Supplementary Information for

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

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Optimization of Fabrication Parameters for Porous PDMS Sponge

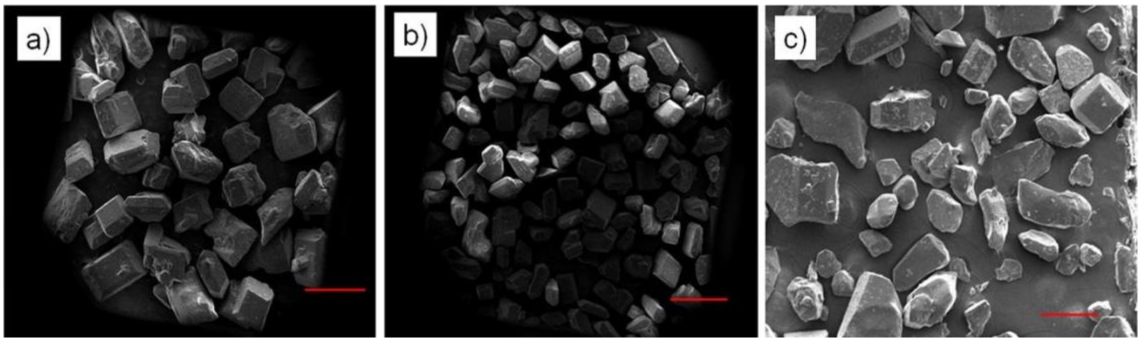

Figure S1. SEM images of granulated sugar from left to right 850–500 μm, 500–200 μm, and 200–30 μm. (Scale bar is 1 mm)

Commercial granulated sugar was crushed using mortar and sieved into particle size ranges (850–500 μm, 500–200 μm, and 200–30 μm). Scanning Electron Microscopy (SEM) images (see, Fig. S1) confirmed that the sugar particles achieved desired size ranges.

Granulated sugar with size ranges 850–500 μm, 500–200 μm, and 200–30 μm was used as a porogen for fabrication of PDMS sponges. The amount of sugar plays a critical role in determining pore size and morphology of the sponges. PDMS sponges were prepared using varying PDMS to sugar weight ratios, and SEM images were obtained to evaluate the pore size, pore distribution, and homogeneity (Fig. S2). At a 1:2 ratio, sugar templated pores averaged 0.46 ± 0.1 mm, accounting for ~60% of all the pores, while spontaneous pores averaged 0.26 ± 0.1 mm. This ratio resulted in sparsely distributed, smaller pores, consistent with the literature (2013) at a similar ratio of 1:1.8. [1] At 1:4 ratio, sugar templated pores measuring 0.38 ± 0.1 mm and non-templated pores 0.24 ± 0.1 mm, resulting in balanced and homogeneous porous structure. [2]At a ratio of 1:6, sugar-templated pores (0.39 ± 0.1 mm) comprises only 33% of total, indicating less controlled pore forming. At a ratio of 1:8, sugar-templated pores 0.33 ± 0.1 mm and only 32% of the total pores, while the average size of other pores was 0.4 ± 0.2 mm. This excessive sugar content compromised the structural integrity of the sponge due to oversized and non-uniform pores. These findings are consistent with previous report indicating that excessive sugar leads to irregular and non-uniform pore structures. Overall, a 1:4 PDMS-to-sugar ratio was identified as optimal, well-distributed porous network that closely replicates the sugar particle dimension.

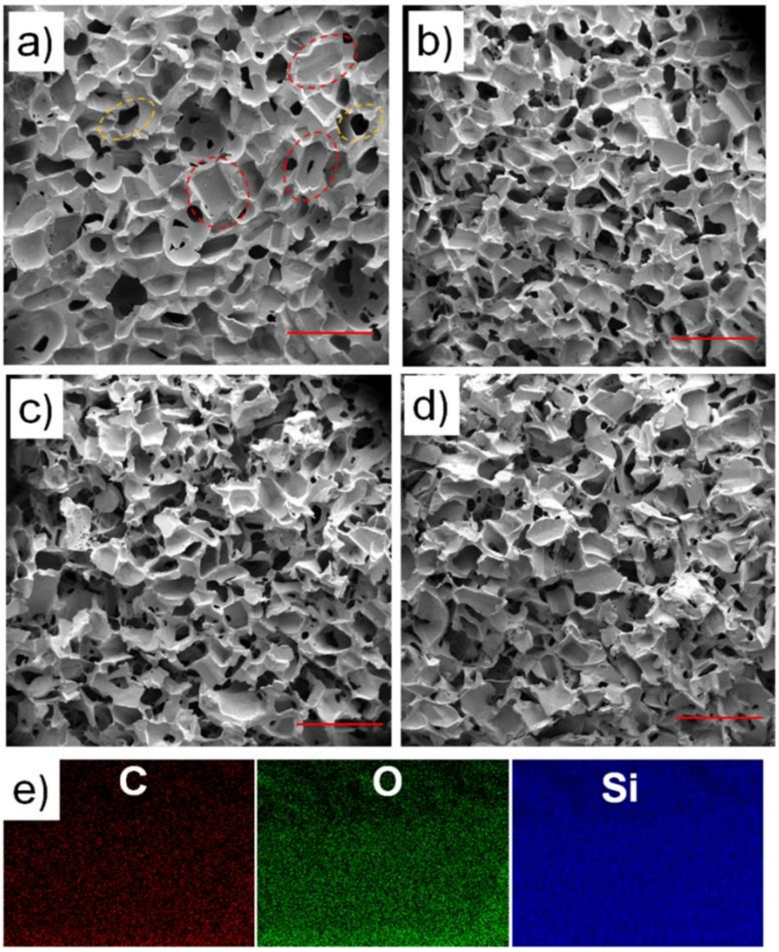

Figure S2. SEM micrograph of PDMS sponge prepared with a) 1:2, b) 1:4, c)1:6, d)1:8 PDMS:granule sugar weight ratio, Scale bar 1 mm, Sugar template pores are indicated by the red dashed lines, while the other pores are highlighted with yellow dashed lines e)EDX maps of a PDMS sponge.

The mechanical strength of the PDMS sponges is strongly influenced by pore size and PDMS to curing agent weight ratio. To investigate the effect of crosslinking density, sponges were prepared at 40:1, 20:1, 10:1, and 5:1 PDMS polymer-to-curing agent weight ratios Compression tests (Fig. S3b) revealed clear correlation between the PDMS polymer-to-curing agent ratio and sponge stiffness; [3,4]: the 40:1 ratio exhibited the lowest Young's modulus (6.6

± 0.3 MPa), producing a soft, flexible material with the lowest crosslinking density. At the 20:1 ratio and 10:1 ratios, the modulus increased from 16.3 ± 0.5 MPa to 51.4 ± 1.6 MPa. The 5:1 ratio exhibited the highest stiffness (58 ± 1.4 MPa), reflecting a densely crosslinked polymer network. These findings confirm that higher curing agent concentrations increase crosslinking density, restrict polymer chain mobility and enhancing the mechanical integrity of the sponges.

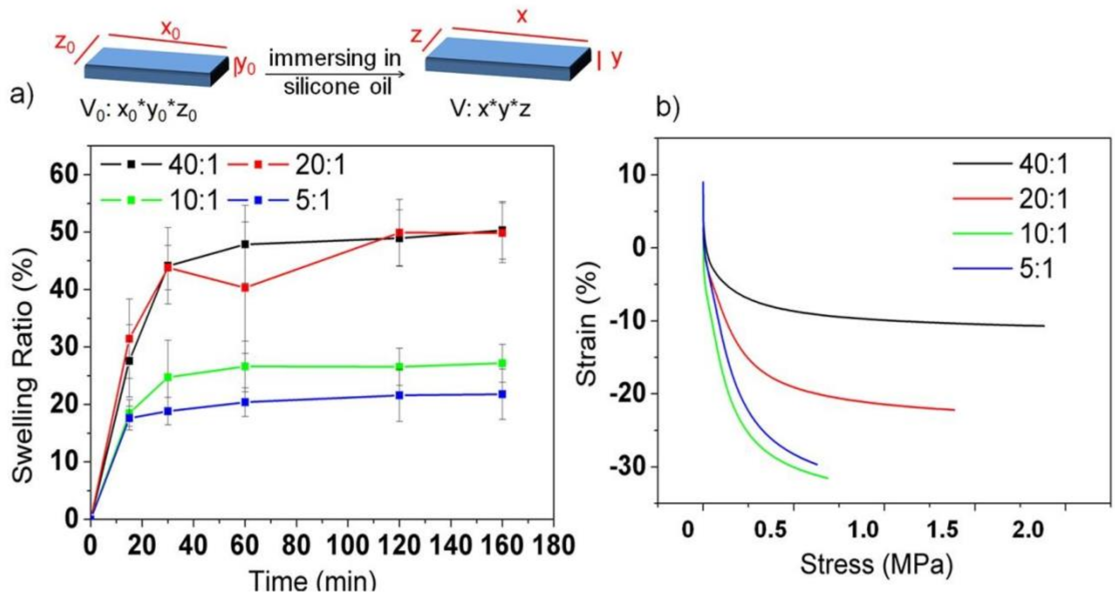

Figure S3. a) Time dependent swelling ratio and b) strain–stress curve of sponges of PDMS polymer-curing agent weight ratios (40:1, 20:1, 10:1, and 5:1)

The swelling behavior of PDMS sponges prepared at PDMS: curing agent ratios of 40:1, 20:1, 10:1, and 5:1 was evaluated by immersing samples in silicone oil. The swelling ratio (% increase in volume) was calculated using the Equation 1:

$$\%\ swelling\ ratio = \frac{V-V_0}{V_0} \times 100 \quad (1)$$

where V is the swollen volume, and V_0 is the initial dry volume of the sponge. Each measurement was conducted in triplicate, and the results were given as mean values with standard deviations.

The swelling ratio decreased with increasing curing agent content, reflecting higher crosslinking density (Fig. S3a). The 40:1 and 20:1 showed the highest swelling (~%50), while 10:1 and 5:1 ratio exhibited reduced swelling (%27 ± 3 and %22 ± 4, respectively). Combined with compression tests, the 10:1 ratio was selected as optimal providing a balance between mechanical stiffness and moderate swelling, ensuring dimensional stability and suitability for durable applications.

Development of PDMS Sponge-Coated Window Prototype

To demonstrate the practical functionality of the developed system, a prototype window was fabricated (Fig.S4). The device allows switching between opaque and transparent states using a single pump as the control mechanism. The prototype consists of double-glass window incorporating a PDMS sponge-coated glass. Initially, the coated glass and when the pump is run, silicone oil is introduced into the window, filling the pores and glass becomes transparent. To return to the opaque state, the silicone oil is released through the outlet at the bottom resulting in the sponge-coated glass opaque again.

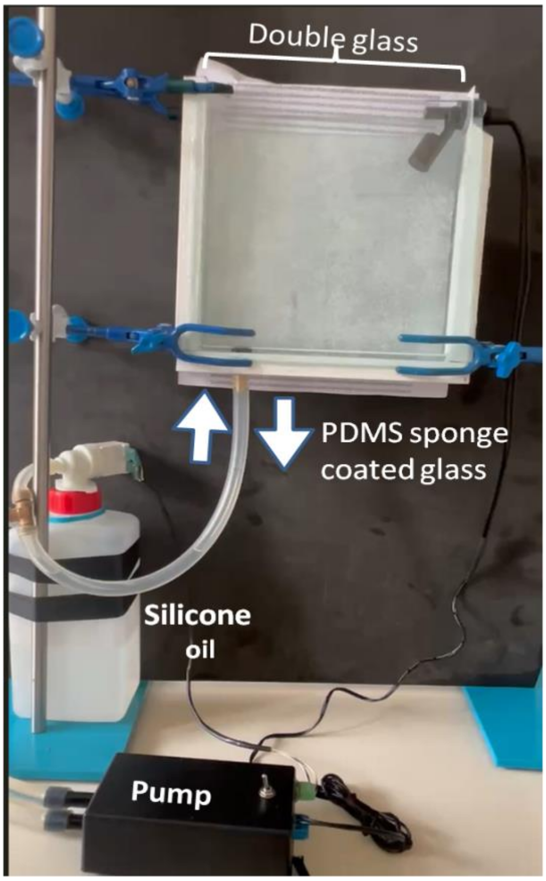

Figure S4. Prototype smart window with PDMS sponge-coated glass

References

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