1. Differential scanning calorimetry

Phase change enthalpy and temperature values were measured by DSC (NETZSH DSC204F1 Phoenix). A DSC program was conducted from −20–80 °C on samples placed in aluminum crucibles, ~5 mg each. Starting from room temperature, a heating step of 5 K per min was applied for four cycles.

2.4.2. Thermal conductivity

A TCi thermal conductivity analyzer (C-Therm) was employed to determine the thermal conductivity of sample foams according to a modified transient plane source method. The samples were 29 mm in diameter and 60 mm in thickness. Thermal conductivity was tested for 7 times from the same positions.

2.4.3. Thermal insulation performance

Insulation and physical properties including structural stability, deformation and phase changes were examined under light irradiation by a thermal camera (FLIR SC7600) and imaged using Research IR Max. Hollow cubic boxes, each side 60 mm with wall thickness of 6 mm, where prepared from PMC (17 g) and PMLC (19 g). Cubes were located under 2 lamps for irradiation process to further evaluate the thermal insulation performance. Two symmetrically located 2000 W halogen lamps were heating the cube at the angle of 45 ° at the distance of 37 cm. The tests were performed at room temperature around 20 ℃. The temperature inside the cubes was recorded by a thermocouple located in the center of the hollow cube. The irradiation heating process was 20 min, followed by the cooling process (off irradiation) for 20 min for each sample.

2.5. Life cycle inventory (LCI) for life cycle assessment (LCA)

Data gathering is an essential part of conducting LCA on the products. Thus, a life cycle inventory is listed as the important step to gather the necessary data for evaluation. The only impact category assessed is climate change, as this corresponds to the objective of this research. The target is to evaluate the environmental impact of the whole life cycle of the developed insulation materials. Climate change is an impact category that is most effectively evaluated through a midpoint assessment. This approach involves measuring the impact using an indicator value at an intermediate stage of the impact process, from which additional impacts can be inferred. The results of this impact assessment are expressed in kg CO_2-eq and refer to a functional unit.

The aim of the LCA was to assess the environmental impact of the entire life cycle of the PMC and PMLC foams, from cradle to grave. In total, three types of materials were assessed: two hybrid foams were compared with traditional PU foam in terms of reducing the impact of climate change. The calculations were performed in OpenLCA using the IPCC 2013 GWP 100a methodology and a causal attribution criterion. The time horizon of the analysis was 2023–2083, with a service life of 60 years for the insulation materials, which corresponds to the use phase of the products. The geographical area considered is the city of Espoo (Finland). The approach used is consequential, so the results represent the change in emissions caused using each of the insulation materials. Additional information on LCA has been provided in the Supplementary Information. Figure S2 depicts the system boundary of the life cycle. All materials have the same volume of 0.04 m^2. The relationship between density, volume and mass is shown in Table S1.

3. Results and discussion

3.1. Comparison of drying methods: Convective drying vs Freeze drying

Samples dried by freeze drying method are shown in Fig. 2a. All samples retain their shape after lyophilisation with freeze drying. There is no deformation or phase segregation of PCM during the drying process, indicating that the PCM is fully embedded in the porous supports. Meanwhile, the introduction of lignin enhances the structural robustness of the hybrids, which can be felt when manually applying pressure to the sample. The compression test, shown in Figure S3, revealed that PMLC has better stability and compressive stress, which indicates the addition of lignin enhanced mechanical resilience of the hybrid foam.

The sample images from the convective drying method are shown in Fig. 2b. All samples have successfully formed into foams. This is primarily attributed to the inclusion of cellulose fibers, which serves to cross-link the material matrix and facilitates the formation of a foam scaffold. Among these samples, the composition consisting of PEG, methylcellulose, lignin, and cellulose fibers exhibits the highest stability. This composition is easy to mold and relatively easy to remove from the mold, especially with the addition of lignin. This may be due to the hydrophobicity of lignin, which improved the release of the samples from the mold [30].

Both freeze drying and convective drying methodologies result in fully dried samples. However, the convective drying yields samples with superior shape retention and resiliency compared to those processed through freeze drying, while freeze-dried samples exhibit an undesirable crispiness. Freeze-drying removes water from a frozen product through sublimation, resulting in a porous structure that gives the product a crispy texture [36]. Therefore, convective drying not only optimizes the properties of the material but also enhances its suitability for potential integration into advanced packaging materials.

3.2. Morphological properties

The densities of the samples are tabulated in Table 2 and are less than 0.04 g/cm^3. All samples show a relatively low density, which is lower than the density of commercial PU foam 0.1 g/cm^3, and other commercial packaging foams, from 0.2 g/cm^3 to 0.42 g/cm^3 [4,37]. The advantageous lightweight characteristic of these hybrid foams proves particularly valuable in packaging. This attribute not only facilitates easy handling, installation, and transportation but also contributes to reduced CO_2 emissions during transit, underscoring their environmental efficiency [18].

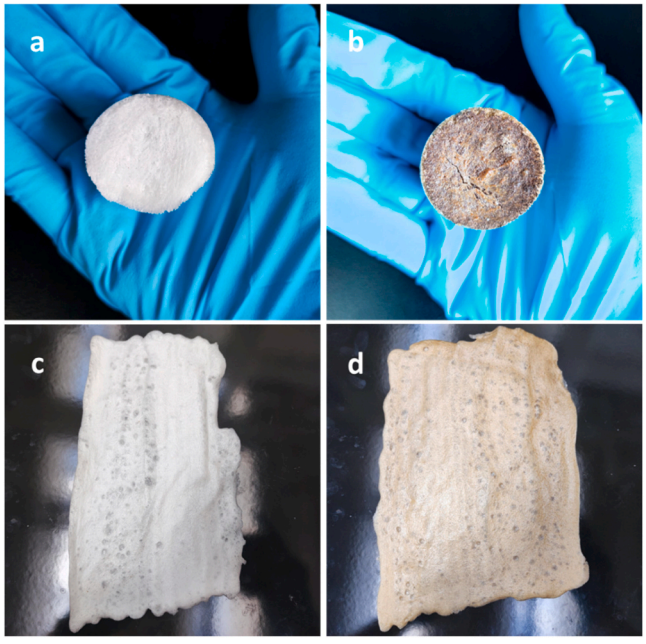

Fig. 2. a) PMC b) PMLC Freeze-dried samples with different compositions in 2.5 cm caliber and 0.8 cm thickness sample size. c) PMC and d) PMLC samples resulted from the convective drying method in upscaling process, with the length of 13.5 cm, width of 18.2 cm and the thickness of 0.15 cm.

Table 2
Density and thermal conductivity of the samples. FD is for freeze drying and CD is for convective drying.

Sample	Density (g/cm^3)	Thermal conductivity (W/mK)
PMC-FD	0.040	0.033
PMC-CD	0.034	0.042
PMLC-FD	0.035	0.037
PMLC-CD	0.037	0.038

SEM is used to study the microstructure of the samples and verify the degree of pore alignment in all samples, which can be seen in Fig. 3. The SEM images reveal that the samples exhibit a porous structure, combining both open and closed cell properties. No sign of phase segregation and inhomogeneity was observed by SEM which means successful foaming and the integration of PEG in the porous structure.

A leakage resistant behavior was observed for the foam samples upon heat exposure by placing the samples in an oven at 70 ℃ for 2 hours. Figure S4 shows the images from the leakage tests. This illustrates the effective retention of PCM melt by the porous media, showcasing a successful absence of seepage or loss. The absence of leakage was also confirmed by thermal camera images under radiation heating discussed in later sections. Consequently, when these hybrids serve as packaging materials to ensure temperature stability, the porous carriers prove proficient at securely storing the PCM. In practical scenarios, even when subjected to temperatures over 50°C or 60°C, the foams exhibit sustained stability, free from any PCM leakage.

The contact angle tests were conducted to assess the hydrophobicity of the hybrid foams, specifically examined on the convective drying samples (as depicted in Fig. 4). This method allows for a an understanding of how water interacts with the hybrids, providing valuable insights into their hydrophobic properties. However, this test could not be measured for the freeze-drying samples due to their pronounced hydrophilicity. As water drop contacted the surface of freeze-dried samples, it was soon immersed into the sample. This can be due to the larger-sized pore foamed during hydrophilization process [38]. Thus, only convective drying samples were measured for contact angle test. For the samples dried with convective drying method, the initial contact angle is more than 90°. While foam mixed with lignin shows better hydrophobicity with the average contact angle more than 88° in 400 s. The addition of lignin increased the hydrophobicity, which indicates its ability to prevent moisture absorbance in real use. It has been reported that the use of lignin enhances hydrophobicity in biopolymer [39] and rubber-based composites [40].

3.3. Thermal properties

DSC thermograms for PMC and PMLC samples are depicted in Fig. 5. The DSC results are compiled in Table 3, which show the measured melting temperatures and enthalpies. The melting heat for the samples from convective drying was between 113 J/g and 123 J/g and the crystallization heat was between −121 J/g to −110 J/g. Even though, the enthalpy values for the samples from freeze drying appear slightly higher which can be due to lyophilization, these samples were rigid and brittle which is not ideal for the target application. While PEG is successfully embedded in the porous structure, the substrate of this porous carrier does not significantly affect the latent heat of the samples. Moreover, there is no significant effect of lignin addition on the latent heat of the samples, which means that lignin does not disturb the phase

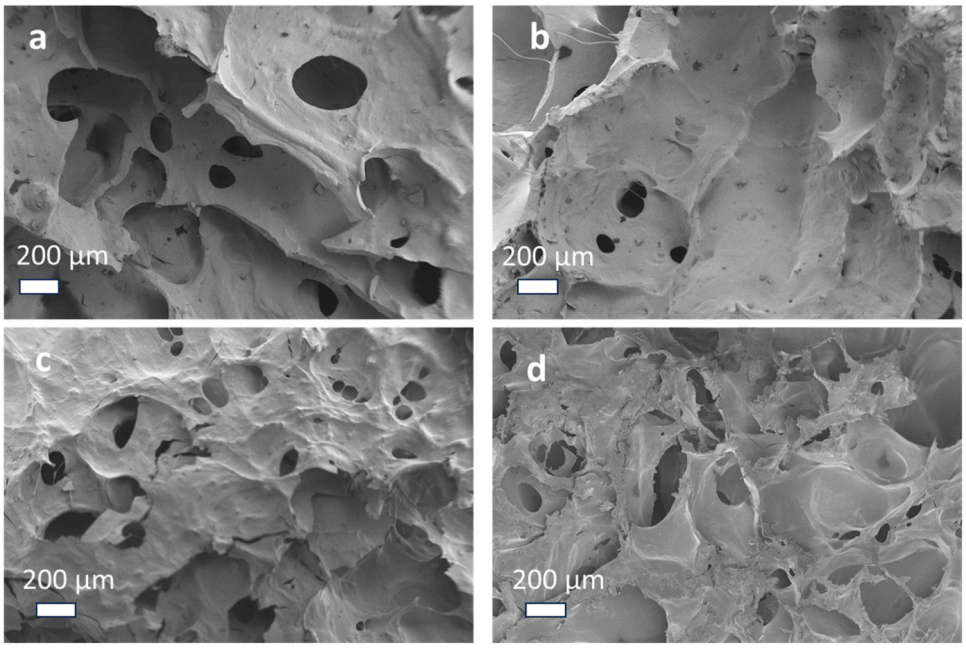

Fig. 3. SEM images of a) PMC and b) PMLC freeze-dried; c) PMC and d) PMLC convective-dried samples.

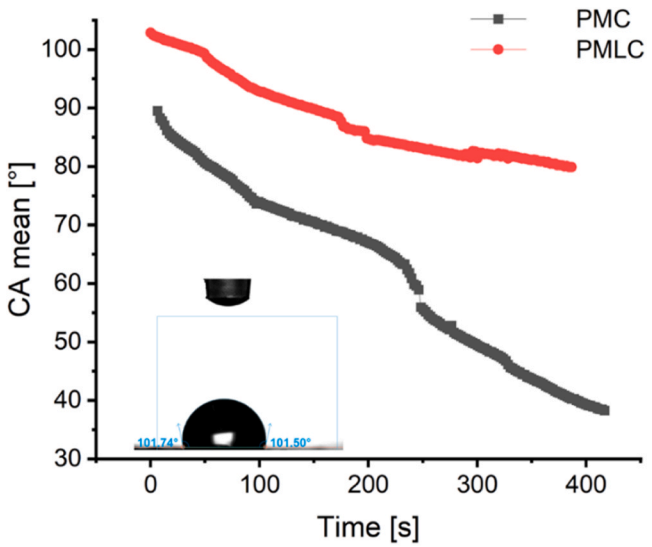

Fig. 4. Contact angle (CA) data of two hybrid samples from convective drying method. This test could not be measured for the freeze-dried samples due to their pronounced hydrophilicity.

transformation of the PCM in the matrix. The two drying methods resulted in no major changes in the phase change properties, indicating the suitability of production method for producing stable porous structures with embedded PCM.

The average thermal conductivity and standard deviation of PMC sample are 0.043 ± 0.008 W/mK and for PMLC sample are 0.038 ± 0.005 W/mK. The previously reported thermal conductivity of a similar phase change hybrid was 0.040 W/mK [18], which agrees with those of the foams developed in this study. The thermal conductivity for PMLC samples containing lignin is lower compared to that of PMC samples. The possible reason could be the synergistic effects of lignin addition to cellulose fibers and methylcellulose system [41]. The hydrophobic effect of lignin can cause a well-preserved porous structure and better pore distribution in these samples which lead to lower thermal conductivity. Lower thermal conductivity is generally beneficial for packaging as it enhances insulation, maintaining stable temperatures, crucial for product quality and safety, while the hydrophobic nature of these samples can preserve the material from moisture absorbance. Therefore, PCM foams containing lignin show a promising prospect for packaging application.

3.4. Thermal insulation performance

Fig. 6 shows the scaled-up samples prepared in a box form for the thermal insulation performance test under irradiation heating. The measurement was also recorded by a FLIR thermal camera and images are presented in Fig. 7. Figures S5 and S6 in the Supplementary Information present more thermal images. The results of thermocouple are shown in the plot in Fig. 6. The temperature inside the hollow boxes increased continuously to a maximum temperature after 20 minutes irradiation, i.e., 92°C in the sample PMC and around 110°C in the sample PMLC. The maximum temperature is different between two samples. Since both samples contain almost the same mass of PCM, the possible influence is from the structural composition. As can be seen, the sample containing lignin has a darker brown color which can lead to higher emissivity, absorbance, and conversion of light to heat resulting in higher temperature. In addition to thermal conductivities, different emissivity of samples may affect to the thermal performance. However, the actual mechanism is not yet clear and further investigation is required. In the cooling step, both samples PMC and PMLC showed a gradual decrease near the temperature at which phase change occurs, suggesting the crystallization of PEG. Upon the removal of the irradiation, the samples held a temperature for a long period due to the energy released upon crystallization. This study indicates that the PCM foam shows appropriate fluidity retention, insulation, and energy capture properties useful for packaging application. Similar results were observed for previously reported PCM bio composites [18]. As can be seen in Fig. 7, a noticeable surface temperature is achieved during irradiation and yet there is no sign of surface leakage or decomposition which is another indication of the stability of the developed hybrid foams.

3.5. Life cycle assessment

Fig. 8a shows the comparison of results in kg CO_2 eq from cradle-to-

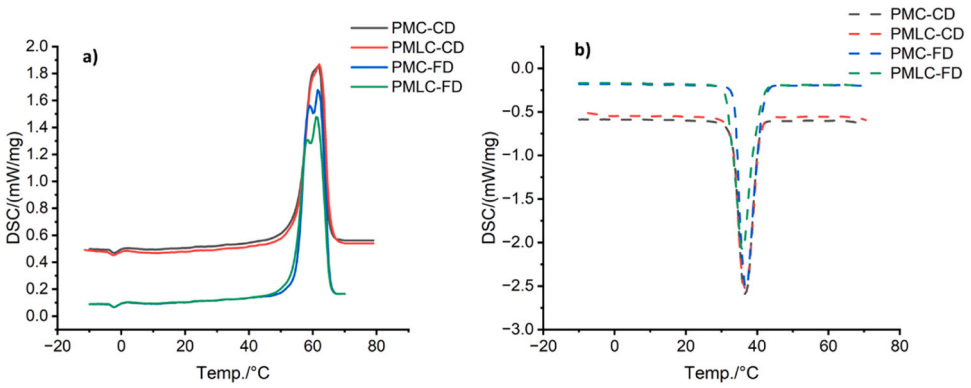

Fig. 5. DSC thermograms for PMC and PMLC samples from freeze drying (FD) and convective drying (CD) in a) heating and b) cooling process.

Table 3
DSC results of the samples from freeze drying (FD) and convective drying (CD) (T_m – Melting temperature, ΔH_m – Melting heat, T_c – Crystallization temperature, ΔH_c – Crystallization temperature).

Sample	T_m (°C)	ΔH_m (J/g)	T_c (°C)	ΔH_c (J/g)
PMC-FD	55	135±1.5	42	−133±1.0
PMLC-FD	54	129±3.5	42	−127±4.3
PMC-FD	54	113±0.7	41	−110±0.6
PMLC-CD	55	118±5.4	41	−117±4.0

gate phase between PU foam and PMC samples from freeze drying and convective drying. The PMC and PMLC have the similar production process. Thus, in this research, the production of PMC is mainly researched on the carbon emission during the process. The detailed information can be found in the Supplementary Information. Fig. 8b shows the comparison results in kg CO_2 eq between the PMC and PU foam from cradle-to-grave phase. Fig. 8a contains the carbon emission from the raw materials and production phases, which indicates the carbon emission caused by the production. While Fig. 8b contains two more phases, use and end-of-life, giving an overlook of carbon emissions from the product's whole life cycle.

From cradle to gate, all hybrid foams show positive carbon emissions. The PMC sample with convective drying method shows the lowest carbon emissions compared to all other foams including PU. This indicate that PMC foam is a more energy efficient foam compared to PU foam, which is an improvement. When considering the cradle to grave phase, all foams are generally carbon negative, while the hybrid foams perform better than PU foam emission-wise. This is due to the carbon reduction from the use phase. Thermal insulation and the energy absorbance performance of the material in real use will help to reduce the energy used to maintain the temperature.

In terms of end-of-life management, there are three commonly used strategies for thermal insulation materials: landfill, recycling, and incineration. Landfilling is a commonly used method but raises concerns due to the long-term environmental impact of non-biodegradable insulation materials. Recycling is a promising approach for certain types of insulation, while incineration can generate energy but requires careful management to minimize harmful emissions. Considering the different materials in this case study, all are assumed to have the same environmental impact during the end-of-life phase due to the challenges of estimating CO_2 emissions or absorption [42]. However, it is important to emphasize that, unlike the product reported in this research, commercial insulations such as PU present challenges related to biodegradation and environmental aspects. End-of-life management for these products requires further comprehensive studies which will be reported in our future works.

From the results shown, the freeze-drying method introduces more carbon emission during the production process, which has a significant impact on the total carbon emission results. Freeze drying methods have been used in biopharmaceutical and food industries, the high energy consumption and carbon emission issues have been discussed widely, which should not be ignored [43,44]. Meanwhile, the convective drying method proves to be a promising method to reduce carbon emission

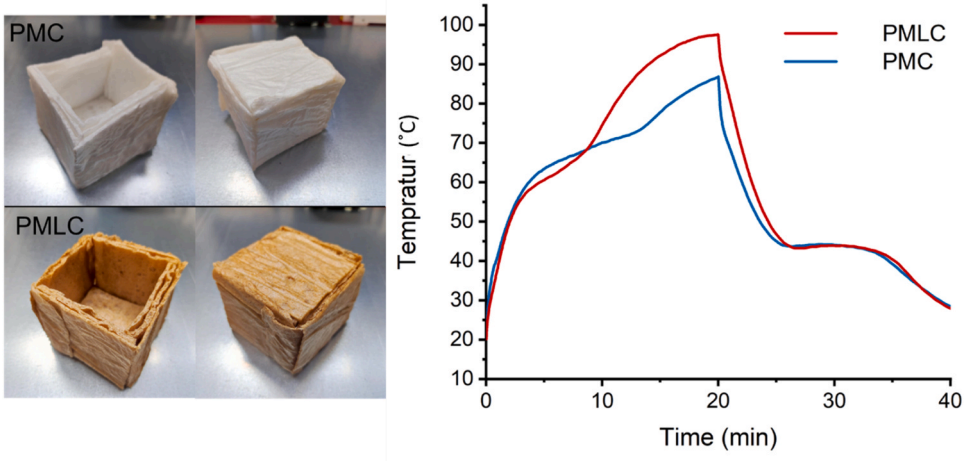

Fig. 6. The images of sample boxes made from the convective drying samples in the upscaling process, with the size of 60*60*60 mm and the wall thickness of 50 mm. The test samples are designed with three layers to enhance the stability under heat lamp. The right plot shows the measured temperature evolution inside the hollow cubes insulated with PMC and PMLC samples exposed to heating radiation.

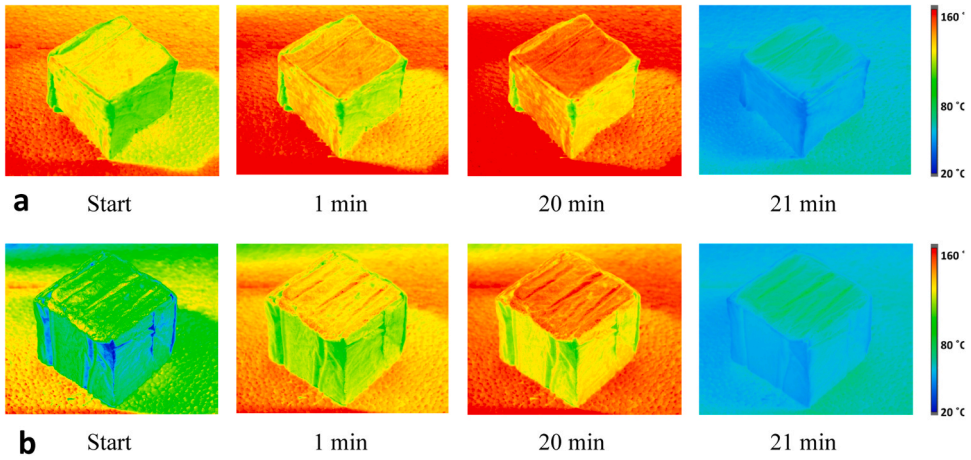

Fig. 7. Thermal behavior of sample boxes under irradiation captured by IR camera: a) PMC and b) PMLC samples.

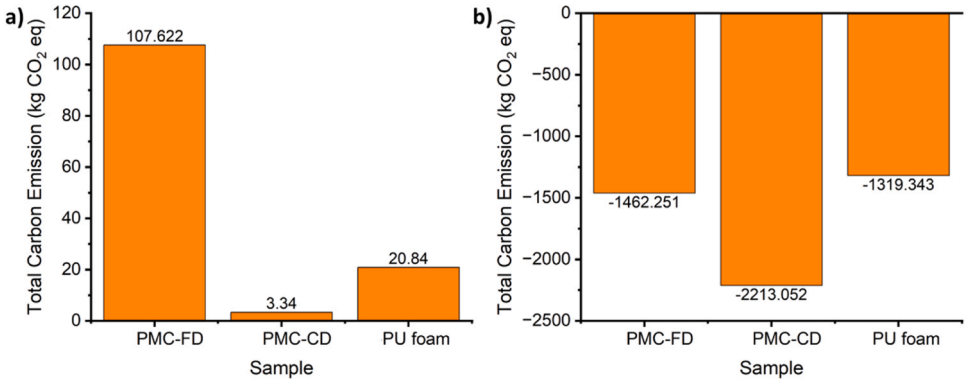

Fig. 8. a) Total carbon emission from cradle-to-gate phase and b) Total carbon emission from cradle-to-grave phase.

during the production process. For these thermal insulation materials, the total life cycle is found to have a negative carbon emission, which is mainly due to the negative carbon emission during the use phase. However, further LCA research are necessary due to the limitations and assumptions made during this research.

A key limitation of the LCA method was the lack of databases for certain chemicals and processes in the OpenLCA. Despite this, the method effectively explained the manufacturing and use stages of the materials. Variations in LCA results can stem from an incomplete list of materials or processes in various databases. While the method elucidated the manufacturing and use stages, it has several shortcomings and requires improvement for better application in this sector. Payback time is also an important aspect that needs to be investigated further. Different PCMs in different applications can have varying environmental and economic payback times [45]. Heidari et al. [46] analyzed the environmental impacts of wood based PCM panels, finding that PCM and heating energy had the highest environmental impacts, with energy consumption identified as a hotspot. They concluded that developing comprehensive databases, including sector-specific inventories for bio-based materials like chemical components and forestry operations, is essential for future LCAs. They also emphasized the need to investigate uncertainty in LCA methods and develop a tool suitable for projects with time and cost constraints. Serrano et al. [47] studied the enhancement of stabilized rammed earth walls with microencapsulated PCM. Their study included thermal analysis of different material compositions and their environmental impact assessed by LCA. Their findings indicated improved thermal properties, but the LCA impact points significantly increased due to the use of microencapsulated PCM. They suggested that using macro-encapsulated PCM and carefully selecting the PCM could improve future LCA results.

4. Conclusions

The up-scaling process to produce thermally insulating hybrid foams proved to be highly efficient, with lower greenhouse gas emissions, good thermal insulation, energy storage capacity and hydrophobicity of the products. In the material preparation part, PEG is used as a PCM, methylcellulose and cellulose fibers are used as carrier materials with porous structure. Lignin is then introduced to enhance the stability and hydrophobicity of the hybrid foams. Different drying methods, namely freeze drying and convective drying, are investigated to select the best option for upscaling. Considering factors of shaping, thermal properties, energy consumption and sustainability, the convective drying is found to be a promising method for production.

The shape of all hybrid foams from the convective drying method is maintained with relatively good flexibility in real use. The density of all samples was between 0.03 and 0.04 g/cm^3, which proves the relatively low-density hybrid foams. The SEM showed the porous structure of the samples, with open and closed cell structures. The contact angle test demonstrated the hydrophobicity, indicating potential use in packaging applications.

The melting point varied between 53 °C and 56 °C, while the latent heat ranged between 110 J/g and 123 J/g. This proves the promising application in the field of energy storage and thermal regulation for all the prepared PCM hybrid foams. As for thermal conductivity, it varied from 0.037 W/mK to 0.042 W/mK. This indicates potential applications from a thermal insulation perspective. There was no sign of PCM leakage for the hybrid samples under heat exposure, indicating potential real-

world applications for such foam substrate with embedded PCMs. The thermal insulation performance test indicated excellent thermal protection property under irradiation and heating.

The LCA results proved that there is a considerable energy consumption in the freeze-drying process, which is not suitable for scaling up considering the energy saving factor. While for convective drying of the samples, there was less carbon emission in the production phase. Based on the property and LCA research, the convective drying method proves promising for upscaling production with positive results in physical shape, thermal properties, and carbon emission reduction. The potential use of the products in packaging is not only based on the thermal insulation and energy storage property, but also on the hydrophobicity and resilient shape of the formed materials.

CRediT authorship contribution statement

Xiang Hu: Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data Acquisition. **Ari Kankkunen:** Writing – review & editing, Validation, Methodology, Investigation, Formal analysis. **Ari Seppälä:** Writing – review & editing, Methodology, Investigation, Formal analysis. **Maryam R. Yazdani McCord:** Writing – original draft, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.mtcomm.2024.110281.

References

[1] I.D. Ibrahim, Y. Hamam, E.R. Sadiku, J.M. Ndambuki, W.K. Kupolati, T. Jamiru, A. A. Eze, J. Snyman, Need for sustainable packaging: an overview, Polymers 14 (20) (2022) 4430, https://doi.org/10.3390/polym14204430.

[2] H. Cheng, H. Xu, D. Julian McClements, L. Chen, A. Jiao, Y. Tian, M. Miao, Z. Jin, Recent advances in intelligent food packaging materials: principles, preparation and applications, Food Chem. 375 (2022) 131738, https://doi.org/10.1016/j.foodchem.2021.131738.

[3] L.K. Ncube, A.U. Ude, E.N. Ogunmuyiwa, R. Zulkifli, I.N. Beas, Environmental impact of food packaging materials: a review of contemporary development from conventional plastics to polylactic acid based materials, Materials 13 (21) (2020) 4994, https://doi.org/10.3390/ma13214994.

[4] P. Nechita, S.M. Nastac, Overview on foam forming cellulose materials for cushioning packaging applications, Polymers 14 (10) (2022) 1963, https://doi.org/10.3390/polym14101963.

[5] H. Liu, H. Du, T. Zheng, K. Liu, X. Ji, T. Xu, X. Zhang, C. Si, Cellulose based composite foams and aerogels for advanced energy storage devices, Chem. Eng. J. 426 (2021) 130817, https://doi.org/10.1016/j.cej.2021.130817.

[6] B. Regubalan, P. Pandit, S. Maiti, G.T. Nadathur, A. Mallick, Potential Bio-Based Edible Films, Foams, and Hydrogels for Food Packaging, in: S. Ahmed (Ed.), In Bio-based Materials for Food Packaging: Green and Sustainable Advanced Packaging Materials, Springer, Singapore, Singapore, 2018, pp. 105–123.

[7] I.Y. Miranda-Valdez, S. Coffeng, Y. Zhou, L. Viitanen, X. Hu, L. Jannuzzi, A. Puisto, M.A. Kostiainen, T. Mäkinen, J. Koivisto, M. Alava, Foam-formed biocomposites based on cellulose products and lignin, Cellulose 30 (4) (2023) 2253–2266, https://doi.org/10.1007/s10570-022-05041-3.

[8] M. Reichler, S. Rabensteiner, L. Törnblom, S. Coffeng, L. Viitanen, L. Jannuzzi, T. Mäkinen, J.R. Mac Intyre, J. Koivisto, A. Puisto, M. Alava, Scalable method for bio-based solid foams that mimic wood, Sci. Rep. 11 (1) (2021) 24306, https://doi.org/10.1038/s41598-021-03764-0.

[9] J. Cucharero, S. Ceccherini, T. Maloney, T. Lokki, T. Hänninen, Sound absorption properties of wood-based pulp fibre foams, Cellulose 28 (7) (2021) 4267–4279, https://doi.org/10.1007/s10570-021-03774-1.

[10] V.K.R. Kodur, P.P. Bhatt, M.Z. Naser, High temperature properties of fiber reinforced polymers and fire insulation for fire resistance modeling of strengthened concrete structures, Compos. Part B: Eng. 175 (2019) 107104, https://doi.org/10.1016/j.compositesb.2019.107104.

[11] H. Baniasadi, J. Seppälä, A. Kankkunen, A. Seppälä, M.R. Yazdani, Water-resistant gum-based phase change composite for thermo-regulating insulation packaging, J. Energy Storage 61 (2023) 106725, https://doi.org/10.1016/j.est.2023.106725.

[12] S. Raoux, Phase Change Materials, Annu. Rev. Mater. Res. 39 (1) (2009) 25–48, https://doi.org/10.1146/annurev-matsci-082908-145405.

[13] M.R. Yazdani McCord, H. Baniasadi, Advancements in form-stabilized phase change materials: stabilization mechanisms, multifunctionalities, and applications-a comprehensive review, Mater. Today Energy (2024) 101532, https://doi.org/10.1016/j.mtener.2024.101532.

[14] A. Seppälä, K. Turunen, M.R. Yazdani, Thermal conductivity of sugar alcohols, Sol. Energy Mater. Sol. Cells 243 (2022) 111796, https://doi.org/10.1016/j.solmat.2022.111796.

[15] W.T. Le, A. Kankkunen, O.J. Rojas, M.R. Yazdani, Leakage-free porous cellulose-based phase change cryogels for sound and thermal insulation, Sol. Energy Mater. Sol. Cells 256 (2023) 112337, https://doi.org/10.1016/j.solmat.2023.112337.

[16] S. Yu, X. Li, T. Zhang, X. Yuan, L. Chen, B. Yang, Y. Wang, Improving the shape stability and photothermal conversion performance of phase change materials through three-dimensional metal-organic network, Mater. Today Commun. 38 (2024) 108194, https://doi.org/10.1016/j.mtcomm.2024.108194.

[17] D.M.S. Al-Homoud, Performance characteristics and practical applications of common building thermal insulation materials, Build. Environ. 40 (3) (2005) 353–366, https://doi.org/10.1016/j.buildenv.2004.05.013.

[18] M.R. Yazdani, R. Ajdary, A. Kankkunen, O.J. Rojas, A. Seppälä, Cellulose nanofibrils endow phase-change polyethylene glycol with form control and solid-to-gel transition for thermal energy storage, ACS Appl. Mater. Interfaces 13 (5) (2021) 6188–6200, https://doi.org/10.1021/acsami.0c18623.

[19] R. Jilte, A. Afzal, Ü. Ağbulut, S. Shaik, S.A. Khan, E. Linul, M. Asif, Battery thermal management of a novel helical channeled cylindrical Li-ion battery with nanofluid and hybrid nanoparticle-enhanced phase change material, Int. J. Heat. Mass Transf. 216 (2023) 124547, https://doi.org/10.1016/j.ijheatmasstransfer.2023.124547.

[20] T. Wu, C. Wang, Y. Hu, X. Zeng, M. Song, Flexible solid-solid phase change materials with high stability for thermal management, Int. J. Heat. Mass Transf. 211 (2023) 124202, https://doi.org/10.1016/j.ijheatmasstransfer.2023.124202.

[21] M.R. Yazdani McCord, A. Kankkunen, D. Chatzikosmidou, A. Seppälä, J. Seppälä, H. Baniasadi, Polypyrrole-modified flax fiber sponge impregnated with fatty acids as bio-based form-stable phase change materials for enhanced thermal energy storage and conversion, J. Energy Storage 81 (2024) 110363, https://doi.org/10.1016/j.est.2023.110363.

[22] M. Kenisarin, K. Mahkamov, Solar energy storage using phase change materials, Renew. Sustain. Energy Rev. 11 (9) (2007) 1913–1965, https://doi.org/10.1016/j.rser.2006.05.005.

[23] J. Liu, D. Jiang, H. Fei, Y. Xu, Z. Zeng, W. Ye, Preparation and properties of lauric acid-octadecanol/expanded graphite shape-stabilized phase change energy storage material, Mater. Today Commun. 31 (2022) 103325, https://doi.org/10.1016/j.mtcomm.2022.103325.

[24] I.Y. Miranda-Valdez, M.R. Yazdani, T. Mäkinen, S. Coffeng, L. Viitanen, J. Koivisto, M.J. Alava, Cellulose foams as scalable templates for phase change materials, J. Energy Storage 73 (2023) 109036, https://doi.org/10.1016/j.est.2023.109036.

[25] K. Turunen, M.R. Yazdani, A. Santasalo-Aarnio, A. Seppälä, Exceptional cold-crystallization kinetics of erythritol-polyelectrolyte enables long-term thermal energy storage, Sol. Energy Mater. Sol. Cells 230 (2021) 111273, https://doi.org/10.1016/j.solmat.2021.111273.

[26] A. Kumar, G. Varshney, Thermally switchable metal-free THz electromagnetic shield using phase change material, Mater. Today Commun. 38 (2024) 108007, https://doi.org/10.1016/j.mtcomm.2023.108007.

[27] H. Baniasadi, M. Madani, J. Seppälä, J.B. Zimmerman, M.R. Yazdani, Form-stable phase change electrospun nanofibers mat with thermal regulation and biomedical multi-functionalities, J. Energy Storage 68 (2023) 107660, https://doi.org/10.1016/j.est.2023.107660.

[28] G. Reyes, R. Ajdary, M.R. Yazdani, O.J. Rojas, Hollow filaments synthesized by dry-jet wet spinning of cellulose nanofibrils: structural properties and thermoregulation with phase-change infills, ACS Appl. Polym. Mater. 4 (4) (2022) 2908–2916, https://doi.org/10.1021/acsapm.2c00177.

[29] M. Yazdani, N.M. Mahmoodi, M. Arami, H. Bahrami, Surfactant-modified feldspar: isotherm, kinetic, and thermodynamic of binary system dye removal, J. Appl. Polym. Sci. 126 (2012) 340–349, https://doi.org/10.1002/app.36883.

[30] M.S. Al-Homoud, Performance characteristics and practical applications of common building thermal insulation materials, Build. Environ. 40 (3) (2005) 353–366, https://doi.org/10.1016/j.buildenv.2004.05.013.

[31] J. Zhang, W. Liu, O. Gauthier, S. Sourice, P. Pilet, G. Rethore, K. Khairoun, J.-M. Bouler, F. Tancret, P. Weiss, A simple and effective approach to prepare injectable macroporous calcium phosphate cement for bone repair: Syringe-foaming using a viscous hydrophilic polymeric solution, Acta Biomater. 31 (2016) 326–338, https://doi.org/10.1016/j.actbio.2015.11.055.

[32] X. Fan, Y. Guan, Y. Li, H.-Y. Yu, J. Marek, D. Wang, J. Militky, Z.Y. Zou, J. Yao, Shape-stabilized cellulose nanocrystal-based phase-change materials for energy storage, ACS Appl. Nano Mater. 3 (2) (2020) 1741–1748, https://doi.org/10.1021/acsanm.9b02441.
[33] Z. Shen, M. Qin, F. Xiong, R. Zou, J. Zhang, Nanocellulose-based composite phase change materials for thermal energy storage: status and challenges, Energy Environ. Sci. 16 (3) (2023) 830–861, https://doi.org/10.1039/D2EE04063H.
[34] M. Cheng, J. Hu, J. Xia, Q. Liu, T. Wei, Y. Ling, W. Li, B. Liu, One-step in-situ green synthesis of cellulose nanocrystal aerogel based shape stable phase change material, Chem. Eng. J. 431 (1) (2022) 133935, https://doi.org/10.1016/j.cej.2021.133935.
[35] M. Song, J. Jiang, J. Zhu, Y. Zheng, Z. Yu, X. Ren, F. Jiang, Lightweight, strong, and form-stable cellulose nanofibrils phase change aerogel with high latent heat, Carbohydr. Polym. 272 (2021) 118460, https://doi.org/10.1016/j.carbpol.2021.118460.
[36] X. Duan, X. Yang, G. Ren, Y. Pang, L. Liu, Y. Liu, Technical aspects in freeze-drying of foods, Dry. Technol. 34 (11) (2016) 1271–1285, https://doi.org/10.1080/07373937.2015.1099545.
[37] S. Arif, G. Burgess, R. Narayan, B. Harte, Evaluation of a biodegradable foam for protective packaging applications, Packag. Technol. Sci. Int. J. 20 (6) (2007) 413–419, https://doi.org/10.1002/pts.770.
[38] K. Li, M. Gao, Z. Li, H. Yang, L. Jing, X. Tian, Y. Li, S. Li, H. Li, Q. Wang, Multi-interface engineering of solar evaporation devices via scalable, synchronous thermal shrinkage and foaming, Nano Energy 74 (2020) 104875, https://doi.org/10.1016/j.nanoen.2020.104875.
[39] E.M. Zadeh, S.F. O'Keefe, Y.T. Kim, Utilization of lignin in biopolymeric packaging films, ACS Omega 3 (7) (2018) 7388–7398, https://doi.org/10.1021/acsomega.7b01341.
[40] R. Shorey, A. Gupta, T.H. Mekonnen, Hydrophobic modification of lignin for rubber composites, Ind. Crops Prod. 174 (2021) 114189, https://doi.org/10.1016/j.indcrop.2021.114189.
[41] L. Liu, B. Cui, L. Tan, W. Wang, Improving the combination of cellulose and lignin using xylan as a compatibilizer, Cellulose 28 (9) (2021) 5335–5349, https://doi.org/10.1007/s10570-021-03864-0.
[42] A.B. Gala, M. Raugei, P. Fullana-i-Palmer, Introducing a new method for calculating the environmental credits of end-of-life material recovery in attributional LCA, Int. J. Life Cycle Assess. 20 (2015) 645–654, https://doi.org/10.1007/s11367-015-0861-3.
[43] F. Franks, T. Auffret, Freeze-drying of pharmaceuticals and biopharmaceuticals, R. Soc. Chem. (2008), https://doi.org/10.1039/9781847557704.
[44] T.M. Oyinloye, W.B. Yoon, Effect of freeze-drying on quality and grinding process of food produce: A review, Processes 8 (3) (2020) 354, https://doi.org/10.3390/pr8030354.
[45] K. Struhala, M. Ostrý, Life-Cycle Assessment of phase-change materials in buildings: a review, J. Clean. Prod. 336 (2022) 130359, https://doi.org/10.1016/j.jclepro.2022.130359.
[46] M.D. Heidari, D. Mathis, P. Blanchet, B. Amor, Streamlined life cycle assessment of an innovative bio-based material in construction: a case study of a phase change material panel, Forests 10 (2019) 160, https://doi.org/10.3390/f10020160.
[47] S. Serrano, C. Barreneche, L. Rincón, D. Boer, L.F. Cabeza, Optimization of three new compositions of stabilized rammed earth incorporating PCM: thermal properties characterization and LCA, Constr. Build. Mater. 47 (2013) 872–878, https://doi.org/10.1016/j.conbuildmat.2013.05.018.