Electronic Supplementary Information for "Colour-Tunable Spiral Photonic Actuators"

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Materials and sample preparation. A transparent poly(dimethylsiloxane) (PDMS) (Dow Corning, Sylgard 184®) elastomer with a refractive index that matched that of silica was selected as the hydrophobic layer material for the actuator. For the hydrophilic layer, a ultra-violet (UV)-curable elasomeric polyurethane (PU) precursor (Noland product, NOA 71) and a hydrogel 2-hydroxyethyl methacrylate (HEMA) monomer (Sigma Aldrich) with its 5 wt% darocure-1173 initiator (Chiba Co.) was blended in a series of compositions. We chose a hydrophilic transparent layer consisting of 70 wt% PU and 30 wt% HEMA (PU70/HEMA30) which is elastic and responds to hydrophilic solvents. The glass transition temperatures were measured utilizing both differential scanning calorimetry and dynamic mechanical analysis as well as by the evaluating the tensile properties the of PU/HEMA layers at a various compositions at room temperature. The results are shown in Figs. S1 and S2. In order to combine the multi-faceted environmental responsiveness of polymer gels with photonically active structures, silica colloidal FCC crystals (silica sphere diameter = 270 nm, Bangs Laboratories) were first assembled on the fluorinated silane (tridecafluoro-1,1,2,2-tetrahydrooctyltrichlorosilane, Sigma Aldrich) monolayer treated mold with dimensions of 2 mm × 40 mm × 0.2 mm for the spiral opal actuator. Utilizing Colvin's method

multiple times, we controlled the thickness of the silica colloidal PC to be 0.08 ± 0.01 mm (~ 300 silica sphere layers). The hydrophobic PDMS precursor infiltrated the silica colloidal template and was then thermally cured at 65 °C for 12 hr. Before applying the hydrophilic PU70/HEMA30 layer, a methacryloxypropyl-trimethyoxysilane (Sigma Aldrich) monolayer was created on the oxygen plasma treated PDMS as an adhesive between the hydrophobic PDMS and a hydrophilic PU/HEMA layer. In order to keep the sample thickness even, parafilm and a glass plate were placed on the top of the mold during the UV-curing process of the hydrophilic PU70/HEMA30 layer. The final opal planar switches were peeled from the molds and stored in vacuum before carrying out characterization and analysis.

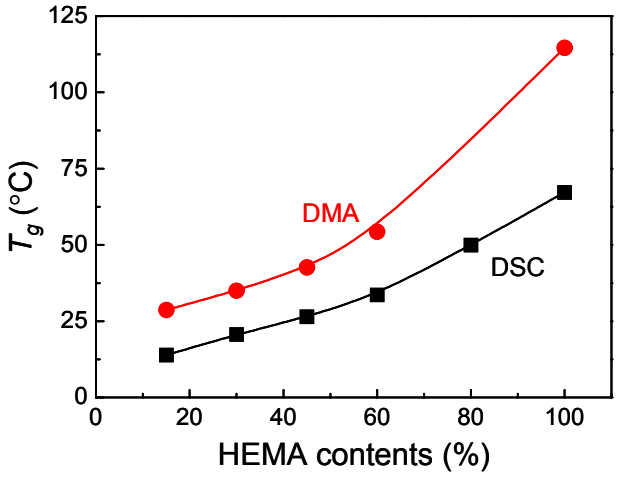

Fig. S1 Glass transition temperatures of HEMA in PU/HEMA blends at different HEMA concentrations, which is measured by dynamic mechanical analysis and differential scanning calorimetry.

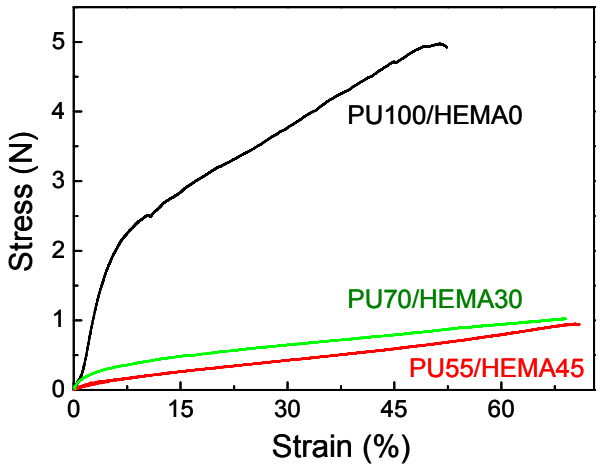

Fig. S2 Mechanical tensile tests of PU/HEMA blends at various compositions.

Equipment and Experiments. The silica colloidal opal and the PDMS imbedded opal morphologies were investigated by SEM (JEOL 6060 and FESEM JSM-7401). The pictures of spiral opal switches were taken using a digital camera (EOS 5D, Canon). Utilizing oxygen plasma (PDC-32 G, HARRICK) for 4 min at 75 mTorr and 150 W, hydroxyl functions, which can react with methacryloxypropyltrimethoxysilane in ethanol, were created on the surface of the PDMS-embedded opals. Swelling tests and gel content measurements for PDMS and PU70/HEMA30 in hexane, acetic acid and ethyl acetate were carried out on samples with dimensions of 5 mm × 30 mm × 1 mm. Reflectivity spectra were obtained using an optical microscope (Zeiss Axioscop) equipped with a fiber-optic spectrometer (Stellarnet EPP2000) and a near infrared spectrometer (6000i, Varian) using a silver-coated metallic mirror as a 100% reference.

Preparation of photonic actuator: Colloidal silica face-centered cubic (FCC) single crystals (silica sphere diameter = 270 nm) were self-assembled on the fluorinated silane monolayer treated

mold with dimensions of 2 mm × 40 mm × 0.2 mm. The sample geometry is identical with the one illustrated in Fig. 1a. The *y*-axis is more or less parallel to the $[0\,\bar{1}\,1]$ direction of the silica colloidal FCC structure. To photograph close to the [200] zone of the silica colloid FCC structure (Fig. S3b), the [111] zone sample (Fig. S3a) was tilted 45°-clockwise around the *x*-axis toward y-axis. The reddish color reflected from the [111] zone was found to have a peak reflection wavelength of 590 nm (Fig. 2) which matches the calculated reflection band peak at 596 nm. The greenish reflected color from the [200] zone is also well matched with the calculated peak reflection wavelength of 507 nm, which is deduced from the analysis of silica colloid FCC structure. The silica/air PC reflected red and green light close to the [111] and [200] zones of silica colloidal photonic crystal, respectively.

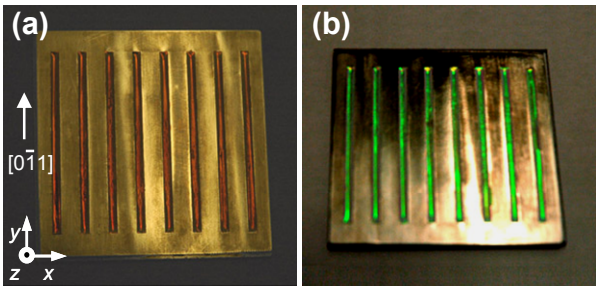

Fig. S3 Photographs of 270 nm silica colloidal photonic crystal. (a) from the [111] zone and (b) close to the [200] zone of the FCC structure.

Swelling tests: In order to test the reversibility and to evaluate the gel content of the PDMS and PU70/HEMA30 layers in different solvents, such as hexane, acetic acid and ethyl acetate, swelling tests were conducted with respect to swelling and deswelling times, as shown in Fig. 3. The maximum swelling expansion ratio (V_{max}/V_0, here V_{max} is the expanded volume at swelling time, $t = t_{max}$ and V_0, is the initial volume at $t = t_0$) of the hydrophobic PDMS layer is 2.28 in hexane, 1.66 in ethyl acetate, and 1.10 in acetic acid. On the other hand, the V_{max}/V_0 of hydrophilic

PU70/HEMA30 is 1.00 in hexane, 1.66 in ethyl acetate, and 2.05 in acetic acid. All the samples swelled more than 90% of their maximum swelling expansion ratio in 10 min. During the deswelling process, PDMS samples in all the solvents returned back to the original planar dimension in 20 min, while PU70/HEMA30 samples in acetic acid and ethyl acetate took more than 1 hr. The gel content of the thermally cured PDMS and UV-cured PU70/HEMA30 layers was also evaluated by the before and after swelling weight ratio in good solvents. The gel content of PDMS measured in hexane and PU70/HEMA30 in acetic acid is 100% and 99%, respectively.

Define the handedness: In order to determine the handedness of scrolled opal switches, we need to define the directions of the PDMS and PU70/HEMA30 layers. Using the right-handed 3D Cartesian coordination system which is illustrated in Fig. 1 and Fig. S4, the + *z*-axis and – *z*-axis are defined as the PDMS and PU70/HEMA30 layers, respectively. The simplest way to recognize which side is the PDMS or PU70/HEMA30 layer in the scroll opal actuator is mechanical stretching. When the sample is released after mechanically stretching about 10% along the *y*-axis (long axis of scroll actuator), a right-handed scroll is first formed and returns back to the original planar shape (Fig. S4a). This is because the elastic recovery of the PDMS layer (+ *z*-axis) is faster than that of the PU70/HEMA30 (– *z*-axis) layer.

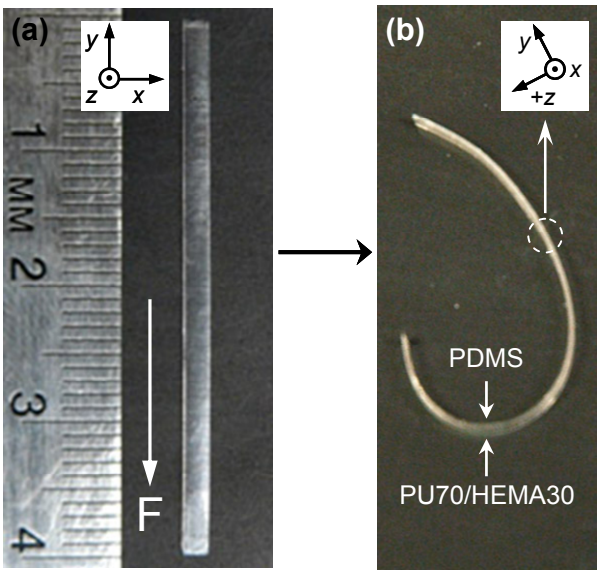

Fig. S4. Photographs of scroll actuators (a) before and (b) after mechanically stretching along the y axis.

Fig. S5. Left-handed photonic spiral actuator in ethyl acetate: (a) side view, (b) 45°-tilted view, and (c) top view, respectively.