Fabrication of membrane-type microvalves in rectangular microfluidic channels *via* seal photopolymerization†

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Rectangular fluidic channels have rarely been used in microfluidic devices which use PDMS membrane-type microvalves, since the rectangular channel shape does not perfectly match the round shape of the membrane deformation. We present a polymer sealing method to fabricate PDMS membrane-type microvalves for rectangular microchannels. After fabricating the microfluidic device, photocurable oligomer is introduced into the fluidic channel and gas pressure is applied to the pneumatic channel to deform the membrane. The polymer seal is then locally polymerized by photolithography producing a structure matching the shape of the deformed membrane curvature. We compare the flow leakage between the membrane-type microvalve with and without a polymer seal. We also demonstrate a micropump and droplet generator using this embedded polymer membrane-type microvalve in a rectangular microfluidic channel. This polymeric seal technique enables the use of easily fabricated rectangular channel membrane microvalves with all the functionality of their curved channel counterparts with negligible flow leakage.

Introduction

Rapid prototyping of microfluidic devices has been popularized by the development of soft lithography, which is a simple sol–gel based replica molding process. In soft lithography, reusable molds are fabricated by simple photolithography and many microfluidic channel devices are replicated from the mold by thermal curing silicone based elastomers such as PDMS.[1] For reliable production of uniform devices the mold should be thermally stable and structurally strong. SU-8 is very popular for use as a replica-molding material since it is an epoxy-based negative photoresist, which forms a highly cross-linked structural layer *via* UV patterning. Due to this high degree of cross-linking, SU-8 is thermally stable, chemically resistive, and structurally strong, making it a good material for molds. Moreover, the optical transparency of SU-8 enables the fabrication of thick molds, up to several hundreds of micrometres, where as other positive photoresists can only form shallow molds on the order of several tens of micrometres. The chemical and mechanical stability of the SU-8 makes it possible to make multilevel molds by repeating the lithography steps on the same mold.[2,3] The cross-sectional shape of the microfluidic channels from the resulting SU-8 mold is a simple rectangle or a combination of multiple rectangular patterns. Rectangular channels are easy to design, with flow dynamics that are more easily visible using a microscope since they lack the shadowing that results from a curve channel architecture.

However, one drawback to rectangular microfluidic channels developed *via* the SU-8 molding process is the difficulty in the integration of active components such as microvalves.[4] Membrane type pneumatic microvalves are the most common microvalves in PDMS microfluidic devices and are used in many applications.[5–9] When the fluidic channel is rectangular it is impossible to completely shut off the flow since the curvature of the expanding membrane is round. For this reason, rounded microfluidic channels are used. Rounded microfluidic channels are molded from rounded molds fabricated through a resist reflow process.[10,11]Reshaping happens due to surface tension of the melted ‘reflowed’ resist on the unmelted resist. The maximum channel thickness that can be achieved by using conventional positive resist is tens of micrometres, limiting the height of the rounded fluidic channel. Other resists such as SU-8 can allow for high aspect ratio structures, but cannot be reflowed since they are highly cross-linked and thermally stable. Since thermal ‘reflow’ is not required using the method presented here, materials such SU-8 can be used

In this paper, we present a PDMS membrane microvalve with an *in situ* polymerized seal embedded in a rectangular channel. We locally fabricated the polymer sealed valve and compare it to a typical microvalve in a rectangular channel without a polymeric seal. We also replicate microcomponents, formed by traditional membrane-based microfluidic valves such as a droplet generator and a micropump.

Results and discussion

A PDMS chip was constructed so that a membrane-type microvalve in a rectangular microfluidic channel contained an *in situ* polymerized seal as shown in Fig. 1a. The rectangular channel allowed for high accuracy fabrication without resist reflow processing. The polymeric seal is fixed on the bottom of the channel and forms a hemi-cylindrical shape when viewed from a cross-sectional vantage point (Fig. 1b). This polymeric seal structure enables perfect leak-free operation in a rectangular microchannel. When the gas pressure is applied to the pneumatic

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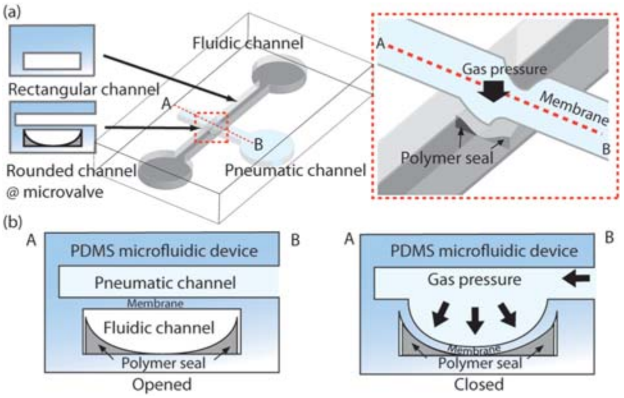

Fig. 1 Schematic diagram of a polymer-sealed microvalve. (a) 3D view of polymer-sealed membrane microvalve (A–B). (b) Cross-sectional view of a rectangular PDMS microfluidic channel integrating a polymer-sealed membrane-type microvalve, with a hemi-cylindrical cross-section. Pneumatic pressure causes shape matching of the membrane valve to the polymer seal.

channel, the membrane is deformed to a hemi-cylindrical shape to form a tight seal against the curved polymer.

We fabricated a rectangular microfluidic channel with a membrane-type microvalve in order to compare the operation of a polymer-sealed valve to an unsealed valve (Fig. 2a). Red dye is introduced into the fluidic channel in both devices that intersects a pneumatic channel on an adjacent layer. When the valve without the polymer seal is closed, the leakage of red dye can be found along the wall of the fluidic channel (Fig. 2c). A shadow from the deformed membrane of the microvalve interferes with observing the leakage of the red dye as shown in Fig. 2c, therefore a fluorescent nanoparticle solution (Fluorescent Nile Red Particles 1.0 w/v 0.04–0.06 μm, Spherotech) is used for observing the leakage of flow through the microvalve using fluorescence imaging. As can be seen in Fig. 2e, without the polymer seal the fluorescent nanoparticle solution still flows along the wall of the fluidic channel even though the microvalve is closed. In order to prevent microvalve leakage, a polymeric seal is fabricated in the rectangular channel (Fig. 2b). When the valve with a polymer seal is closed, the red dye disappears in the layer vertically adjacent to the microvalve (Fig. 2d). As shown in Fig. 2f, there is no leakage along the walls of the channel as in the valve without the polymer seal.

The microdevice with microvalves we use in this experiment is composed of three layers: a pneumatic channel layer, a fluidic channel layer, and a glass substrate. The pneumatic channel layer and fluidic channel layer are fabricated through a soft lithography process. The glass substrate is pretreated with 3-(trimethoxysilyl)propylmethacrylate (TMSPMA) to enhance bonding between the glass and the polymer seal[12,13] (Fig. 3a). All the layers were aligned and bonded after plasma treatment. To generate a polymer seal, the prepolymer, trimethylolpropane triacrylate (TMPTA), is introduced into the fluidic channel of the microfluidic device (Fig. 3b). After filling the fluidic channel with prepolymer, gas pressure is applied to the pneumatic channel in order to deform the membrane into a round shape. A rectangular UV light pattern generated by a digital micromirror device (DMD) is illuminated on the prepolymer contacting the microvalve. Alternatively, a fluorescent microscope with a mercury lamp can be used for generating a UV pattern by locating a mask in front of the field stop of the microscope.[14] The polymer seal is polymerized in a rectangular UV pattern 3-dimensionally matching the shape of the deformed membrane (Fig. 3c). The uncured prepolymer is washed out with 1 ml ethanol and dried for 1 minute at 100 °C on a hot plate (Fig. 3d). A gap, resulting from oxygen diffusion through the channel during polymerization appears between the PDMS wall and polymer seal. The cross-section of the microvalve is observed in Fig. S1† in order to analyze the polymer-sealed microvalve's on/off flow ratio taking into account the gap between the PDMS wall and polymer seal (Fig. S1†).

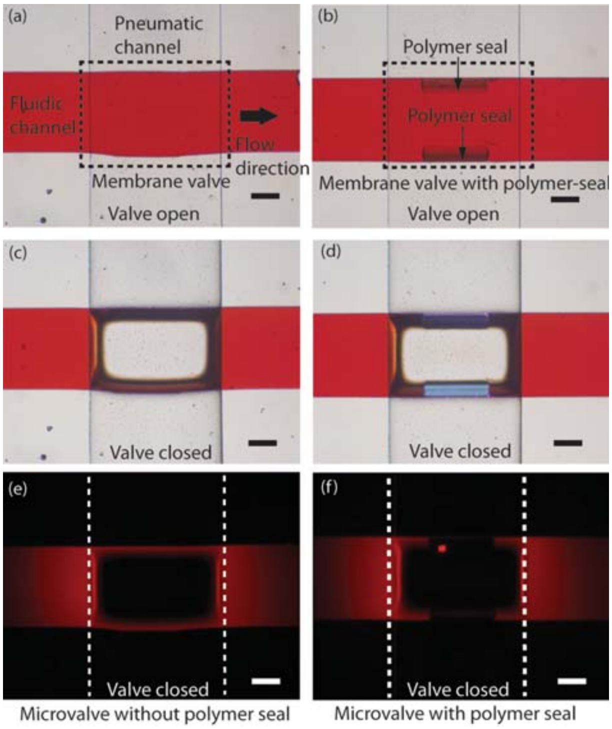

Fig. 2 Comparison between a polymer-sealed valve and an unsealed valve in a rectangular microfluidic channel. (a) An unsealed valve in a rectangular microfluidic channel. Red dye is used to image the fluidic channel, and to observe the flow leakage. (b) Polymer-sealed valve. (c) When the valve without a polymer seal is closed, red dye is leaked along the wall of the fluidic channel. (d) When the valve with a polymer seal is closed, the red dye disappears on the microvalve and the fluidic channel is perfectly closed. (e, f) For a more clear observation, a fluorescent nanoparticle solution is used. The input pressure for red dye flow is 1 psi while the pressure in the pneumatic channel when activated is 15 psi. Scale bar: 200 μm.

As shown in Fig. 4, flow rate can be controlled from 0 to 1 (0: valve close, 1: valve open) using the microvalve with a polymer seal (blue line). Red dye solution is injected into the microfluidic channel at 1 psi pressure while the pneumatic channel pressure is varied from 0 to 14 psi. The flow rate is measured at various pneumatic channel pressures. As the pneumatic channel pressure is increased, the flow rate decreases. At 10 psi, the graph bottoms out at 0 (as far as can be detected), which means that the flow is completely stopped by the microvalve. The microvalve

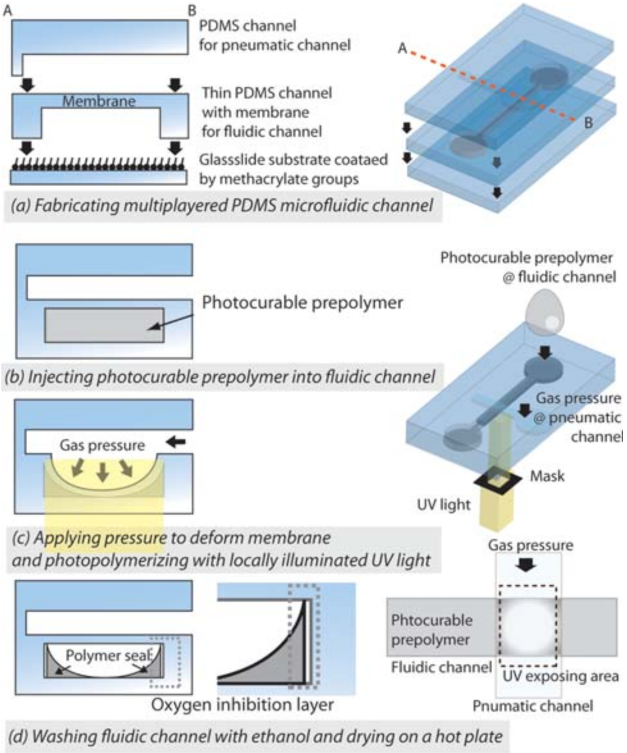

Fig. 3 Steps for fabricating a microvalve with an *in situ* polymerized seal. (a) Microfluidic channel fabrication. A PDMS layer for the fluidic channel is placed on a thin PDMS layer for the pneumatic channel and a glass substrate coated with TMSPMA. (b) Injecting photocurable prepolymer. (c) Applying pressure to the pneumatic channel in order to deform a membrane then polymerizing a polymer seal by illuminating UV light using a rectangular pattern. (d) Stopping applying the pressure and washing the fluidic channel with ethanol.

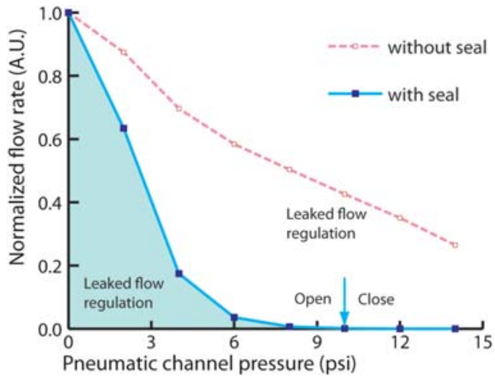

Fig. 4 The leaked flow rate comparison between the microvalve with a polymer seal and without. The graph for the flow rate in the microvalve with a polymer seal (blue line) falls steeply until it reaches 10 psi compared to the graph for the flow rate in the microvalve without a polymer seal (red dotted line). After 10 psi, the flow rate of the microvalve with a polymer seal (blue) is saturated at 0. On the other hand, the graph for the microvalve without a polymer seal (red dotted line) continuously falls. (fluidic pressure: 1 psi).

lacking a polymer seal cannot decrease the flow rate to 0, limiting it to use as a flow regulator because the leakage of fluid along the wall of the fluidic channel.

Membrane-type microvalves have previously been used for advanced microcomponents, such as a droplet generation and for a peristaltic micropump (Fig. S2†). Fig. 5 shows on-demand droplet generation in a rectangular microfluidic channel valve with an embedded polymer seal. In order to generate the aqueous droplet inside the microfluidic channel, the surface inside the microfluidic device is required to be hydrophobic. PDMS-coated glass is used to make all of the surfaces inside the microfluidic channel hydrophobic. Therefore another approach is necessary to fix the microstructure to the glass substrate without surface treatment. In order to circumvent this issue we designed an anchored structure in a rectangular cavity in the fluidic wall. The two gaps on the wall fix the polymeric seal without chemical surface treatment for bonding enhancement. In the T-junction microfluidic channel, the microvalve squeezes the red dye stream and generates a droplet with temporal control. When the pneumatic valve is opened, the red dye goes into the oil stream and when the pneumatic valve is closed, the valve cuts the red dye stream and generates a droplet.

The integration of multilayered membrane-type microcomponents in a rectangular channel is useful for microfluidics devices requiring several microvalves using a rectangular channel fabricated with SU-8. The polymer seal alleviates the necessity for reflow processing, which may limit the channel dimension due to the selection of a positive photoresist. Additionally, the polymer seal approach can be used for non-PDMS microfluidic devices fabricated by the cutting method for rapid prototyping since the cross-section of a microfluidic device made using this technique has a rectangular shape. However, without an automated stage and optical setup it would be laborious to use the polymer seal technique for making arrays of PDMS membrane

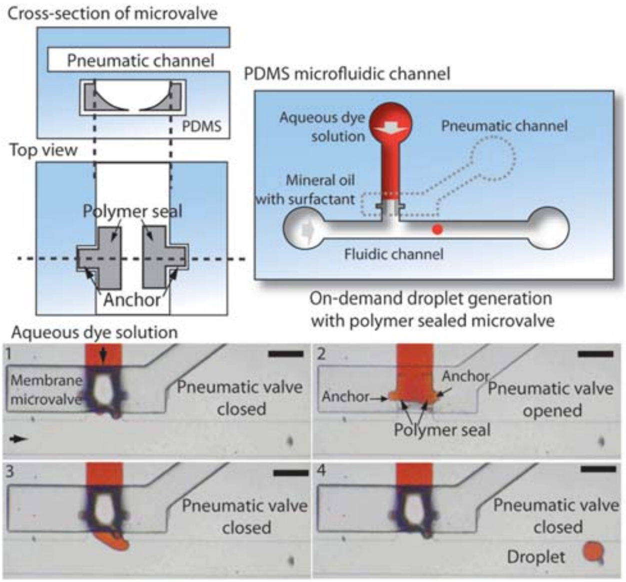

Fig. 5 On-demand droplet generation. In a T-junction microfluidic device, a polymer-sealed microvalve squeezes the red dye stream and generates a droplet with temporal control. When the pneumatic valve is opened, the red dye goes into the oil stream and when the pneumatic valve is closed, the valve cuts the red dye stream and generates a droplet (red dye phase flow rate: 1 μl min^{-1}, oil phase flow rate: 10 μl min^{-1}, pneumatic channel pressure: 15 psi). Scale bar: 200 μm.

microvalves since the polymer seals would need to be individually fabricated. Additionally the use of a polymer seal may cause particle accumulation in some cases because it interrupts flow. Despite these limitations, polymeric-sealed microvalves can be widely used for various applications requiring the use of a rectangular fluidic channel.

Experimental

Fabricating microfluidic devices

A soft lithography replica molding process is used for the fabrication of all PDMS-based microfluidic devices. We fabricated an SU-8 (Microchem Corp., Newton, MA) mold on a silicon wafer using photolithography. Trichloro-(1*H*,1*H*,2*H*,2*H*-perfluorooctyl)silane (Sigma aldrich) is vapor deposited on the wafer for easy removal of the subsequently poured PDMS (Sylgard 184, Dow Corning). After pouring, the PDMS is baked at 150 °C for 15 min before it is peeled off the mold. For fabrication of thin membranes for the fluidic channel layer, the mold is coated with PDMS and spun at 1000 rpm for 30 s and baked under the same conditions as above. Each PDMS layer is bonded after plasma surface treatment by an RF plasma generator (Cute-MP, Femtoscience). The height of the pneumatic microchannel and the membrane thickness were 40 μm, and 60 μm, respectively.

Chemical pretreatment for fixing the *in situ* polymerized seal

In order to enhance the bond between the polymer seal and the glass, 3-(trimethoxysilyl)propyl methacrylate with 2% of ethanol is pretreated on the glass substrate before fabricating the microfluidic device. After 15 min, the glass substrate is washed with 100% ethanol several times and baked at 100 °C on a hot plate for 1 min. Alternatively, this treatment can be done *in situ* after fabricating the microfluidic device.

Fabricating an *in situ* polymerized seal

Trimethylolpropane triacrylate (TMPTA, Sigma-Aldrich) mixed with 10 wt% of a photoinitiator (2,2-dimethoxy-2-phenylacetophenone) is introduced into the channel. Gas pressure (8 psi) is applied to the pneumatic channel to deform the membrane. To polymerize the prepolymer as a seal in the fluidic channel, a rectangular UV light pattern is illuminated on the deformed microvalve. The UV light pattern is generated using an optofluidic maskless lithography system. 365 nm UV light from a UV light source (LC8, Hamamatsu, 200 W) is used for photopolymerization. An objective lens (×10) is used to demagnify the UV light pattern. The exposure time is 500 ms and the UV illumination energy on the pattern is 340 mW cm^{-2}. Alternatively, a fluorescent microscope with an illumination system in the UV range can be used for generating arbitrary UV patterns by placing an arbitrary shaped transparent mask in front of the field stop of the microscope.[14] UV exposure time in alternative photolithography using conventional fluorescent microscopy (IX71, Olympus) is within 5 s. UV exposure time depends on the UV energy from the illumination source, which is longer than that of optofluidic maskless lithography system[15] since the mercury lamp (U-LH100HG, Olympus, 100W) is of lower power than the UV light source. After fabricating the seal, the channel is washed out with 1 ml of ethanol to remove TMPTA in the microfluidic channel, and dried it at 100 °C on the hot plate in order to remove the ethanol.

Conclusions

We presented a method to fabricate a PDMS membrane-type microvalve for rectangular microchannels using an *in situ* polymerized seal. We also demonstrated the equivalent functionality of our approach to curved channel microvalves by demonstrating a well functioning micropump and droplet generator.

Acknowledgements

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