Supporting Information for:

Multifunctional Actuation Systems Responding to Chemical Gradients

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Experimental

Sample preparation and characterization

Acrylamide (AAm), acrylic acid (AAc), *N*,*N*'-methylenebisacrylamide (bis-AAm), glycidyl methacrylate (GMA), 2-hydroxy-4-(2-hydroxyethoxy)-2-methylpropiophenone, and (tridecafluoro-1,1,2,2-tetrahydrooctyl) triethoxy silane were purchased from Sigma-Aldrich (St. Louis, MO). Polydimethylsiloxane (PDMS) (Dow-Sylgard 184) was purchased from Ellsworth (Germantown, WI) and UVO-114 was purchased from Epoxy Technology (Billercia, MA). All the chemicals were used as received.

Silicon masters of the microfin and micropost array were fabricated using the Bosch process[1,2] and replicated in GMA-modified epoxy via PDMS molding as described elsewhere.[3] Each microfin was 1.2 µm wide and 11.5 µm tall and separated side-to-side and front to back by 5 µm. Micrposts were 1.5 µm in diameter, 10 µm tall with an 8 µm pitch.

To pattern hydrogel within the microfluidic channels, a photomask was used. The photomask with rectangular area (7 mm x 750 µm) was created with a Versa laser cutter in a double sided, pressure sensitive adhesive sheet (Graphix, Maple Heights, OH). The cut sticker was colored black and then pressed onto a No. 2 cover slip to create a mask.

A few microliters of UV-initiated pH responsive hydrogel precursor solution (20% AAm, 20% AAc, 2% bis-AAm, 1% 2-hydroxy-4-(2-hydroxyethoxy)-2-methylpropiophenone by weight in deionized water) was placed on the GMA-epoxy microstructure sample surface. To make surfaces that actuate in response to bulk pH, the hydrogel layer had to be very thin and the coverslip should touch, or come close to touching, the structures. Upon hydrogel curing, the structures bend over. To make surfaces which do not respond to bulk pH, the coverslip distance from the structures was controlled by using spacers. The cover slip mask was gently put over the droplet to induce wetting of the hydrogel over most of the sample surface. The sample was cured under a UV lamp (100W, Black-Ray with no filter) at 5 inches away for 4 minutes and afterwards the cover slip mask was removed. The sample was rinsed with water to remove excess precursor solution.

Microchannels were created in a similar fashion to the mask by using a laser cutter to cut T-channel patterns or rectangular patterns (7 mm long and 750 µm wide) into the same double sided, pressure sensitive adhesive. Two or more adhesive sheets were used together for the T-channel depending on the depth of channel desired. PDMS was used to seal off the top of the channel. Solution was introduced to the channels using a syringe pump and polyethylene tubing. Typical flow rates ranged from 10-50 µL per minute.

Optical imaging and video recording was done on an Olympus IX71 inverted microscope using StreamPix v.3 software and QImaging Retiga 2000R color camera.

Confocal Microscopy

Pyrromethene 546 (Exciton, Inc.) was mixed into the GMA-modified epoxy and used for structure replication. 1μm red fluorescent carboxylate-modified microspheres (Invitrogen, 2% solids) were diluted 10x in deionized water and a few drops were let to dry on the surface of the hydrogel after sample fabrication but before integration into a microfluidic cell. Confocal microscopy was done on a Leica DMI3000 SP5 TCS in conventional mode.

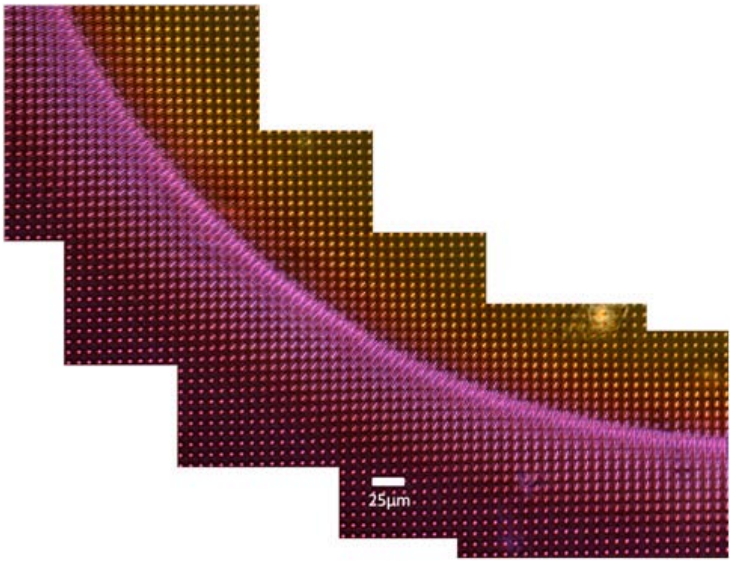

Figure S1. Laminar flow interface of acid (yellow) and base (purple) shows that the embedded microposts always bend perpendicular to the interface, even when the interface is curved, providing chemical control over the bending direction. Color arises from bromophenol blue pH indicator.

[1] T. N. Krupenkin, J. A. Taylor, T. M. Schneider, S. Yang, *Langmuir* **2004**, *20*, 3824..
[2] S. A. McAuley, H. Ashraf, L. Atabo, A. Chambers, S. Hall, J. Hopkins, G. Nicholls, *J. Phys. D: Appl. Phys.* **2001**, *34*, 2769–2774.
[3] P. Kim, L. D. Zarzar, X. Zhao, A. Sidorenko, J. Aizenberg, *Soft Matter* **2010**, *6*, 750-755.