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Supporting Information

Simultaneous Control of Gaussian Curvature and Buckling Direction by Swelling of Asymmetric Trilayer Hydrogel Hybrids

Seog-Jin Jeon^{a,b} and Ryan C. Hayward^{b,*}

^a Department of Polymer Science and Engineering, Kumoh National Institute of Technology, Gumi, Gyeongbuk 39177, South Korea

^b Department of Polymer Science and Engineering, University of Massachusetts Amherst, Amherst 01003, USA

*hayward@umass.edu

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Fig S1. ¹H NMR spectra of photo-crosslinkable polymers. (a) PDEAM copolymerized with 1 mol% BP, (b) PDEAM copolymerized with 5 mol% BP, (c) PDEAM copolymerized with 7 mol% BP and 0.3 mol% RhBMA, and (d) PpMS copolymerized with 5 mol% BP. The sharp single peak is chloroform.

Fig S2. (a) The diameter of a circular sheet of PDEAM hydrogel is (a) 520 μ m in deswelled state (60°C) and (b) 860 μ m in swelled state (10°C), leading to swelling ratio of 1.65.

Fig S3. (a) Bottom and (b) top mask designs for concentric rings with constant dash and gap lengths of 35.8 and 7.2 μ m, respectively. To reduce alignment between neighbouring gaps on the same surface, each ring moving outward from the centre is azimuthally offset by 15° from the previous ring, but there are still 'seams' of alignment between gaps in neighboring rings, as denoted in (a) by the red shaded regions. To prevent alignment between gaps between top and bottom PpMS patterns, the PpMS patterns on these surfaces are also azimuthally offset by 15°. (c) However, overlaying the patterns in (a) and (b) reveals that gaps in the 4th ring are still aligned. (d) Thus, an additional azimuthal offset of 7.5° is applied only to the 4th ring in the top PpMS pattern, preventing alignment between gaps on the top and bottom surfaces as shown by the overlay.

Fig S4. Mask design used for samples in Fig 3a and Fig 5 of the main text to ensure that gaps do not align either between neighboring concentric rings, or between the top and bottom surfaces on the same ring.

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Mov S1. The trilayer structure with stripes of PpMS seen in Fig 1b and c is showing anisotropic expansion and is remained in a flat geometry during the swelling-deswelling cycle. Playback is at 8 times real speed.

Mov S2. The trilayer structure with concentric continuous lines of PpMS seen in Fig 3b is forming a cone with unwanted distortion, a dimple near the apex.

Mov S3. Morphing three connected cones in a row seen in Fig 5a and b is demonstrated for temperature sweep between 60°C and 10°C. The heding direction of cones is consistent with the pre-programmed heading direction, upward (left) / downward (mid) / upward (right). Playback is at 8 times real speed.



Fig S1. ¹H NMR spectra of photo-crosslinkable polymers. (a) PDEAM copolymerized with 1 mol% BP, (b) PDEAM copolymerized with 5 mol% BP, (c) PDEAM copolymerized with 7 mol% BP and 0.3 mol% RhBMA, and (d) P*p*MS copolymerized with 5 mol% BP. The sharp single peak is chloroform.



Fig S2. (a) The diameter of a circular sheet of PDEAM hydrogel is (a) 520 μ m in the deswelled state (60°C) and (b) 860 μ m in the swelled state (10°C), corresponding to a linear swelling ratio of 1.65.



Fig S3. (a) Bottom and (b) top mask designs for concentric rings with constant dash and gap lengths of 35.8 and 7.2 μ m, respectively. To reduce alignment between neighbouring gaps on the same surface, each ring moving outward from the centre is azimuthally offset by 15° from the previous ring, but there are still 'seams' of alignment between gaps in neighboring rings, as denoted in (a) by the red shaded regions. To prevent alignment between gaps between top and bottom PpMS patterns, the PpMS patterns on these surfaces are also azimuthally offset by 15°. (c) However, overlaying the patterns in (a) and (b) reveals that gaps in the 4th ring are still aligned. (d) Thus, an additional azimuthal offset of 7.5° is applied only to the 4th ring in the top PpMS pattern, preventing alignment between gaps on the top and bottom surfaces as shown by the overlay.



Fig S4. Mask design used for samples in Fig 3a and Fig 5 of the main text to ensure that gaps do not align either between neighboring concentric rings, or between the top and bottom surfaces on the same ring.