Properties of Hydrogel-Wood Composite as a New Thermochromic Glazing Material

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ABSTRACT

Recently, thermal-response hydrogel smart window is widely studied because of its high luminous transmittance (τ_{lum}) and high solar modulation ability ($\Delta\tau_{sol}$). However, its liquid state is undesirable for window applications. Wood has strong mechanical strength and low thermal conductivity. Due to the unique features of the thermal-response hydrogel and wood, a thermochromic hydrogel wood composite (HWC) that can smartly regulate solar irradiation is proposed by impregnating a thermal-response hydrogel into delignified wood. The novel HWC demonstrates advanced optical properties (i.e. τ_{lum} = 83% and 40% at the cold transparent and hot opaque states & $\Delta\tau_{sol}$ = 38%) and low transition temperature (i.e. T_c = 23 °C). Moreover, the HWC is highly flexible and easily fitted into existing windows frames. Overall, the HWC with its impressive features shows great promise for energy-efficient material for smart windows in buildings.

INTRODUCTION

Energy used to achieve thermal comfort in indoor environments accounts for 60% of all energy used in buildings (Catrini, Curto, Franzitta, & Cardona, 2020; Gholamzadehmir, Del Pero, Buffa, Fedrizzi, & Aste, 2020). Nearly 50% of this energy consumption is caused by heat transfer to the building envelopes, especially heat loss through inefficient glass windows (Wang et al., 2018). To improve the energy efficiency of buildings, thermo-, electro- and photo-chromic smart windows that dynamically modulate light transmittance have been widely investigated (Ke et al., 2019; Sun, Wu, & Wilson, 2018). They can be achieved by coating chromic materials (e.g. VO_2, WO_3, AgI, etc.) on conventional glass for light regulation ability. Among them, thermochromic smart window is the most promising, because it can regulate transmittance in response to ambient temperatures, which is totally passive and does not consume any energy. Various thermos-responsive materials have been investigated including vanadium dioxide (VO_2) (M. Li, Magdassi, Gao, & Long, 2017; Warwick & Binions, 2014; Xu, Cao, Luo, & Jin, 2018), perovskites (De Bastiani et al., 2017; Lin et al., 2018; Wheeler et al., 2017; Zhang et al., 2019) and ionic liquids (Y. Chen et al., 2019; J. Zhu et al., 2016). However, there are some drawbacks of these thermochromic materials. For example, the transition temperature of VO_2 is 68 °C, which is considerably high for window applications because normally, the temperature of windows can only go up to 40-50 °C. In addition, the thermochromic effect of VO_2 is significant in the near infrared region that account for about 50% of the solar irradiance. Reflecting 50% of the solar irradiance limits the performance of the smart windows. Similarly, ionic liquid also has high transition temperature (60-70 °C) which makes it hard to apply to smart windows in buildings. With respect to the perovskites, since it is susceptible to humidity so perovskites thermochromic smart windows are required a double-glazed window system to well control the humidity surrounded by the smart windows. Therefore, perovskites thermochromic smart windows are hard to realize in real applications at the current stage. Therefore, a thermochromic material with suitable transition temperature, from 20-30 °C, and stable chemical properties is needed for developing thermochromic smart windows to achieve energy saving.

Recently, researchers found that thermochromic hydrogel demonstrates great potential to be applied as smart windows, and Poly(N-isopropylacrylamide) (PNIPAM) is one of the great candidates. The size of hydrogel particles increases at a high temperature and decreases at a low temperature and this change in size of hydrogel particle is reversible. The particle size at the high temperature is comparable to the wavelength of the visible light, which is from 380-780 nm, so the hydrogel particles can significantly reflect the solar irradiance by the light scattering. Thus, the temperature changes and shows significant transmittance contrast between transparent and opaque states, which contributes to high luminous transmittance (τ_{lum}) and solar modulation ability ($\Delta\tau_{sol}$). In other words, the thermochromic effect of the hydrogel takes place in the visible region which can effectively reflect the solar irradiance and reduce the solar heat gain to indoor environment, so, the energy consumption in the heating, ventilation and air conditioning (HVAC) system can be reduced. In addition, its reasonable transition temperature (i.e. 32 °C (Zhou et al., 2020)) is much lower than traditional VO_2 thermochromic smart windows (e.g. 68 °C for VO_2 (Kong et al., 2020)), exhibiting practical application potential. However, PNIPAM hydrogel is in gel or liquid state, causing poor mechanical strength. Therefore, the hydrogel needs to be sealed in a double-glazed structure to prevent leakage, resulting in a high installation and maintenance cost. Even though the hydrogel can be cross-linked with chemicals to reduce the viscosity, the mechanical strength is still poor. Therefore, the development of a solid transparent thermochromic hydrogel with strong mechanical properties is urgently needed.

Wood as a common construction material has been widely used because the manufacturing processes are simple, and the wood is renewable, mechanically strong and offers low thermal conductivity. It was found that wood can be transparent after being delignified by bleaching chemicals and then impregnated with refractive-index matching polymers (e.g. epoxy (M. Zhu, Song, et al., 2016), polymethyl methacrylate (PMMA) (Y. Li, Fu, Yu, Yan, & Berglund, 2016) and polyvinylpyrrolidone (PVP) (M. Zhu, Li, et al., 2016)). Most importantly, the transparent wood demonstrates high mechanical strength due to the strong cellulose nanofibers and aligned cell skeleton structure in native wood (Fu, Chen, & Sorieul, 2020).

So, in this study, a thermal-response hydrogel integrated with balsa wood for a strong and flexible thermochromic hydrogel transparent wood (HWC) is proposed. The delignified wood acts as the skeleton to encapsulate the hydrogel and enhance the mechanical strength. The novel HWC exhibits advanced optical properties with a high τ_{lum} of 83% and 40% at the transparent and opaque state respectively, high $\Delta\tau_{sol}$ of 38 %, and low transition temperature of 23 ^{o}C. Moreover, with the wood skeleton, the mechanical strength has been significantly improved compared with the thermal-response hydrogel. The HWC is also highly flexible, which makes it can be directly pasted on the existing windows. Overall, the HWC eliminates the mechanical weakness of hydrogel and most importantly, it maintains high transparency and high $\Delta\tau_{sol}$ of thermal-response hydrogel, showing promising future as smart window materials.

METHODS

Fabrication of the HWC

Balsa wood was selected as the raw material in this study, because it is one of the lightest woods (i.e. 100-250 kg m^{-3}) and is widely used in construction and decoration areas. In addition, the fast growth rate and thick trunk of balsa wood make it suitable for large-scale cutting methods (e.g. rotary cutting) (Borrega, Ahvenainen, Serimaa, & Gibson, 2015). The fabrication process of HWC is shown in Figure 1. The balsa wood slices were delignified by immersing in 5% sodium hypochlorite solution for 12 hours to remove cellulose, hemicellulose, and lignin (C. Chen et al., 2020). The delignification process significantly reduces the thickness of cell walls and makes it more porous. The delignified wood is white because of the strong light scattering in the microchannels and the reflection on the surface of the wood. Next, the samples were washed three times by ethanol and deionized water. Subsequently, for the thermo-response hydrogel, 0.33g N-isopropylacrylamide (BIS) was dissolved in 13.7 ml deionized (DI) water at 80 ^{o}C in a reaction vessel. Then, the 0.76ml of 0.19M sodium dodecyl sulfate was added into the reaction vessel. The chemical reaction was initiated with 0.76 ml of 0.36 M ammonium persulfate (APS). 1 minute later, 3.36g NIPAm and 168 mg BIS dissolved into 21.8 ml DI water was pumped into the reaction vessel at the rate of 100 µl/min using a syringe pump. After that, the PNIPAM solution can be achieved. Next, 3 g acrylamide, 2 mg BIS and 20 mg KPS were dissolved in 10 ml PNIPAM solution. At last, 30 µL of N,N,N,N-tetramethylethylenediamine (TEMED) was added to the above homogeneous solution to finish the fabrication of the thermo-response hydrogel. To achieve thermochromism and transparency, the delignified balsa wood was immersed in the hydrogel solution and impregnated under a vacuum condition. The impregnation process was repeated several times for the full impregnation. Last, the wood slice was solidified at room temperature to obtain the HWC.

Characterizations of the HWC

To measure the transition temperature, the samples were heated and cooled between 10 °C to 40 °C at intervals of 1.5 °C. Simultaneously, the visible light transmittance (550 nm) of the samples was measured using a Lens Transmission meter at each step. T_c was determined by plotting the first derivative of the transmittance to the temperature as a function of temperature, and the T_c is the temperature showing the minimum value at the first derivative. The transmittance spectra were measured by a UV-VIS-NIR spectrophotometer. The τ_{lum} and $\Delta\tau_{sol}$ are two important indicators for thermochromic smart windows (Liu et al., 2020). τ_{lum} is the amount of visible light transmitted by the windows that is useful for human vision under normal conditions, which is defined as

$$\tau_{lum} = \frac{\int_{\lambda=380nm}^{780nm} \bar{y}(\lambda)T(\lambda)d\lambda}{\int_{\lambda=380nm}^{780nm} \bar{y}(\lambda)d\lambda}, \qquad (1)$$

where $\tau(\lambda)$ is the transmittance of the windows at wavelength λ. $\bar{y}(\lambda)$ is the photopic luminous efficiency of the human eyes defined by the CIE (International Commission on Illumination) standard. The wavelength range of 380 nm - 780 nm corresponds to the limits of human vision. The τ_{sol} is the integral transmittance under AM 1.5 solar irradiation and is given by

$$\tau_{sol} = \frac{\int_{\lambda=300nm}^{2500nm} AM_{1.5}(\lambda)T(\lambda)d\lambda}{\int_{\lambda=300nm}^{2500nm} AM_{1.5}(\lambda)d\lambda}, \qquad (2)$$

The solar modulation ability ($\Delta\tau_{sol}$) of a thermochromic window describes the solar transmittance between transparent ($\tau_{sol,t}$) and opaque ($\tau_{sol,o}$) states and it is calculated as $\Delta\tau_{sol} = \tau_{sol,t} - \tau_{sol,o}$. The tensile stress and strain were characterized by a tensile strength machine where a pulling force was applied to the HWC sample. The applied pulling force was recorded and the deformation of the HWC sample was measured to establish the properties including the stress, strain and Young's modulus. The Young's modulus of the samples was computed by the strain and stress in the elastic elongation region in the pulling process. For mechanical properties, the tensile stress and Young's modulus were examined 5 samples of the HWC and average values of the tensile stress and Young's Modulus was used to characterize its mechanical properties.

Figure 1. Fabrication process of HWC

RESULTS AND DISCUSSION

HWC undergoes a reversible phase transition between transparent and opaque states at a certain temperature. In order to measure the transition temperature, the transmittance at 550 nm against temperature of the HWC was measured. As shown in Figure 2a, the transmittance of the HWC decreased with the increase of temperature (orange solid line), and the transmittance increased back to the original value with the temperature decrease (blue solid line). And the transition temperature upon heating and cooling process appears at the minimum value of the transmittance derivative (lowest point of the orange and blue dash line). The average transition temperature of the HWC is around 23 °C which is the mean value of the transition temperatures of the heating process and cooling process. The transition temperature of HWC is suitable as a thermochromic smart window because the optical switch between 20 °C and 25 °C can result in maximal energy saving in HVAC systems. (Long & Ye, 2014; Warwick, Ridley, & Binions, 2014)

For the optical properties of the HWC, the τ_{lum} and $\Delta\tau_{sol}$ were characterized by an UV-Vis-NIR spectrometer. The optical properties, at the opaque state and transparent state, of the HWC were measured to study its thermochromic properties. The HWC samples at 20 °C were characterized for the transparent state luminous transmittance ($\tau_{lum,t}$) and solar transmittance ($\tau_{sol,t}$), and then the samples were heated to 40 °C for measuring the opaque state luminous transmittance ($\tau_{lum,o}$) and solar transmittance ($\tau_{sol,o}$). Figure 2b shows the transmittance spectrum, from 300 to 2500 nm, of the HWC at the opaque state and transparent state. It was found that the $\tau_{lum,t}$ of the HWC was 83% while its $\tau_{lum,o}$ was 40%. The high $\tau_{lum,t}$ illustrates a transparency of the HWC whereas the low $\tau_{lum,o}$ demonstrates opacity of the HWC as shown in Figure 2c. With the HWC at the transparent state, the letters on the paper can be clearly observed through the HWC, but at the opaque state, the letters cannot be seen through the HWC. The $\Delta\tau_{sol}$ is about 38%, and this high $\Delta\tau_{sol}$ implies the HWC can block the solar irradiance at the opaque state while allowing solar irradiance to transmit through the HWC at the transparent state to regulate the indoor air temperature. The significant optical contrast of HWC between transparent and opaque states can be attributed to the phase transition of the thermal-response hydrogel. When the ambient temperature is higher than the transition temperature, the thermal response hydrogel transfers from a swollen state to a shrunken state. This phenomenon leads to the significant change of refractive index in the microgels of hydrogel, and the strong light scattering causes the opacity of the HWC. It should be noted that the significant thermochromic effect was observed at the wavelength from 300-1400 nm, accounting for 89% of the solar irradiance. Therefore, the HWC has the strong solar modulation ability, showing high energy saving potential.

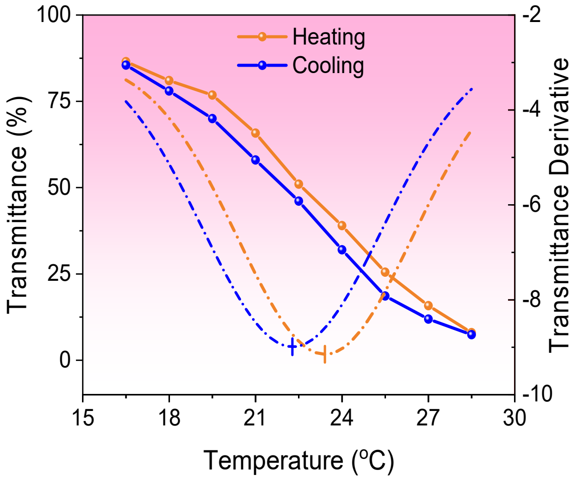

Figure 2. (a) Transmittance (solid lines) and transmittance derivative (dash lines) with function of temperature for determining the transition temperature.

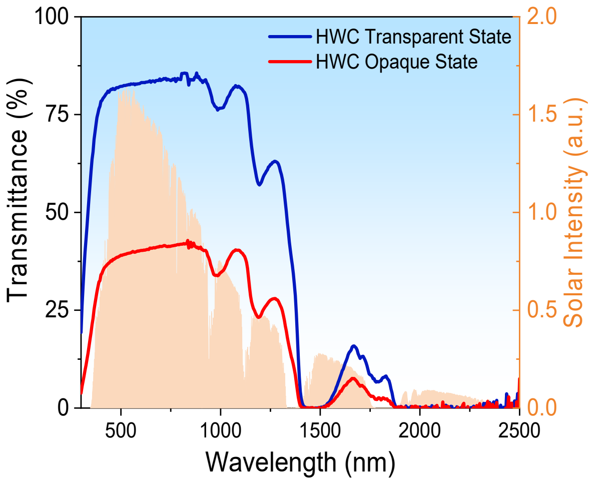

Figure 2. (b) Transmittance spectrum of the HWC at the transparent state and opaque state

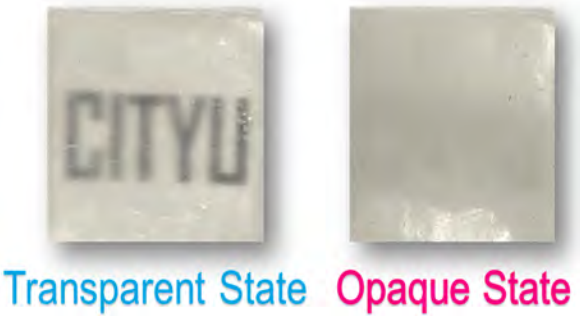

Figure 2. (c) Transparency and opacity of the HWC at the transparent state and opaque state.

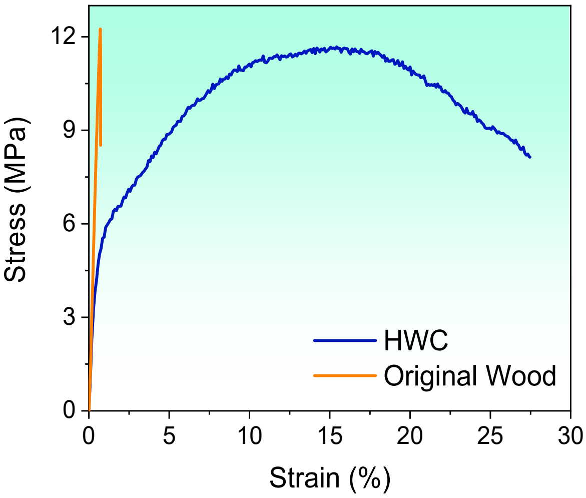

Figure 2. (d) Stress-strain curve of HWC and original wood.

Figure 2. (e) Flexibility of HWC.

Regarding the mechanical properties of the HWC, HWC demonstrates robust mechanical strength and high flexibility. In order to characterize its mechanical properties, the stress-strain profile of HWC was measured by a tensile test machine, and the result was compared with the original balsa wood. As shown in Figure 2d, it was found that the stress of the HWC was 11 MPa, which can be comparable to the original wood (stress of 12 MPa), implying that the fabrication process of the HWC, including delignification and impregnation, did not deteriorate the mechanical strength of the balsa wood. In addition, the HWC also inherits the merit of thermo-response hydrogel, which is strong toughness. The HWC demonstrates much higher breaking strain than that of the original wood. The breaking strain is the percentage of the elongation applying a pulling force when the material is failed. The breaking strain of the HWC is 14% while that of the original wood is only 0.7%. The breaking strain of the HWC is much higher than that of the original wood, so the HWC can result in a better shatter-proof effect than the original wood so that the potential risk when failure of the HWC can be reduced. Additionally, the Young's moduli, which is defined as the tensile stress divided by the tensile strain in the elastic deformation in the pulling process, of original wood and HWC were measured. The Young's moduli of the original wood and the HWC were 1700 MPa and 90 MPa, respectively. The Young's modulus of the original wood was almost 19 times of that of the HWC. The larger Young's modulus means that the material requires a larger force to be elongated. In other words, the HWC is relatively elastic and easy to be deformed and restored. Besides, the HWC is also soft and easy to be stretched, so it can be safely attached on the exiting windows. Furthermore, the HWC also exhibits excellent flexibility as shown in Figure 2e. It can be easily rolled and then recovers to the original shape without any damage, making it to be conveniently transported and installed. Therefore, in the real application, the HWC is easily attached to existing windows, so the retrofitting cost, installation cost and the maintenance cost of applying HWC to the current windows is low. Meanwhile, using the HWC can achieve the thermochromic effect to reduce the solar heat gain to the indoor environment and saving the energy of the HVAC system in buildings. So, in the following section, the energy saving performance of using HWC in buildings was determined numerically and is discussed.

HWC can smartly control the transmittance of solar radiation according to ambient temperature, therefore, it can effectively block the solar radiation to reduce the indoor air temperature in the hot weather. Conversely, the HWC can allow the solar radiation to pass through to heat the indoor environment in the cold weather. To quantitively study the energy saving of an HWC smart window applied in buildings, EnergyPlus, a simulation tool, was used to estimate the energy consumption in Hong Kong. A conventional double-glazed system, as a reference window, was created via the WINDOW algorithm developed by the Lawrence Berkeley National Laboratory. HWC was attached on the double-glazing system and named as HWC smart window. The τ_{sol} and τ_{lum} of the conventional window were 60% and 78% as the input parameters to the EnergyPlus simulation. The optical properties, such as the τ_{lum} and τ_{sol} of the HWC window investigated in "Characterization of the HWC" section, were employed in the EnergyPlus simulation. In the EnergyPlus simulation, a 12-floor large office reference building (Building information is described in Table 1) established by U.S. Department of Energy was used to evaluate the energy consumption using the conventional window and the HWC smart window (DOE, 2015).

Table 1. Information of the building used in the EnergyPlus simulation

Building type	12-floor office building
Length × Width	73.2 m × 48.8 m
Window Fraction (Window-to-Wall Ratio)	40%
Thermostat setpoint for HVAC system	23.9 ^{o}C cooling/21.1 ^{o}C heating
Lighting setpoint	500 lux

In the simulation, four aspects of the energy consumed in a year in buildings were analysed such as cooling, heating, lighting and fans/ pumps. Figure 3 shows that the energy consumption of cooling is the major sector in buildings, but using the HWC smart window, the energy consumption of cooling was 7% smaller than that using the conventional window. Regarding the energy consumption of fans/ pumps in buildings, 14% of energy can be saved using the HWC smart window compared to using the conventional window. The energy saving in cooling and fans/ pumps using the HWC is due to the indoor air temperature reduction to achieve thermal comfort, so that the energy consumption of air conditioning system can be significantly reduced comparing with using conventional windows. It should be noted that owing to the solar modulation ability, a part of the visible light is reflected by the HWC smart window. Therefore, in order to maintain the luminance of the indoor environment, the energy consumption of lighting using the HWC smart window was slightly larger than that of using the conventional window. Overall, the energy consumption using the HWC was 7% smaller than that using the conventional window. These analysis prove that employing the HWC smart windows in buildings is an effective way to save energy.

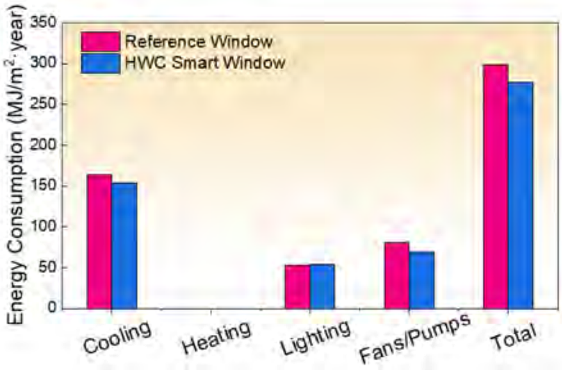

Figure 3. Energy consumption per year in different aspects in buildings equipped with the HWC and conventional window simulated by EnergyPlus.

CONCLUSIONS

In this study, a flexible transparent hydrogel wood with robust mechanical strength and smart optical regulation ability is developed. The HWC demonstrates high transparency with T_{lum} of 83% in the transparent state, high opacity with T_{lum} of 40% in the opaque state, and high solar modulation ability of ΔT_{sol} = 38%. The transition temperature of HWC was 23 ^{o}C, demonstrating great potential in real applications. The HWC also exhibits the tensile strength that can be comparable with original wood. With respect to the Young's modulus of the HWC, it was about 90 MPa where the Young's modulus of the original wood was 20 times of that of the HWC. Therefore, the HWC is highly flexible, making it conveniently to be used on the existing windows. In addition, the energy saving potential of HWC was successfully proven by the EnergyPlus simulation and 7% of total energy in buildings can be saved. Notably, HWC was made by renewable wood as well as non-toxic hydrogel, which is friendly to the environment and safe for the users. Overall, HWC has potential to be a new generation material for smart windows applied in energy-efficient buildings in the future.

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