Transparent Wood for Thermal Energy Storage and Reversible Optical Transmittance

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ABSTRACT: Functional load-bearing materials based on phase-change materials (PCMs) are under rapid development for thermal energy storage (TES) applications. Mesoporous structures are ideal carriers for PCMs and guarantee shape stability during the thermal cycle. In this study, we introduce transparent wood (TW) as a TES system. A shape-stabilized PCM based on polyethylene glycol is encapsulated into a delignified wood substrate, and the TW obtained is fully characterized, also in terms of nano- and mesoscale structures. Transparent wood for thermal energy storage (TW-TES) combines large latent heat (∼76 J g⁻¹) with switchable optical transparency. During the heating process, optical transmittance increases by 6% and reaches 68% for 1.5 mm thick TW-TES. Characterization of the thermal energy regulation performance shows that the prepared TW-TES composite is superior to normal glass because of the combination of good heat-storage and thermal insulation properties. This makes TW-TES composites interesting candidates for applications in energy-saving buildings.

KEYWORDS: wood, encapsulation, phase-change material, energy storage, biocomposite

INTRODUCTION

The global energy consumption in the building sector exceeds 30%, and technologies for reduced energy consumption and CO₂ emission are therefore under development. Transparent wood (TW) composites are of interest in energy-saving buildings because of low thermal conductivity (0.15 W m⁻¹ K⁻¹), high optical transmittance (92% for 1.5 mm thick), and haze (80% for 1 mm thick), as well as favorable mechanical properties. To prepare TW, a wood template is first chemically treated for removing lignin, which is the major light-absorbing component in wood. In a second step, light scattering is reduced by infiltrating the pore space with a polymer component matching the wood refractive index. In comparison to glass, which is generally used for light-transmitting building structures, the TW shows better thermal insulation and could reduce heat loss. Furthermore, the TW exhibits a combination of high optical transmittance and haze, which offers uniform indoor lighting while preserving privacy. Most of the reported studies on TW for energy-saving applications focus on the excellent thermal insulating properties of this material. Thermal energy storage (TES) has emerged as a key technology to harvest and store solar energy. Latent heat storage systems based on phase-change materials (PCMs) are among the most efficient in reducing energy consumption because of their large latent heat capacity over a narrow temperature range. PCMs act as temperature regulators that can store and release thermal energy in order to shift the heating and cooling loads over time and thus reduce heat energy loss. In particular, organic PCMs store latent heat energy during a reversible solid–liquid phase-change process that occurs at the melting temperature of the material. The present study focuses on the incorporation of polyethylene glycol (PEG) in the TW. PEG is nontoxic and biodegradable, with a large latent heat and a tunable solid–liquid transition temperature that can be varied by tuning PEG molecular weight. However, solid–liquid PCMs such as PEG suffer from leakage issues during melting, which limits their long-term thermal reliability. In order to address leakage issues, PEG can be entrapped by a surrounding polymer matrix, such as polymethyl methacrylate (PMMA). Porous supporting materials can be used to encapsulate the PCM, such as silica and cellulosic networks. Here, wood is investigated as a PCM substrate. Wood is an abundant and renewable material, which has a unique hierarchical structure with organized cells formed during tree growth. Wood cells have structurally advantageous micropore sizes for chemical modification and polymer impregnation. The diameter could be 10–50 μm, with length at the scale of millimeters. Because delignified wood (DW) has a unique mesoporous cell wall structure, a high specific area, and a high adsorption capacity, it is an attractive supporting material for PCM encapsulation. Therefore, in this study, the PCM was encapsulated into a DW scaffold. Very few studies report the use of wood scaffolding as a porous supporting material for...
Excellent TES properties are reported in the present study, associated with high optical transmittance and haze. A multifunctional TW material was prepared via a two-step procedure, which involves wood delignification followed by encapsulation of the PEG/PMMa polymer blend into the porous DW scaffold. A schematic representation of the preparation procedure of the multifunctional TW, labeled as a transparent wood for thermal energy storage (TW-TES), is shown in Figure 1. TW-TES can store thermal energy even for small temperature variations and combines excellent latent heat storage properties with low thermal conductivity, tunable optical transmittance, and robust mechanical performance. These properties make the multifunctional TW-TES composite a potential candidate for heat-transfer control in building applications, such as light-transmitting structures with reduced heat energy consumption and improved thermal comfort indoors. The objectives are to investigate the TW-TES concept and its potential. Also, the mesoscale structures are investigated so that the role of PCM distribution in TW-TES can be assessed.

**EXPERIMENTAL SECTION**

**Materials.** Silver birch wood (*Betula pendula*, purchased from Glimakra of Sweden AB) with an oven-dried density of 620 kg/m³ was used as a raw material. Specimens with dimensions of 2 cm × 2 cm × 0.5 mm and 2 cm × 2 cm × 1.5 mm were prepared. More specimens were prepared for mechanical testing with dimensions of 5 mm × 60 mm × 1.5 mm. Sodium chlorite (*NaClO₂*, Alfa Aesar), 2,2′-azobis(2-methylpropionitrile) (AIBN, Sigma-Aldrich), and PEG (Sigma-Aldrich) with a molecular weight of 1000 g mol⁻¹ were used as received. Methyl methacrylate (MMA, Sigma-Aldrich) was passed through a column of neutral aluminum oxide (Sigma-Aldrich) prior to use to remove the inhibitor.

**Delignification of the Wood Samples.** The delignification process was performed using NaClO₂ (1 wt %) in an acetate buffer solution (pH 4.6) at 80 °C. The reaction was ended once the wood samples became white. The DW samples were then thoroughly washed several times with deionized water and sequentially dehydrated with ethanol and acetone.

**Preparation of TW-TES Composites.** PEG 1000 was heated at 45 °C until complete dissolution. MMA (30 wt %) was added to the molten PEG (70 wt %) solution and kept under stirring at 45 °C for 10 min. The initiator, AIBN (0.3 wt % based on MMA), was then added to the PEG/MMA solution, and the blend was continuously stirred at 45 °C for 10 min. The maximum PEG/PMMa ratio without any leakage during phase transition was 70/30 (w/w). The DW sample was then immersed in the blend solution for infiltration. Infiltration was performed under vacuum at room temperature for about 15 min; it was repeated three times. The blend solution containing the DW samples was heated at 40 °C for a few minutes between each infiltration to delay PEG crystallization. The infiltrated DW template was finally sandwiched between two glass slides, packaged with aluminum foil, and polymerized in an oven at 70 °C for 4 h. The final TW-TES composites prepared have thicknesses of 0.5 and 1.5 mm and possess a thin layer of PEG/PMMa at their surfaces. The density of the final composite was determined by measuring their dimensions and weight. The DW volume fraction was determined and is presented in Table S1.

**Characterization.** The cross-sectional morphologies of TW-TES composites were characterized by field emission scanning electron microscopy (Hitachi SEM S-4800, Japan). A differential scanning calorimeter (Mettler Toledo DSC1, Switzerland) was used to analyze the thermal properties of neat PEG, PEG/PMMa polymer blend, and TW-TES composite under nitrogen with a flow rate of 50 mL min⁻¹. The samples were heated from 0 to 60 °C with a heating rate of 2 °C min⁻¹. The latent heat was determined by numerical integration of the area of the thermal transition peak. The thermal reliability of the TW-TES composite was investigated by performing 20 heating–cooling cycles. The data were recorded from the second scan. The thermal stability of the TW-TES composite was studied by thermogravimetric analysis (TGA) using a Mettler Toledo TGA/DSC1 (Switzerland) with a heating rate of 10 °C min⁻¹ from room temperature to 600 °C under nitrogen atmosphere. A light flash apparatus (LFA 467 HyperFlash Netzsch, Germany) was employed to determine cross-plane thermal diffusivities (α) on square samples (20 × 20 × 1.5 mm). The measurements were performed at 45 °C in order to avoid the PCM phase transition. The sample surfaces were graphitized using a graphite spray before the measurements. Thermal conductivity, κ, was then calculated from the equation \( κ = \rho C_p \alpha \), where \( \rho \) is the density and \( C_p \) is the specific heat capacity of the material. The chemical structure of native wood (NW), DW, neat PEG, neat PMMA, and TW-TES composite was evaluated by Fourier transform infrared spectrometry (FTIR) using a PerkinElmer spectrum 100 FT-IR equipped with a MXTI Golden Gate, single-reflection accessory unit with a diamond ATR crystal (Graseby Specac Ltd., UK). The spectra were recorded at room temperature in the range of 4000–600 cm⁻¹. Tensile tests and three-point bending tests were performed using an Instron 5944 (USA) instrument equipped with a 500 N load cell. The tests were carried out with a 10% min⁻¹ strain rate and span of 30 mm. All samples were preconditioned for 24 h and tested in a room at a temperature of 22 ± 1 °C and 50 ± 2% relative humidity. The optical measurements were performed using an integrating sphere in the visible and NIR wavelength region (400–1000 nm). A Quartz tungsten halogen light source (model 66181 from Oriel Instruments) with strong, stable output mainly in the visible and NIR region was used. The visible transmittance and haze were measured according to ASTM D1003 “Standard Method for Haze and Luminous Transmittance of Transparent Plastics” by placing the sample at the entrance port of the integrating sphere and following the same procedure as previously reported in another study. Thermal images were captured using an infrared camera (FLIR A655sc).
ended libriform fibers provide mechanical support, whereas water conduction is supplied by open-ended vessels. Birch fibers and vessels have a mean diameter of 18 and 82 μm, respectively. At the ultrastructural level, the cell wall structure of the wood cells consists of layers with oriented cellulose microfibrils bonded by a mixed hemicelluloses-lignin matrix. Figure 2a shows the SEM image of the cell wall structure of birch wood. The delignification process adds nanoscale porosity in the middle lamella (lignin-rich region) and in the cell wall, while preserving the anisotropic structure of wood (Figures 2b and S1). PEG with a molecular weight of 1000 g mol⁻¹ was chosen as the PCM owing to its excellent energy storage capacity at a suitable working temperature range; in addition, PEG is highly miscible with wood.32–34 In order to guarantee high-energy density and avoid PEG leakage during the phase transition, PEG (70 wt %) was mixed with MMA monomer (30 wt %) and AIBN as an initiator, the blend was infiltrated in the DW scaffold, and then MMA polymerized, so that a polymer blend termed PEG/PMMA was formed. The DW scaffold is used as a supporting structure for encapsulation of the PEG/PMMA polymer blend. TW-TES composites with thicknesses of 0.5 and 1.5 mm were prepared. The morphology of the TW-TES composite was investigated using SEM and is presented in Figure 2c. Upon infiltration, the wood microstructure is preserved and the PEG/PMMA blend is homogeneously distributed at the scale of 20 μm so that the cell lumen is completely filled by the polymer phase. PEG/PMMA is of immiscible two-phase polymer blend.19,20 No interface gaps were apparent between PEG/PMMA and the wood cell wall. This indicates favorable interfacial interaction between PEG/PMMA polymer blend and the wood cell wall (Figure 2d). This may be attributed to the good miscibility between the hydrophilic PEG phase and the wood cell wall. Figure 2e shows the interior of TW-TES wood cell wall where polymer domains are apparent, indicating the successful impregnation of the wood cells.

In order to assess if the PEG phase is located in the cell wall, a DW piece was soaked in a 30 wt % PEG 1000 aqueous solution and the cell wall morphology was evaluated. Figure S2 shows a high-resolution SEM image of the wood cell wall after PEG infiltration. After PEG infiltration, PEG-rich domains could be identified and no pores were detected at this scale in contrast with DW before infiltration (Figure S1). The similarities between the SEM micrographs of the cell wall interior shown in Figures 2e and S2 provide support for PEG diffusion into the wood cell wall. Figure 2e indicates that the PEG-rich domain inside the cell wall is at the scale of 100–200 nm particles.

The chemical interactions between the PCM and the DW template in TW-TES composites were analyzed by the FTIR analysis. FTIR spectra of NW, DW, neat PMMA, neat PEG, and TW-TES composite are presented (Figure 2f). In comparison with NW, the peak at 1505 cm⁻¹ attributed to aromatic skeleton vibrations from lignin is lacking from the DW spectrum.35 This confirms successful removal of lignin by delignification. In addition, all of the major characteristic absorption peaks of DW, PEG, and PMMA are present in the TW-TES composite. The absorption peak of PMMA ascribed to C=O stretching (1725 cm⁻¹) is present. Moreover, shift in the O–H stretching vibration peak in the TW-TES (3419 cm⁻¹) spectrum compared with that of DW (3350 cm⁻¹) and PEG (3468 cm⁻¹) indicates hydrogen-bonding interactions between PEG and DW. The PEG absorption peak at 947 cm⁻¹, attributed to −CH₂ vibrations,36 is shifted in TW-TES, also suggesting intermolecular interaction between PEG and DW. Thus, the DW template can effectively prevent leakage because of capillary forces and intermolecular interactions with the small-scale PCM domains. No new absorption peak is apparent in the TW-TES spectrum, so that only intermolecular interactions are expected between PEG and PMMA and between PEG and the DW template. The stabilization of PEG in PMMA and DW template does not significantly alter the chemical structure of PEG, and the thermal properties of PEG are possibly preserved.

**Thermal Properties.** The TES properties are important for the application of TW-TES in energy-saving buildings.
Thermal storage cycles include two steps: absorption of latent heat by the crystalline polymer (PEG) upon heating above the melting temperature ($T_m$) and latent heat release during cooling until the crystallization temperature ($T_c$). The melting and cooling curves of neat PEG, PEG/PMMA blend, and TW-TES composite are presented in Figure 3a,b, respectively. TW-TES shows excellent thermal properties with latent heat of melting and crystallization of about 76 and 74 J g$^{-1}$, respectively. The melting process of TW-TES occurs over a temperature range between $\sim$27 and $\sim$40 °C, with a $T_m$ of 38 °C. When heated above $T_m$, the TW-TES composite consists of molten PEG encapsulated into the wood template and surrounded by PMMA. At this stage, the TW-TES stores latent heat of the liquid PCM. Cooling until the crystallization point ($T_c = 29$ °C) will initiate the crystallization of PEG and hence release the stored latent heat. The differential scanning calorimetry (DSC) curves show that TW-TES exhibits endothermic and exothermic behavior similar to the PEG/PMMA blend. Neat PEG exhibits a shoulder on the leading edge of the endothermic peak associated with the melting point of crystallites. The presence of a shoulder is a consequence of heterogeneous melting and is attributed to variations in crystal thicknesses.$^{17,37}$ Further support is given in Table 1, which summarizes the latent heat of melting ($\Delta H_m$) and crystallization ($\Delta H_c$) and characteristic temperatures ($T_m$ and $T_c$) of neat PEG, PEG/PMMA, and TW-TES composite. The theoretical data were defined as follow: $\Delta H_{Th} = X \cdot \Delta H_{PEG}$, where $\Delta H_{Th}$ is the theoretical latent heat, $\Delta H_{PEG}$ is the latent heat of neat PEG, and $X$ is the mass fraction of PEG. The enthalpy values of PEG/PMMA blend and TW-TES composite are proportional to the PEG fraction and are shown in Table 1. The experimental results for TW-TES were close to predicted data based on PEG/PMMA behavior. This can be attributed to the preserved thermal performance of PEG in TW-TES, and it indicates that the domain size of PEG is sufficiently large so that PEG preserves its melting and crystallization behavior. If PEG was completely soluble in the wood cell wall, the melting and crystallization behavior could have been altered in a negative manner because of the lower PEG crystallinity.

The phase-change phenomenon is critical to TW-TES performance, and it was therefore investigated if PEG could crystallize in the wood cell wall. A DW template was soaked in 30 wt % aqueous PEG solution in order to selectively infiltrate the wood cell walls, while keeping the lumen pore space free of polymer. The DSC analysis was performed on the DW template with PEG infiltrated cell walls (Figure S3). Latent heat of melting and crystallization of, respectively, 39 and 38 J

Figure 3. (a) DSC scans of neat PEG, PEG/PMMA blend, and TW-TES obtained while heating from 0 to 60 °C followed by (b) cooling process. (c) DSC curves of TW-TES before and after thermal cycling, indicating good thermal reliability after 20 cycles. (d) TG/DTG curves of DW, neat PEG, neat PMMA and TW-TES.

|                  | $T_m$ [°C]$^a$ | $\Delta H_m$ [J g$^{-1}$]$^b$ | $\Delta H_{Th}^c$ [J g$^{-1}$] | $T_c$ [°C]$^d$ | $\Delta H_c$ [J g$^{-1}$]$^e$ | $\Delta H_{Th}^f$ [J g$^{-1}$] |
|------------------|----------------|-----------------------------|-----------------------------|----------------|-----------------------------|-----------------------------|
| neat PEG         | 37.8 ± 0.3     | 159.0 ± 2.1                 |                            | 29.5 ± 0.7     | 161.5 ± 2.5                 |                            |
| PEG/PMMA (70/30 w/w) | 36.9 ± 0.2     | 103.0 ± 2.5                 | 111.3                       | 25.5 ± 1.2     | 101.1 ± 0.2                 | 113.1                       |
| TW-TES composite | 38.3 ± 0.3     | 76.3 ± 3.4                  | 77.3                        | 28.6 ± 1.3     | 74.2 ± 3.1                  | 78.5                        |

$a$Melting temperature. $^b$Latent heat of melting. $^c$Theoretical latent heat of melting. $^d$Crystallization temperature. $^e$Latent heat of crystallization. $^f$Theoretical latent heat of crystallization. The values in the tables are given as mean ± standard deviations.
g⁻¹ were achieved when PEG was located in the wood cell wall only. It is concluded that crystallization of PEG does not only take place in the microscale lumen space but also in the wood cell wall. This provides further information with respect to PEG distribution and supports that the wood cell wall contains PEG-rich domains. The PCM penetrates throughout the whole wood template, ensuring high heat storage efficiency in TW-TES.

The thermal reliability of the TW-TES composite was evaluated by performing 20 heating−cooling cycles. Data are presented in Figure 3c. After 20 cycles, the phase-change temperatures, \( T_m \) and \( T_c \), remain comparable with those at the initial state. After cycling, the latent heat of crystallization \( \Delta H_c \) showed a minor increase of 0.55% because of enhanced nucleation of PEG crystallites after several heating−cooling cycles. The latent heat of melting, \( \Delta H_m \), did not show any variations. TW-TES exhibits excellent thermal reliability after 20 thermal cycles.

The thermal stability was evaluated using TGA in nitrogen, and the TG curves of neat PEG and TW-TES are shown in Figure 3d. From 30 to 150 °C, TW-TES exhibits only 1.0% weight loss attributed to residual moisture evaporation, which is much lower than that for the DW template (5.3%). TW-TES exhibits a two-step degradation starting from \( \sim 290 \) °C, including the degradation of DW template and the degradation of PMMA and PEG compounds. The thermo-oxidative stability was also studied in oxygen (Figure S4). Under these conditions, thermal oxidation of TW-TES starts from \( \sim 190 \) °C. The degradation of TW-TES is initiated by the production of formic esters originating from the degradation of PEG. Overall, these results show that TW-TES is thermally stable below \( \sim 290 \) °C in the absence of oxygen (nitrogen environment), and below \( \sim 190 \) °C in an oxidative environment (oxygen).

Thermal insulating materials are essential in limiting heat loss and thus reducing energy consumption in buildings. A light flash test was employed in order to preliminary evaluate the thermal conductivity of TW-TES composites. TW-TES has efficient thermal insulation properties with a low cross-plane thermal conductivity of 0.30 W m⁻¹ K⁻¹, compared with 1.36 W m⁻¹ K⁻¹ for normal glass (Table S2). The combination of heat storage and thermal insulation properties is favorable.

Optical Function and Mechanical Properties. Wood optical transparency is obtained by first partially removing the lignin component and then introducing a material matching closely the wood refractive index in order to reduce the light scattering sources located at the air/wood interfaces in the lumen area and inside the cell wall. In our study, optical transmittance was obtained by infiltrating the PEG/PMMA blend in the porous DW structure. TW-TES shows good optical properties with switchable optical transmittance during the phase-change process. The optical properties of TW-TES were determined using an integrating sphere at temperatures below and above \( T_m \) to assess the variations in optical properties with temperature (Figure 4a,b). The optical transmittance of TW-TES increases by 6% at \( \lambda = 550 \) nm after the phase change (\( T > T_m \)) as PEG crystals melt in the composite. As the temperature is lowered below \( T_m \), the optical transmittance of TW-TES is reduced because of the crystallization of PEG. The change in transmittance can be visually observed as the TW-TES composite changes from opaque to transparent when undergoing the phase transitions (Figure 4c). Above \( T_{sw} \), the weight of TW-TES is reduced by 68%. Lower thickness results in higher optical transmittance because a shorter light pathway lowers light attenuation. The TW-TES specimen with a thickness of 0.5 mm results in transmittance up to 84% above \( T_{sw} \). It should be noted that the PEG/PMMA blend has
Overall, the work to fracture of TW-TES (1.9 MJ m$^{-2}$ ± failure (0.2 ± 0.3) strength of 74.5 MPa).38,40 Haze is the percentage of transmitted light mismatch between holocellulose (1.53), PEG (1.46), and PMMA (1.49).38,40 Haze is the percentage of transmitted light that is scattered at large angles from the direction of light propagation.41 The haze of 1.5 mm thick TW-TES (77% at $\lambda = 550$ nm) did not significantly vary with temperature. As the haze was measured by subtracting the transmitted light at small scattering angles, the visual difference might be due to transmitted light which is scattered to larger angles. A relative decrease in haze of 3% was observed for the thinner specimens when heated above $T_m$. This can be attributed to a reduction in scattering centers, such as voids, upon melting of PEG crystals. The visual change in transparency of TW-TES with increased temperature can be advantageous for applications such as temperature sensors for the environment and light-transmitting structures with homogeneous lighting.

The mechanical properties were evaluated below $T_m$ using three-point bending and tensile tests. The reduced stress–strain curves of TW-TES from flexural tests for longitudinal properties are shown in Figure S5. TW-TES shows a flexural strength of 74.5 ± 10.2 MPa and a strain to failure as high as 3.2 ± 0.6%. Glass shows brittle nature with a low strain to failure (0.2 ± 0.1%), whereas TW-TES is much more ductile (3.2%). Overall, the work to fracture of TW-TES (1.9 MJ m$^{-3}$, area under stress–strain curve) is about 10 times higher than that for glass (0.2 MJ m$^{-3}$) because of more favorable mechanisms of failure in TW-TES composites. Tensile test results for NW and TW-TES are shown in Figure 4d. TW-TES shows high elastic modulus (14.9 ± 1.3 GPa) compared with polymers, and this is similar to NW (14.5 ± 3.4 GPa). TW-TES shows a strength of 70.5 ± 6.6 MPa, which is lower than that for NW (129.6 ± 10.4 MPa). The lower strength of the TW-TES composite could be attributed to the presence of residual nanoporosity in the cell wall because of delignification. In addition, PEG impregnation reduces mechanical strength because molar mass and PEG glass-transition temperature are low, so that stress transfer between cellulose fibrils in the cell wall is compromised.38 In general, TW-TES shows satisfactory mechanical strength and modulus for load-bearing applications. An important reason is the relatively low porosity of birch wood, which means that the content of wood cell wall material is high.

In order to demonstrate the TES performance of TW-TES in real applications, an infrared camera was used to record temperature changes in TW-TES during heating. Specimens at room temperature were placed on a heating plate set at a temperature of 60 °C, and the time for each specimen to reach the heating plate temperature was recorded. Figure 5 shows thermal images of TW-TES and glass specimens when placed in contact with a surface heated at 60 °C. The samples were transferred simultaneously from room temperature to the heated surface (Figure 5a). After 20 s, the glass specimen reached the temperature of the heated surface (Figure 5b), indicating the fast temperature equilibration. In contrast, TW-TES required 210 s of heating in order to reach the equilibrium temperature (Figure 5c), showing the heat storage capacity and thermal energy regulating property of TW-TES. The equilibration temperature of TW-TES was compared with TW without PCM, prepared as previously reported.6 The results show that temperature equilibrium of the specimens is first reached by glass, followed closely by TW without PCM, and finally by TW-TES. Compared with TW without PCM, TW-TES shows great efficiency in terms of TES. These differences between glass and TW-TES are attributed to the lower thermal conductivity of TW-TES and to the ability of the TW-TES composite to absorb and store thermal energy. TW-TES is a candidate as heat-storage and light-transmitting material for applications in energy-saving buildings.

### CONCLUSIONS

This study presents a novel concept of multifunctional TW-TES that combines mechanical properties, TES, and optical transmittance. A mesoporous DW template was used as a supporting, nonleaking substrate material for encapsulation of the PCM. The TW-TES composite was obtained by impregnation with a liquid PEG/MMA blend, followed by MMA polymerization. The final impregnated polymer component was a 70/30 (w/w) physical blend of PEG/MMMA.

The TES properties of the TW-TES composite were highly favorable and proportional to the 49 wt % PEG content. The TES function was combined with high mechanical properties. In addition, the materials showed tunable optical transmittance when the external temperature was increased because the PEG phase was melting. With increasing thickness, the optical transmittance is reduced because of increasing light attenuation. This can be a limitation for applications of higher thickness TW-TES as light-transmitting materials. The optical transmittance of TW-TES can be enhanced by reducing the wood volume fraction by using higher porosity wood species, so that the number of wood/polymer interfaces is lowered.

Dissolution experiments suggest that the PEG phase was continuous in the wood lumen pore space. The morphological
analysis of TW-TES composites also indicates that a PEG-rich phase was present in the wood cell wall, in the form of 100–200 nm mesoscale domains. The PEG distribution is critical to the mechanical, optical, and TES properties of the material. The design of TW-TES materials requires optimization of mechanical properties with respect to TES capacity. The PEG content could be increased by the selection of higher porosity wood species, but then the mechanical performance would be reduced.

The thermal properties of the TW-TES composite can be further tailored, for instance, by tuning the melting temperature of PEG. The present concept of multifunctional TW materials can expand the applications of wood-based materials to new areas including temperature sensors, heat storage, and light-transmitting structural materials.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.9b05525.

SEM image of the interior of DW cell wall, SEM image of the interior of DW cell wall infiltrated with PEG; DSC curve of PEG infiltrated into DW cell wall; TGA of DW, PEG, PMMA, and TW-TES in oxygen; three-point bending stress–strain curves of glass and TW-TES; weight fraction and volume fraction in the TW-TES composite; and thermal conductivity of glass and TW-TES (PDF).

Thermal evolution of TW-TES, TW without PCM, and glass specimens when placed from room temperature to a surface heated at 60 °C (MP4)

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**Notes**

The authors declare no competing financial interest.

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