Laboratory's deep space network and on spacecraft, transporting signals for detectors, sensors and phase array antennas or carrying long distance signals, to name a few. It is conceivable that ceramic-ribbon waveguide may be the backbone of future communication systems in this frequency band. ☐

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Functional hydrogel structures for autonomous flow control inside microfluidic channels

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Hydrogels have been developed to respond to a wide variety of stimuli[1–6], but their use in macroscopic systems has been hindered by slow response times (diffusion being the rate-limiting factor governing the swelling process). However, there are many natural examples of chemically driven actuation that rely on short diffusion paths to produce a rapid response[7]. It is therefore expected that scaling down hydrogel objects to the micrometre scale should greatly improve response times. At these scales, stimuli-responsive hydrogels could enhance the capabilities of microfluidic systems by allowing self-regulated flow control. Here we report the fabrication of active hydrogel components inside microchannels via direct photopatterning of a liquid phase. Our approach greatly simplifies system construction and assembly as the functional components are fabricated *in situ*, and the stimuli-responsive hydrogel components perform both sensing and actuation functions. We demonstrate significantly improved response times (less than 10 seconds) in hydrogel valves capable of autonomous control of local flow.

Conventional microactuators (using, for example, electromagnetic, electrostatic or thermopneumatic effects) require external power for operation and relatively complex assembly, which limits their use in practical systems[8]. Stimuli-responsive hydrogels have a significant advantage over conventional microfluidic actuators owing to their ability to undergo abrupt volume changes in response to the surrounding environment without the requirement of an external power source. Difficulties in integrating conventional micrometre-scale components into functional systems using traditional approaches[8,9] have limited the potential benefits of emerging microfluidic systems. Unconventional approaches are needed to overcome these difficulties in microscale integration. Recently, several groups have explored new fabrication methods that show promise[10–14]. Our approach combines lithography, photopolymerization and microfluidics to create functional components (valves) within microchannels for local flow control. We believe that this approach has widespread applications for flow regulation in microfluidic systems.

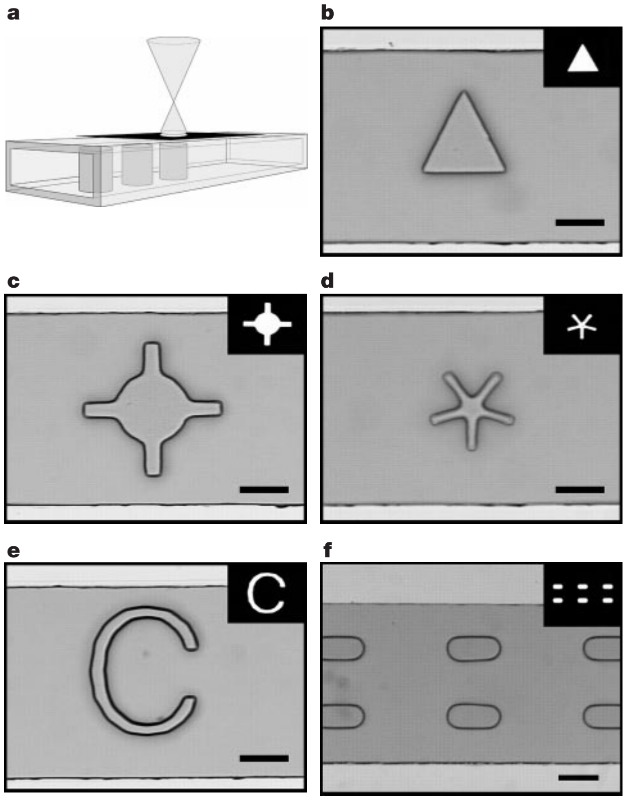

Figure 1 A diagram of the fabrication method and images demonstrating a variety of shapes that were polymerized within 35 seconds. **a**, The fabrication method. **b**, A polymerized hydrogel demonstrating the ability to pattern high-definition straight edges. The corresponding photomask is shown at a reduced size in the upper right corner of each picture. **c**, **d**, Structures illustrating the generation of convex and concave surfaces. **e**, A structure with high-aspect-ratio features. Imperfections in the mask were transferred to the structure, further demonstrating the high fidelity of the photolithographic process. **f**, The simultaneous polymerization of multiple structures with a single exposure of ultraviolet light. Scale bars: **b**–**e**, 250 μm; **f**, 500 μm.

The general approach we use is to flow a mixture of monomers and a photoinitiator into the microchannel, and irradiate the mixture through a photomask (Fig. 1a). In a typical procedure, transparent channels ranging from 500 to 2,000 μm wide and 50 to 180 μm deep are filled with a photopolymerizable liquid consisting of acrylic acid and 2-hydroxyethyl methacrylate (in a 1:4 molar ratio), ethylene glycol dimethacrylate (1 wt%) and a photoinitiator (3 wt%). The liquid is allowed to reach a quiescent state and is then exposed to ultraviolet light through a photomask placed on top of the channel. Polymerization times vary depending on light intensity, photoinitiator and monomer mixture, and can be less than 20 seconds using Irgacure 651 as the photoinitiator and the filtered light source from a standard fluorescence microscope. When the polymerization is finished, the channel is flushed with water to remove the unpolymerized liquid. This method allows pH-responsive hydrogels of different shapes and sizes to be integrated directly into microfluidic systems.

As seen in Fig. 1, the pattern of the photomask is transferred to the polymerized object with high fidelity. The minimum feature size is 25 μm, coinciding with the minimum resolution of the photomask. Confocal micrographs of an object polymerized in a 180-μm-deep channel by irradiation through a circular mask reveal a slightly tapered cylindrical shape, with a smaller diameter at the bottom than the top. The polymerized hydrogel is in contact with both the top and bottom of the channel. The fabrication of multiple structures can be performed either sequentially by moving the mask, or simultaneously by using a mask with a multi-structure pattern (Fig. 1f). Furthermore, multiple hydrogels of different chemical compositions can be fabricated in a single channel by sequential fabrication, as demonstrated below for the flow sorter.

The time response of the volume change approximately follows the square of the dimension as the hydrogel objects reversibly expand and contract, depending on the pH of the surrounding environment. We characterize this dynamic behaviour by measuring the step response of the hydrogel expansion. A step response of less than 10 seconds was observed for a 100-μm-diameter cylindrical structure in a 50-μm-deep channel, but this configuration was mechanically unstable. Hydrogel objects tend to buckle or migrate during a volume change if their lateral dimensions are smaller than the channel height. To fabricate stable objects with fast response times, we polymerize the hydrogel structures around prefabricated posts. The posts provide a robust support, and also improve time response owing to the short diffusion path of the hydrogel jackets surrounding the posts. An array of hydrogel-coated posts can control the flow in large channels, as shown in Fig. 2. The step response for expansion of this array valve design is 8 seconds (the contraction step response is of the same order). In contrast, an alternative valve design that uses a single larger cylindrical structure in the same size channel has a step response of 130 seconds over the same pH range. In this case, the post design accelerates the response time by a factor of 16 (Fig. 2e). This integration of hydrogels into microfluidic systems provides the scaling necessary to overcome the primary drawback (slow time response) of hydrogels.

Expansion and contraction of the hydrogels can regulate the fluid flow in microchannels. Measuring the pressure drop at constant flow rate over a channel containing hydrogel structures reveals this behaviour. To illustrate this, we use a flow rate of 0.15 ml min^{-1} to pump solutions of various pH values past oval-shaped structures (300 × 700 μm, contracted state) polymerized in a 1 × 9 array along the length of a 1,000-μm-wide glass channel. In an acidic

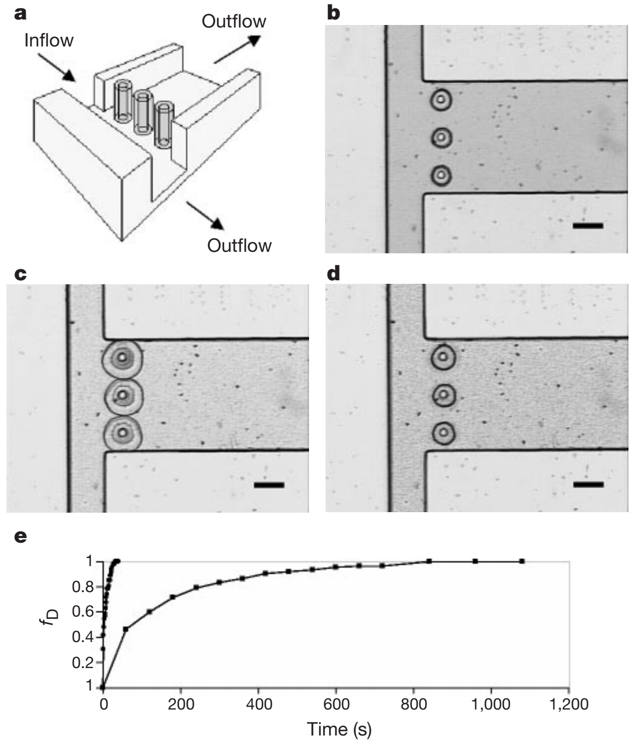

Figure 2 Prefabricated posts in a microchannel serve as supports for the hydrogels, improving stability during volume changes. **a**, A diagram of the hydrogel jackets around the posts. **b**, The actual device after polymerization of the hydrogel. **c**, The hydrogel jackets block the side channel branch in their expanded state. **d**, The contracted hydrogels allow fluid to flow down the side branch. **e**, The improvement in time response of the hydrogel jacket design (circles) versus an alternative design that uses a single larger cylindrical structure in the same size channel (squares). f_D is the fractional change in diameter. Scale bars, 300 μm.

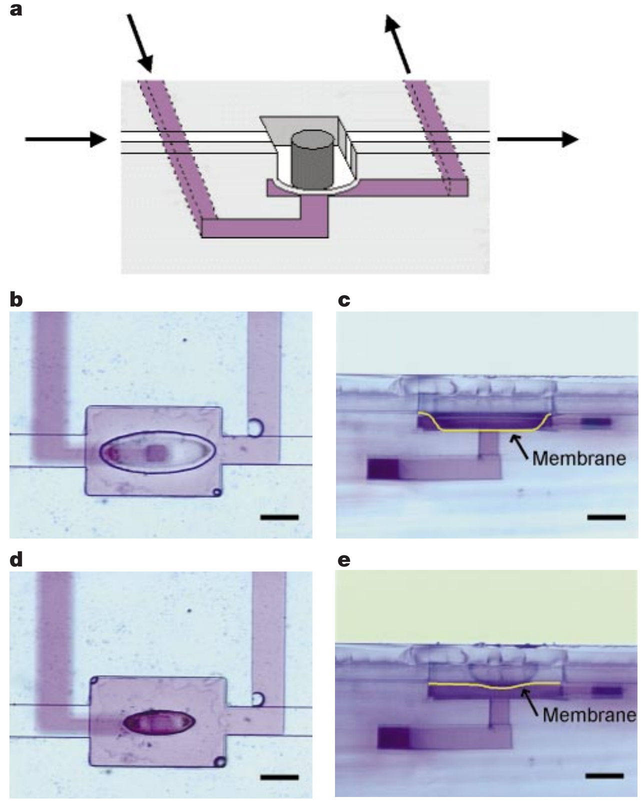

Figure 3 A shut-off valve. **a**, A diagram of the valve design. The arrows denote the direction of fluid flow. **b**, **c**, The hydrogel structure expands and deforms a membrane, blocking flow in an adjacent channel. The images on the left show a top view of the device, and the images on the right show the side view. The fluid in the blocked channel has been dyed for visualization purposes. **d**, **e**, The hydrogel contracts, and the membrane returns to a position that allows flow in the adjacent channel. In **c** and **e**, the boundary of the membrane has been outlined for clarity (yellow). Scale bars, 250 μm.

environment, the objects are in a contracted state and produce a pressure drop of 0.09 pounds per square inch (p.s.i.). Upon raising the pH above the transition point of these hydrogels, the objects fully expand, causing the pressure drop to increase almost eightfold. This simple-to-fabricate device thus functions as a pH-sensitive throttle valve for microfluidic systems.

Through appropriate design, a single hydrogel component can sense the chemical environment in one channel and regulate the flow in an adjacent channel, as shown in Fig. 3. This device contains a flexible membrane that can deform to block the flow in an adjacent channel. The hydrogel structure polymerized in the channel above the membrane expands or contracts as the surrounding pH is changed. The force associated with these volumetric changes is sufficient to deform the membrane and consequently control the flow in the lower channel. It is easy to imagine extension of this demonstration to antigen-responsive hydrogels[6] that could serve as devices in self-regulated drug delivery or biosensors.

An additional demonstration of the versatility of this approach is the fabrication of a self-regulated 'flow sorter'. This device consists of a 'T' channel in which the entrance to each branch is gated with a hydrogel structure of unique chemical composition. The hydrogel for one branch expands at high pH and contracts at low pH, while a hydrogel of a different composition gates the other branch and exhibits an inverse behaviour (that is, contracts at high pH and expands at low pH). The device and a graph of its resulting output responses are shown in Fig. 4. This device automatically directs the flow in the centre channel down one branch or the other depending on the pH. In a certain pH range (5.7–6.8), both hydrogel valves swell to seal the channel. Each hydrogel valve performs the sensing, actuating and regulating functions normally performed by discrete components (valve, sensors, electronics) in a traditional system. By tailoring the chemical composition of the hydrogel, the output response (the slope and position of the volume transition) can be modified, allowing pH-sensitive hydrogels to be used in a variety of applications.

The ability to fabricate functional structures within microfluidic channels has the potential to simplify greatly the processes required to build complex microfluidic systems. Our approach eliminates difficult microscale assembly and the need for electronics for sensing and actuation. In addition, fast response times are achieved owing to the short diffusion paths in microchannels. As noted above, the approach is not limited to pH-response hydrogels, thus enabling a wide range of functional components. The microfluidic/photopolymerization fabrication method described here provides an approach that could be extended to build multifunctional microfluidic systems, allowing complex fluidic processes to be performed autonomously. □

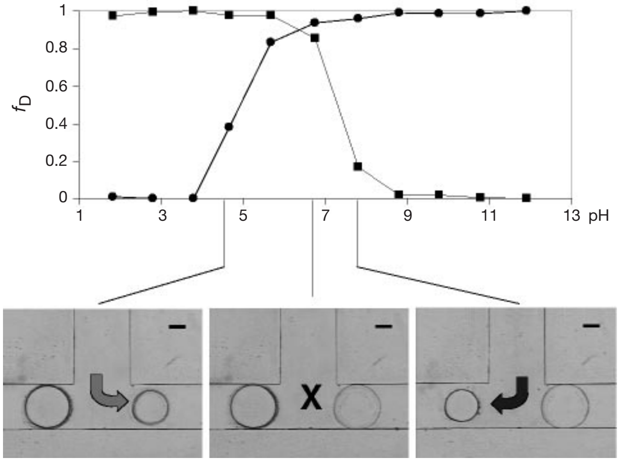

Figure 4 The volume response of two different hydrogels with respect to the pH of the surrounding fluid. Top, the fractional change in diameter (f_D) of the hydrogels with respect to pH. Bottom, images showing a device that directs ('sorts') a fluid stream on the basis of its pH. The hydrogel gating the right branch (circles) expands in base and contracts in acid. The hydrogel gating the left branch (squares) behaves in the opposite manner (expands in acid and contracts in base). The fluid enters from the centre channel at a rate of 0.05 ml min^{-1}. At a pH of 7.8, the flow is directed down the left branch. At a pH of 4.7, the flow is directed down the right branch. Both hydrogels expand to shut off the flow when the pH is changed to 6.7. Scale bars, 300 μm.

Methods

Photopolymerization

The photomasks were prepared by printing patterns on transparency films using a high-resolution commercial printer system (Linotype Herkules Imagesetter) with a resolution of 5,080 dots per inch. The photoinitiator, Irgacure 651, is the registered trade name of 2,2-dimethoxy-2-phenyl acetophenone (Ciba Speciality Chemicals). A near-ultraviolet filter cube (U-MNUA, type BP360-370) was used on an Olympus Epi-Fluorescent microscope (BX-60) to provide the energy necessary for polymerization. The band pass of the cube was 360–370 nm. The resulting hydrogel objects had side walls that deviated from the surface normal by $\sim$6°. All images are unprocessed. The purple hue in Fig. 3 is produced by a dye added for visualization purposes. Arrows and lines have been added in Fig. 3 to highlight the edge of the membrane to clarify its location.

Device fabrication and characterization

The channels in Fig. 1 were constructed by bonding two no. 1 coverslips to a glass substrate with a UV-curable adhesive. The coverslips were placed so that their edges were parallel and separated by the desired channel width. A third coverslip served as the channel top, producing channels that were $\sim$180 μm deep. The channel and posts for the device shown in Fig. 2 were fabricated by etching a 200-μm-thick photosensitive epoxy layer (Nano XP SU-8, Microchem Corp.) spin-coated on a Pyrex substrate. Transparent adhesive tape served as the top of the channel. Step response is the time required for the hydrogel to reach $(1 - 1/e)$ (63.2%) of its total volume change. The device in Fig. 3 was constructed using a fabrication technique previously developed[15]. The device shown in Fig. 4 has no. 1 coverslips as top and bottom, with a 200-μm-thick poly(dimethylsiloxane) gasket in between to define the channel height. The hydrogel on the left was prepared from 2-(dimethylamino)ethyl methacrylate and 2-hydroxyethyl methacrylate (in a 1:4 molar ratio), ethylene glycol dimethacrylate (1.4 wt%) and Irgacure 651 as the photoinitiator (3 wt%). The hydrogel on the right was prepared using the components listed in the text. The pressure measurements were taken using a Validyne model DP-15 pressure transducer with a 0–1 p.s.i. nickel plated diaphragm. The pressure drop measurements were taken over a channel containing hydrogel ovals with their major axes parallel to the channel walls, similar to the configuration shown in Fig. 1f. To verify fluid flow in the various device designs, a variety of flow visualization techniques such as particle tracking, dyes and air bubbles were used.

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