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Solvent Operated Polydimethylsiloxane (PDMS) Sponge Coatings for Tunable Transmittance Smart Windows

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Abstract

Heating and cooling systems account for approximately 30 % of the world's total energy consumption. Various smart window designs with adjustable optical properties have been investigated to control light and regulate heat transfer in buildings. Yet, the high production costs and complex fabrication methods of existing technologies limit their widespread application. Here, we present a sustainable and scalable approach using polydimethylsiloxane (PDMS) sponges and coatings enabling tunable light transmittance. The tunability is achieved through silicone oil infusion and release. The fabrication process includes only PDMS and granulated sugar as a template, providing a simple and environmentally friendly method. The PDMS sponges were applied to glass surfaces, and their stability and light-regulating performance were evaluated. The coated glass surfaces exhibited an 84.6% solar light blocking and a 4.40 ± 1.05 °C difference between the front and back surfaces in the opaque state, compared to 15.2% and 0.8 ± 0.2 °C in the transparent state. They maintained structural integrity after 6 months of exposure to silicone oil and 200 cycles of mechanical abrasion with a 20 g weight. Overall, this low-cost, scalable, and durable coating has significant potential for smart window applications to enhance building energy efficiency.

Introduction

Global energy consumption continues to rise each year, due to technological developments and growing global urbanization. Currently, 30% of global energy consumption is used for heating and cooling buildings, so reducing this energy is vital for sustainability. Windows are primary sites of heat exchange in buildings.[1,2] During the summer, sunlight entering through windows raises indoor temperatures, while in winter, over 30% of heat is lost through windows. Cooling/heating systems consume energy and generate greenhouse gases, further contributing to global warming.[3]

Radiative cooling has emerged as a promising technology to enhance energy efficiency in summer. This approach passively releases heat to the environment while reflecting incoming solar radiation to maintain lower indoor temperatures. Achieving high solar reflectance typically requires advanced nanoporous or microporous materials, often produced using nanoparticles. [4,5] These methods are expensive and complex, so simple, cost-effective, and sustainable alternatives are needed.[6]

Researchers have explored materials with tunable transparency for smart windows, including electrochromic, mechanochromic, thermochromic, photochromic, and liquid crystal-dispersed polymer systems. Electrochromic materials, change their optical properties in response to an electric field, enabling dynamic light control. However, their inherent toxicity, handling requirements,[7,8] sensitivity to undesired reactions, low durability, and high production costs, restrict their practical application.[9–11] Thermochromic materials, such as vanadium dioxide (VO_2), titanium dioxide (TiO_2), and zinc oxide (ZnO), often combined with organic dyes, can respond to temperature changes. VO_2 is one of the most promising materials due to its excellent optical and electrical properties, but its high transition temperature (~68°C), operational complexity, and limited light-regulated performance (22.3% at 90°C) constrain its widespread use.

Thermoresponsive hydrogels such as poly(*N*-isopropylacrylamide) (PNIPAm), can modulate light near their lower critical solution temperature (LCST).15,16 However, their rapid opacity change near room temperature and[16] low mechanical strength pose significant limitations for practical applications. Recent combinations of thermoresponsive hydrogels, vanadium dioxide, and liquid crystals enable dynamic control[17,18] , but they still face challenges with cost, durability, and scalability. In parallel, the refractive index matching method has been widely studied in the

literature to control optical properties [19–23] in gels [24], glass fibers [25], and inorganic particle-containing films, [26] yet its direct application to glass for smart windows remains limited.

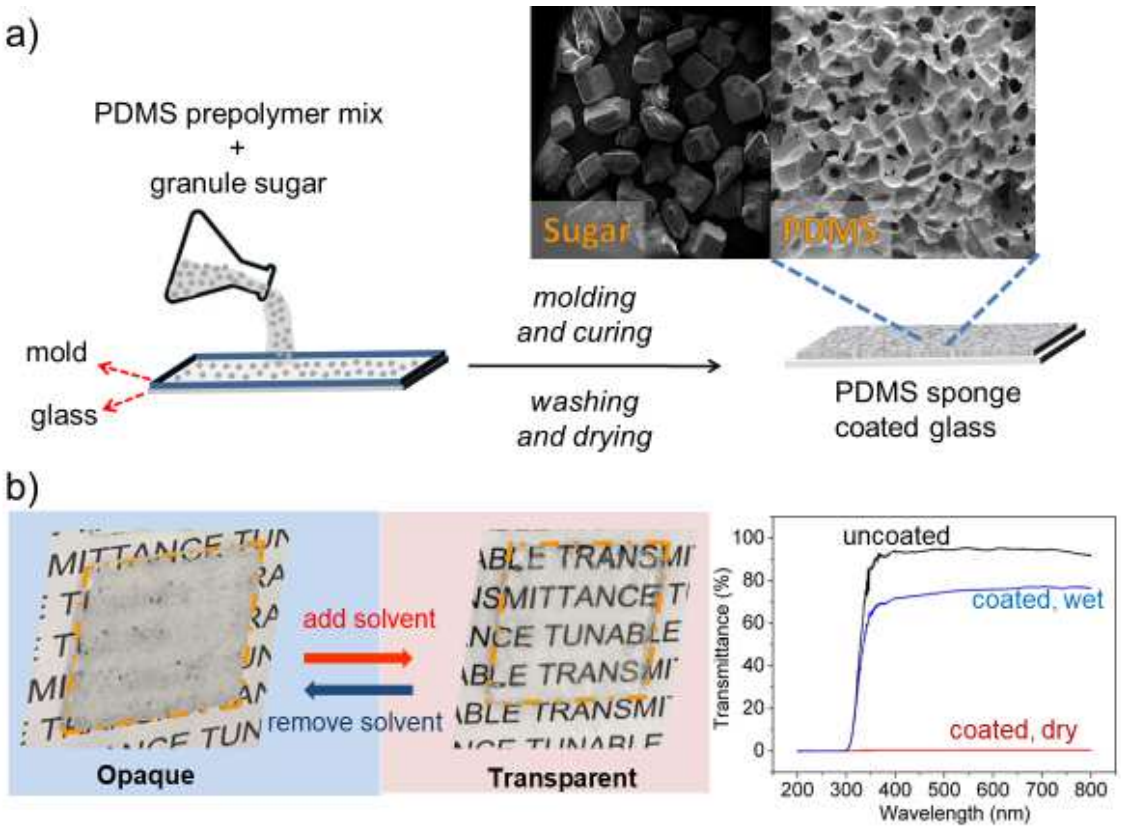


Figure 1. a) The preparation of PDMS sponge-coated glass b) Reversible opaque and transparent states of the PDMS sponge-coated glass exhibiting reversible opaque and transparent states controlled by silicone oil infusion and release, as shown by UV-vis spectra of the uncoated and coated glass on the right. The glass substrate supporting the PDMS sponge is encircled in orange dashed lines as a visual guide.

In this study, we present a sustainable and scalable approach for developing polydimethylsiloxane (PDMS) sponge coatings on glass to enable tunable light transmittance and reduce building energy consumption. Unlike conventional methods relying on hazardous chemicals, our fabrication uses only granulated sugar -as a template- and PDMS (Fig. 1a), providing a more sustainable, environmentally friendly, and cost-effective approach. Based on refractive-index matching, the coatings exhibit dual functionality: in their opaque state, they efficiently block infrared light, thereby reducing cooling energy demands. Upon absorbing silicone oil, the sponges become transparent to visible light, supporting decreased energy consumption for heating (Fig. 1b). To our knowledge, this work presents the first detailed optimization of the sponge-based system and its

application on glass surfaces, demonstrating tunable optical performance, solar light and heat blocking capabilities, and mechanical and chemical resistance. This platform provides a simple solution for smart windows, enabling an integrated functionality not previously achieved.

Experimental

Materials and Methods

Polydimethylsiloxane (PDMS) Sylgard 184 (Dowsil), silicone oil (500 cSt), hexanol (98%), dimethylformamide (≥ 99.8%), ethylene glycol (≥ 99%), and ethanol (≥ 99.9%) were obtained from Sigma-Aldrich. Granulated sugar was purchased from a local market.

The transmittance of the sponges was measured using a UV-Vis spectrometer in the wavelength range of 400 – 800 nm. For the transparent state, samples were immersed in silicone oil within a cuvette, and spectra were recorded with identical alignment to ensure consistent measurements.

The thermal performance of PDMS sponge-coated glass surfaces was evaluated using an infrared (IR) lamp (220 W). Front and back surface temperatures were measured using Fluke TİX560 after 6 min of IR lamp exposure, with samples 55 cm away from the IR source.

Scanning electron microscopy was conducted using an FEI Quanta 200 FEG at an accelerating voltage of 10 kV and a spot size of 3.0. Samples were coated with Au-Pd using a Precision Etching Coating System (PECSTM). Mechanical compression tests were conducted on cylindrical samples using TA Instruments - Water LLC-Q Series Dynamic Mechanical Analyzer with a ramp force rate of 2.0 N/min. UV light illumination was provided by a CAMAG light source (366 nm, 8 W) and solar radiation simulated using a Xe lamp (100 mW cm^{-2} with an AM 1.5 filter).

Fabrication of PDMS Sponges:

Sylgard 184, a commercially available kit consisting of a linear PDMS prepolymer with terminal vinyl groups and a curing agent, was used to prepare the sponges. Crosslinking relies on a hydrosilylation reaction between the vinyl groups ($SiCH=CH_2$) of the PDMS prepolymer and the hydrosilane groups (Si-H) in the curing agent. [27,28] A porous sponge structure can be obtained using granulated sugar as a removable template. During curing, PDMS forms an elastic network, while the sugar particles determine the pore size and structure. After curing, the sugar template is

removed by washing with water, yielding lightweight, flexible, and mechanically robust porous sponges.

We systematically investigated the key parameters- including PDMS: curing agent: granulated sugar weight ratio and sugar granule sizes and sponge thickness- to achieve the lowest light transmittance in their opaque form, highest transmittance after liquid absorption, low swelling degree, and high mechanical strength (The detailed optimization is provided in Supplementary Information). To fabricate the sponges, PDMS prepolymer, curing agent, and granule sugar were mixed and poured into a silicone mold. Air bubbles were removed under vacuum for 15 min, and the samples were cured at 80°C for 3 hours. The cured sponges were washed in an ultrasonic bath with water and dried at 60°C to obtain a porous sponge structure.

Application of PDMS Sponges to Glass Surfaces:

To prepare PDMS sponge-coated glass, a mixture of PDMS prepolymer, curing agent, and granulated sugar (in a 10:1:4 weight ratio) was poured into the mold (3D-printed, 20 mm × 20 mm × 5 mm) fixed onto 22 mm × 22 mm glass substrates, and flattened to achieve uniform thickness. The samples were degassed under vacuum and cured at 80°C for 3 hours. After curing, the glass substrates were immersed in an ultrasonic water bath to remove the sugar template, and subsequently dried in an oven.

Results

Fabrication and UV-Vis Light Transmission of the PDMS Sponges:

To optimize the optical and mechanical performance of PDMS sponge and its coating, key fabrication parameters were systematically investigated. PDMS sponges were prepared with PDMS-to-sugar weight ratios (1:2, 1:4, 1:6, 1:8), and their morphology was analyzed by SEM. The 1:4 ratio provides the most uniform and well-distributed pores. Mechanical properties and swelling behavior were assessed for PDMS-to-curing agent ratios 40:1, 20:1, 10:1, and 5:1, with the 10:1 ratio offering the best balance between stiffness and swelling. Based on these findings, the PDMS: granulated sugar ratio of 1:4 and the PDMS : curing agent ratio of 10:1 were selected as the optimal conditions.

The optical performance of PDMS sponges is strongly influenced by microstructure and thickness.[29–31] While the coatings from nanometers to millimeters can be prepared,[32–35] thicker coatings increase absorption or scattering, reducing transmittance.[36–38] To investigate and optimize optimal performance, PDMS sponges were fabricated using sugar granules with a size range of 850–500 µm, 500–200 µm, and 200–30 µm, and thicknesses of 0.5 mm, 1.0 mm, and 2.0 mm. UV-Vis measurements showed that (Fig. 2), for all thickness, values, larger pores resulted in higher transmittance (~7 % for 850–500 µm), while 500-200 µm and 200-30 µm pores maintained similarly low transmittance (~0.2 %) due to increasing light scattering. Among these two low-transmittance structures, silicone oil penetration was determining factor: smaller pores and thicker samples hindered oil penetration. Therefore, a pore size 500 – 200 µm film thickness 0.5 mm were selected as the optimal, ensuring low transmittance in the opaque state while enabling efficient liquid infusion.

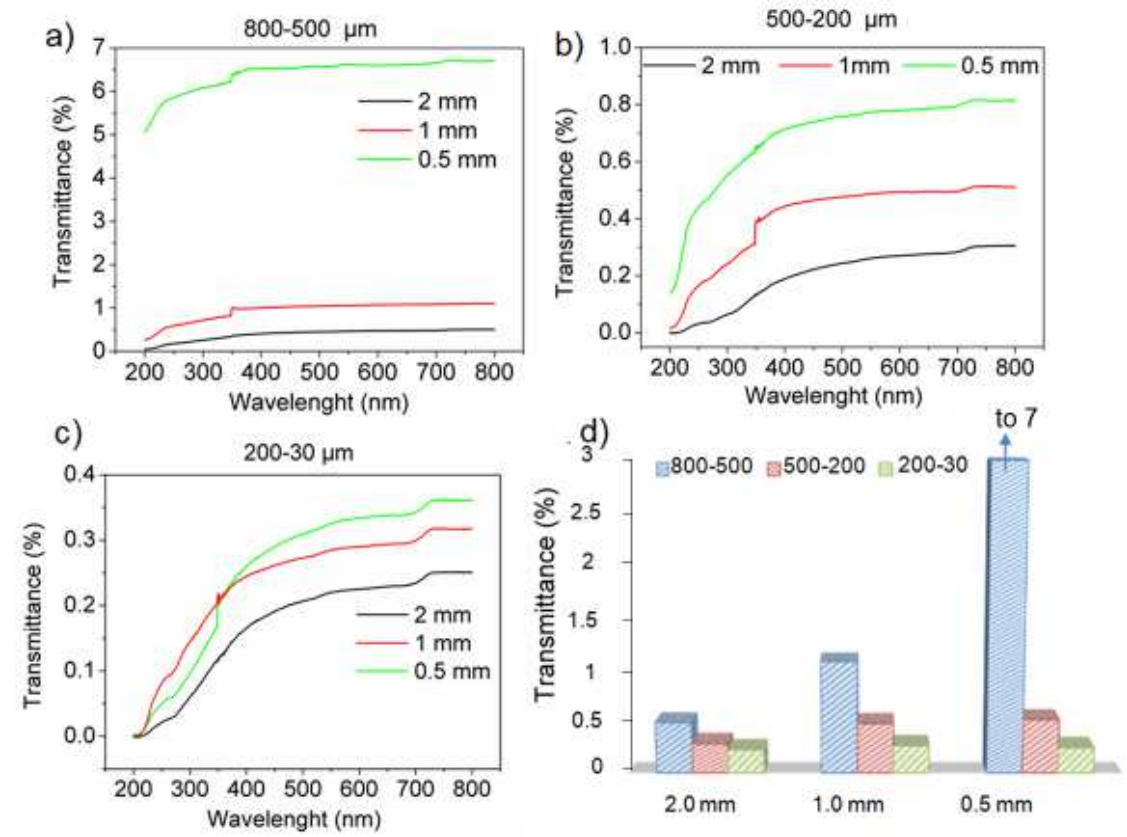


Figure 2. Light transmission control with sample thickness and porosity. UV-vis spectra of 2.0 mm, 1.0 mm and 0.5 mm thick dry PDMS sponges with pore size ranges of 800-500 µm, 500-200 µm, 200-30 µm show; d) with thicker samples of smaller pore sizes the transmittance is lower, 800 nm transmittance values from a-c are utilized. (The abnormality in the spectra at 350 nm is due to a light source change at this wavelength.)

In porous transparent materials, the refractive-index difference between the material and the air pockets causes scattered light, making the material appear opaque. For PDMS sponges, this difference can be calculated as: $\Delta n = n_{PDMS} - n_{air} = 1.42 - 1.0 = 0.42$. Light scattering-and the resulting low transmittance- can be reversed by filling the pores with a medium of similar refractive index to PDMS.[39]. We tested solvents including silicone oil (η:1.40; viscosity: 100 mPas), 1-hexanol (η:1.416),[40,41] dimethylformamide (η: 1.428), ethylene glycol (η:1.431), and ethanol (η: 1.361)[42], considering both their refractive indices and the induced swelling degree. Hexanol achieved nearly 100% transmittance (Fig. 3b), consistent with its minimal refractive index difference from PDMS. It also caused modest swelling (~10%), suggesting its suitability as solvent. However, its potential skin irradiation[43] and strong odor limit practical use. Silicone oil, which poses no health risk and provides 86% light transmittance, emerged as the most suitable alternative. The sponges exhibited a moderate swelling ratio of 27 % in silicone oil. Overall, silicone oil provides an optimal balance between high transparency, controlled swelling, and safety. Therefore, it was selected for the remaining experiments, which demonstrate the application of PDMS sponges as a tunable glass coating.

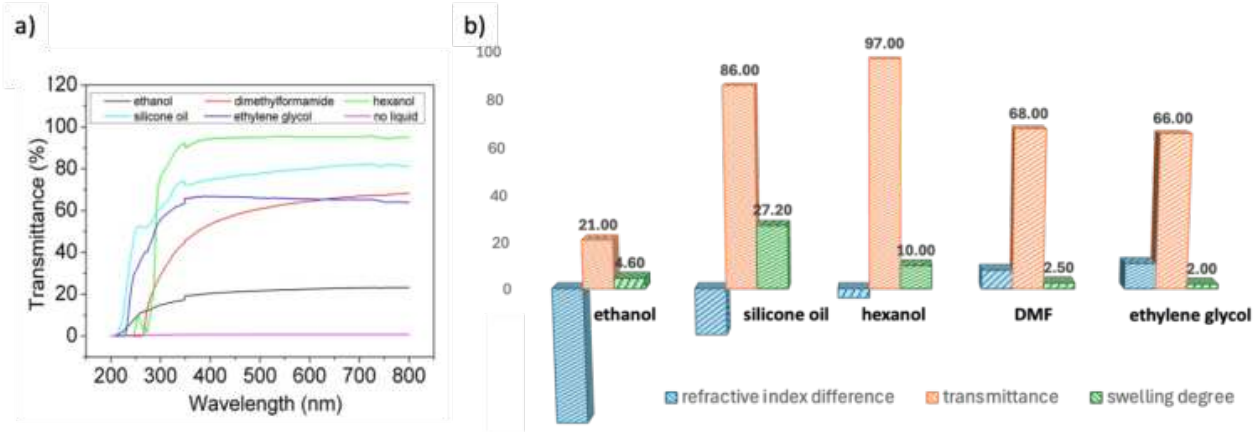


Figure 3. a) UV-vis spectra of PDMS sponges dry and soaked with ethanol, silicone oil, hexanol, DMF, and ethylene glycol. b) The solvents with small PDMS-solvent refractive index difference values yield to higher percent transmittance values as expected. Hexanol is the best solvent among the listed solvents, resulting in the highest transmittance and a low degree of swelling in the material. Silicone oil has the second-highest transmittance with a moderate swelling ratio. The data was acquired using 0.5 mm thickness, 500-200 μm pore size PDMS sponges. The displayed bars for refractive index difference values are for comparison - not to scale with the given axis numbers.

Fabrication of the PDMS sponge-coated glass surfaces and assessment of their tunable transmittance performance:

The simple fabrication of PDMS sponges and their tunable transmittance through silicone oil motivated the development of sponge-coated glass. Glass substrates were coated using the optimized formulation (PDMS:curing agent:granulated sugar = 10:1:4 w/w/w) containing sugar particles with a size range of 500–200 μm. The film thickness was measured using a micrometer at five different locations on glass substrates, both before and after coating. The final coating thickness was calculated as the average of these measurements. Using this method, we prepared coatings with a thickness of 0.57 mm ± 0.10 mm and subjected the coated glass to optical transmittance and mechanical testing, as described below.

Coating a glass slide with the PDMS sponge resulted in an opaque appearance similar to sponge films. Infusion of silicone oil ($n = 1.40$) into the pores, minimized the refractive index difference ($\Delta n = 0.02$), decreasing light scattering. The coating could reversibly switch between the opaque and transparent states through repeated silicone oil infusion and release (Fig. 1).

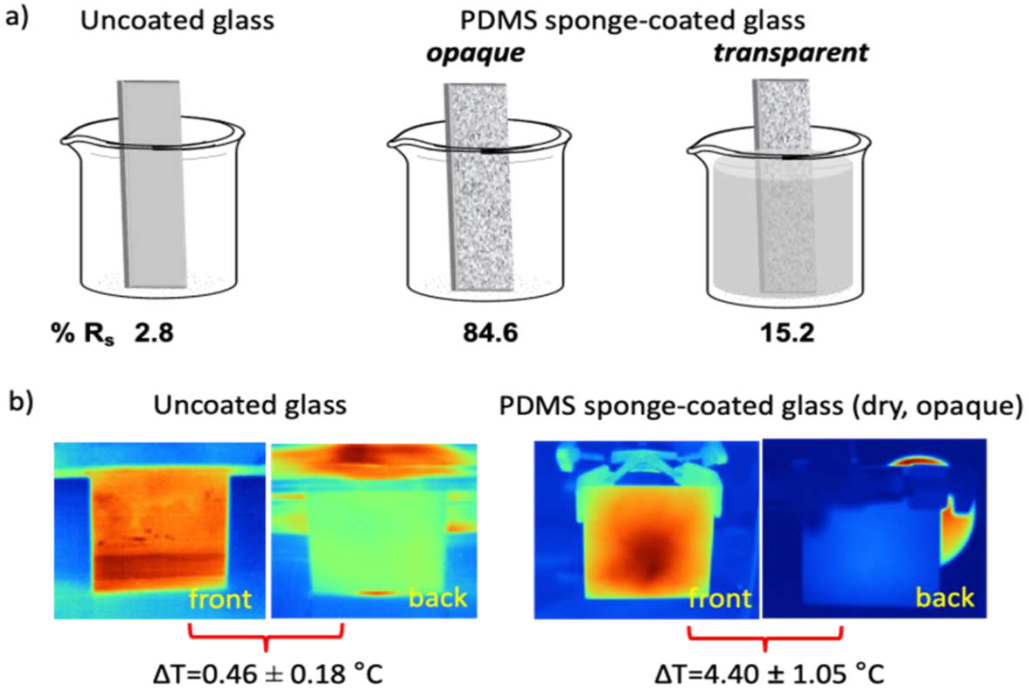


Figure 4. PDMS coatings blocking solar light and heat. a) The solar light blocking ($\%R_s$) values show that the transparent state of PDMS sponge-coated glass behaves similarly to uncoated glass, whereas the solvent-soaked opaque state blocks the solar light. (Solar simulator placed at 30 cm to the samples and a

light meter was for obtaining the transmission values used in Equation 1 to calculate the values shown. b) Infrared images of the front at back sides of the IR-illuminated samples clearly demonstrate the efficient heat blocking in dry PDMS sponge coatings. (The samples were illuminated for 6 min from 55 cm with an IR lamp.)

Evaluating the solar light transmission of PDMS sponges is crucial to understanding their potential for dynamic daylight shading in building windows. To assess this, PDMS sponge-coated glass samples were placed in a beaker and positioned at 30 cm from a solar simulator (380 nm to 705 nm) (Fig. 4a). Their light transmittance was measured by using a light meter (similar to a test reported in a previous study,[44] at the two states: the initial opaque state (U_{dry}) and the transparent state after immersion in silicone oil (U_{wet}). The light blocking performance (R_s) was calculated using Equation 1,

$$R_s = \frac{(U_{dry}, U_{wet}, or\ U_{glass}) - U_{setup}}{U_{setup}} \quad (1)$$

where U_{setup} is the transmittance value for the beaker in which the coated and uncoated glass samples are positioned. The opaque and transparent states blocked the light 84.6% and 15.2%, respectively (uncoated glass: 2.8%).

The thermal insulation was evaluated by measuring the temperature difference (ΔT) between the heat-exposed (front) and back surfaces (Fig. 4b) under IR irradiation at 55 cm. After 6 min, the opaque state showed ΔT: 4.40 ± 1.05 °C, whereas the transparent state was only 0.83 ± 0.15 °C, similar to uncoated glass, 0.46 ± 0.18 °C. (The ΔT values for the initial, before-irradiation states were less than 0.8 °C for all samples.) These results indicate that the transparent state behaves similarly to uncoated glass, enabling daylight and heat transmission, which is beneficial on cold days. In contrast, the opaque state efficiently blocks solar light and heat, making it suitable for reducing indoor heating.

To ensure reliable coating performance, strong adhesion to the substrate is essential. We evaluated mechanical robustness using a simple, cost-effective sandpaper abrasion test.[45] For this test, coated glass samples were placed face-down on 600-grit sandpaper under a 20 g load, and rubbed back and forth over a 10 cm path for 200 cycles. After the test, no significant deformation, damage, or delamination was observed, and the initial sample thickness was retained (Fig. 5a-b). The images before and after abrasion were captured using a digital camera to show the overall

appearance of the surface, rather than the microstructure. The chemical stability of PDMS sponge coatings was evaluated by immersion in strong acidic (pH 1) and alkaline (pH 13)

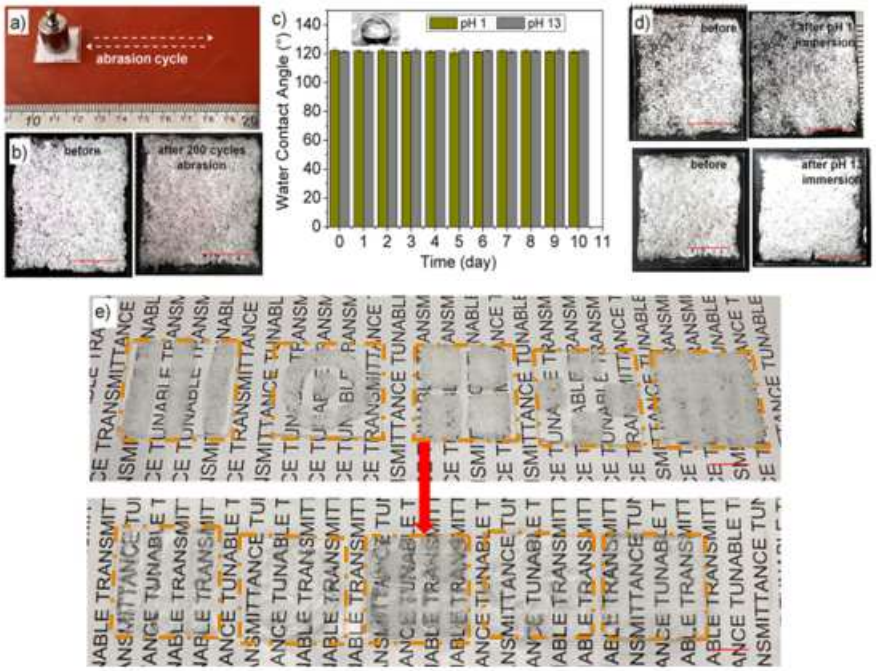


Figure 5. a-b) Mechanical abrasion test for 200 cycles on a sandpaper with a normal weight of 20 g show no visible damage of the sponge coating on glass. c) Contact angle measurement of coating during immersion in pH 1 and pH 13 and d) Images of coating before and after immersion e) PDMS sponge coatings of different shapes display reversible opaque (top) and transparent (bottom) states attained by the liquid absorption-release cycle. Scale bar = 1.0 cm. The glass substrate supporting the PDMS sponge is encircled in orange dashed lines as a visual guide.

The system operates in the presence of silicone oil, and we evaluate long-term performance on sponge-coated glass surfaces. The coated samples were immersed in silicone oil for 1-6 months, with weekly removal, drying, and re-immersion. No surface deformation or deterioration was observed, indicating excellent stability. The performance retention was also achieved with customized patterns - the sponge system can be applied with a total match of geometry and shape of the substrate, enabling a straightforward application to both large and small surfaces with all types of geometries (Fig. 5e).

Performance comparison with similar state-of-the-art systems:

A research group developed a silicone film capable of reversible transitions between porous and transparent states.[46] When combined with solar-absorbing carbon black particles (CBPs) in a bilayer configuration, the material exhibited 95% sunlight absorption in the transparent state and 93% solar reflection with 94% long-wave infrared emission in the porous state. This dual functionality enables passive cooling and heating without energy consumption. However, the need for mechanical modulation restricts its practical application in smart windows.

Another study utilized thermochromic (pNIPAm-AEMA) microparticles that were synthesized and placed between two glass slides.[47] These films exhibited high visible-light transmittance (87.2%) and enabled indoor cooling (~5 °C) driven by a phase transition around 32 °C. However, their temperature-dependent switching and complex fabrication pose a challenge in real-world applications, making them less effective in unstable climates. VO_2–SiO_2 composite films fabricated both luminous transmittance (48.5%) and solar modulation (15.7%).[48] However, switching temperature (~68 °C), limited solar modulation, reduces their practical efficiency.

The PDMS sponge/solvent system offers several key advantages over the materials described above. 1) PDMS sponge-coated glass can effectively control solar transmission and light modulation by switching between opaque and transparent states. 2) This transition between the states works without any need for an external temperature change, offering more consistent performance of environmental conditions compared to temperature-sensitive systems like pNIPAm-AEMA or VO_2-based materials. 3) The PDMS sponge/solvent coatings are relatively more straightforward to fabricate compared to the complex synthesis processes of pNIPAm-AEMA particles or the manufacturing challenges associated with VO2 films. 4) The described system is made up of an easy-to-find and relatively cheap polydimethylsiloxane, sugar, and silicone oil, reducing production cost. 5) The use of silicone-based materials and the application geometry that is well-tolerated for different shapes and geometries also makes the system applicable to large-scale. 6) PDMS sponge coatings have high mechanical durability and long-term stability, making them resistant to environmental degradation. 7) The material can be adapted to the current double-glazed windows. The system also has some disadvantages: in the light-reflecting (passive cooling) state, it is opaque, limiting visibility. Another drawback may be the need to use external pumps to inject and withdraw solvent from the system. Nevertheless, the

coating can be applied to windows where these features can be tolerated, such as on building windows where full-time transparency is not required.

Conclusions

The PDMS sponges can be produced through a simple and low-cost process, offering an attractive alternative to current smart windows. Here, we demonstrate a sponge-coated glass that blocks sunlight by 84.6% in its opaque state while maintaining high transparency. This reversible transition is driven by simple solvent absorption/release cycles. In addition, coatings exhibit high mechanical and chemical durability, highlighting their strong potential for smart window applications.

Author contributions

E.M.; conceptualization, methodology development, data curation, investigation, supervision, visualization, and writing of both the original draft and revised manuscript. H.I.O.; conceptualization, supervision, and writing (original draft and review & editing). B.B.; conceptualization, supervision, and writing (original draft and review & editing).

Conflicts of interest

There are no conflicts to declare.

Data availability

Data supporting this article have been included as part of the ESI,† with original data available on request from the corresponding authors.

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