Supporting Information

On-demand Photo-controlled Motion Enabled by Solvent-driven mesogen alignment Switch

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Figure S1. SEM and corresponding optical images of the fabrication process for Janus AZOIOs-PC side. (A) SiO_2 opal template, (B) composite opal (the interstice of SiO_2 opal template was infiltrated by LC precursor) and (C) PC side of Janus AZOIOs membrane.



Figure S2. SEM images of (A, A_1) PC side and (B, B_1) polymer side.



Figure S3. (A) Thermogravimetric analysis of the Janus AZOIOs membrane. (B) Differential scanning calorimetry curve from the second scan (±10 °C/min) of the Janus AZOIOs membrane.



Figure S4. Change of reflectance spectra of the PC side after being immersed in the solvent of (A) water; (B) ethanol; (C) acetonitrile and (D) toluene.



Figure S5. The variation of the membrane surface temperature in (A) air and (B) toluene with time under UV irradiation (120 mW/cm²).



Figure S6. Schematic illustration of distinct bending modes of the Janus membrane under UV-vis irradiation. (A, A₁) Actuation mechanism of the polymer side under UV-vis irradiation in air. (B, B₁) Actuation mechanism of the polymer side under UV-vis irradiation in toluene.



Figure S7. The finite elemental simulation of Janus AZOIOs membrane when polymer side was UV irradiated in toluene. (The membrane is equivalent to a double-layer structure: the polymer side is the actuating layer, while the PC side is the passive layer. The thickness of PC and polymer layer are 11.5 μm and 29.7 μm, respectively. The length and width of the membrane are 10 mm and 3 mm, respectively. The Young's modulus of the polymer layer is 2.67 GPa, the Poisson's ratio is 0.36, and the thermal expansion coefficient is 106.7 ppm.)



Figure S8. (A) Solvent-responsive actuation of Janus AZOIOs membrane in THF environment and (B) corresponding schematic illustration. (C) Swelling degree of the PC and polymer sides in THF calculated from the swelling equilibrium equation.



Figure S9. (A) Photo-responsive actuation of Janus AZOIOs membrane in acetonitrile environment when the polymer side was UV irradiated. (B) Swelling degree of the PC side in acetonitrile calculated from the Bragg equation.

Solvent	Swelling degree (PC side)	Actuation mode
THF	33.3%	solvent-responsive
toluene	16.6%	photothermal
acetonitrile	1.8%	photochemical

Table 1 Actuation mode of Janus AZOIOs membrane in typical solvents.

The swelling ratio of Janus AZOIOs membrane in different solvents could regulate the actuation mode. For example, solvent-responsive actuation of Janus AZOIOs membrane was observed in THF environment (Figure S8A). In details, the membrane bent toward the polymer side after being immersed in THF, then recovered to its initial flat state in air. The reason of this bending behavior was the larger expansion of the PC side than that of the polymer side based on the porous structure in the PC side (Figure S8B). The swelling degree for the PC/polymer sides were 33.3%/20.3% in THF (Figure S8C), which indicated a value (swelling differences between the PC and polymer sides) of 13% could trigger solvent actuation.

Furthermore, the membrane bent away from the light source and formed a cylindrical shape in toluene when the polymer side was UV irradiated (Figure $2A_2$ in the manuscript), which indicated photothermal actuation. The swelling degree for the PC/polymer sides were 16.6%/9.6% in toluene (Figure 3D in the manuscript). Although the swelling difference between the PC and polymer sides (7%) in toluene was not sufficient to trigger solvent actuation, the significant swelling and permeability of toluene towards the azobenzene polymer network could effectively disrupt mesogen alignment (Figure 4A-B in the manuscript), thereby forming a photothermal actuation mode.

As a comparison, the membrane bent towards the light source in acetonitrile when the polymer side was UV irradiated (Figure S9A), which indicated photochemical actuation. The originally ordered mesogen alignment could be well maintained due to the low swelling (1.8%, Figure S9B) and permeability of acetonitrile towards the azobenzene polymer network. In summary, the different actuation modes of Janus AZOIOs membrane in different solvents were listed in Table 1. The membrane demonstrated solvent actuation in THF based on the larger expansion difference between the PC and polymer sides; the membrane demonstrated photochemical actuation in the swollen polymer network; the membrane demonstrated photochemical actuation in acetonitrile based on the unchanging monodomain mesogen alignment.



Figure S10. Photograph series showing continuously floating and sinking of Janus AZOIOs membrane driven by bubbles under UV light (120 mW/cm²) in the liquid phase.