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Citation: Journal of Applied Physics 117, 174904 (2015); doi: 10.1063/1.4919353
View online: http://dx.doi.org/10.1063/1.4919353
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Macroscopic strain controlled ion current in an elastomeric microchannel

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(Received 27 August 2014; accepted 17 April 2015; published online 6 May 2015)

We report on the fabrication of an ultra-high aspect ratio ionically conductive single microchannel with tunable diameter from \( \approx 20 \mu m \) to fully closed. The 4 mm-long channel is fabricated in a Polydimethylsiloxane (PDMS) mold and its cross-sectional area is controlled by applying macroscopic compressive strain to the mold in a direction perpendicular to the channel length. We investigated the ionic conduction properties of the channel. For a wide range of compressive strain up to \( \approx 0.27 \), the strain dependence of the resistance is monotonic and fully reversible. For strain \( > 0.27 \), ionic conduction suddenly shuts off and the system becomes hysteretic (whereby a finite strain reduction is required to reopen the channel). Upon unloading, the original behavior is retrieved. This reversible behavior is observed over 200 compression cycles. The cross-sectional area of the channel can be inferred from the ion current measurement, as confirmed by a Nano-Computed Tomography investigation. We show that the cross-sectional area decreases monotonically with the applied compressive strain in the reversible range, in qualitative agreement with linear elasticity theory. We find that the shut-off strain is affected by the spatial extent of the applied strain, which provides additional tunability. Our tunable channel is well-suited for multiple applications in micro/nano-fluidic devices. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4919353]

I. INTRODUCTION

Microfluidic and nanofluidic devices have become essential components for many applications in chemical and biomedical engineering research. Their key property is the ability to control ionic, fluid, and molecular flow at the micro and nanoscale. In microfluidic devices, dynamic chemical reactions can be manipulated and analyzed on a chip at the millimeter scale.1 These lab-on-a-chip techniques enable important functionalities, from capillary electrophoresis2 to drug delivery.3 With the development of nanotechnology, the transport can be controlled at the molecular scale and over sub-femtoliter volumes,4–6 leading to a range of nanofluidic devices,7–10 including nanofluidic diodes and transistors.11–14 Transport properties of nanochannels are often different from the properties of their microfluidic counterparts. Thus, studying systems which are able to convert between micro- and nano-scales can provide important insights into the size-effects that impact chemical and physical processes in nanodevices. In addition, characterizing the transition behavior in the simple structure can also facilitate formation of complex hierarchical systems that take advantage of the observed properties to produce both new material responses and interesting technological applications.

Aspect ratio and diameter of micro/nano-fluidic channels play an important role in the functionality of microfluidic and nanofluidic devices. Aspect ratio is defined as the ratio of channel length to channel diameter. There has been a great deal of interest in creating pores/channels with dynamically tunable opening diameter. Conically shaped nanopores fabricated in an elastomer film10 feature opening diameters that can be tuned from a few microns to a few tens of nanometers.15–22 This offers a versatile platform used, for example, as sensors for objects of different sizes. The membrane thickness for the conical tunable pores is 150–250 \( \mu m \) in almost all reports. Currently, there is no technology which allows for a cost-effective preparation of single pores/channels with both dynamically tunable aspect ratio and ultra high aspect ratio structures.23–25 Such ultra-high aspect ratio pores/channels could find application in a wide range of fields,26–28 including gas exchange and reaction in microchannels,29,30 cell growth,31 selective trapping of nanoparticles,32 and DNA linearization.33,34

Here, we present a simple method to fabricate single polydimethylsiloxane (PDMS) channels with tunable diameter in the range of \( \approx 20 \mu m \) to less than 1 \( \mu m \), and ultra-high aspect ratios reaching a few thousand. The paper is organized as follows: In Sec. II, we discuss the geometry and the fabrication method for the device. In Sec. III, the relation between ionic conductance and the compressive strain is characterized by experimental measurements and a

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theoretical linear approximation is presented. In Sec. IV, we further discuss the ionic current behavior and possible explanations for the results.

II. EXPERIMENTAL DETAILS

Single tunable microchannels were extracted from a polydimethylsiloxane (PDMS) mold. PDMS is a rubber-like non-linear viscoelastic material that has been widely used in microfluidics and other bio-related applications. Not surprisingly, the viscoelastic properties are frequency dependent. Nonetheless, it has been shown that the stress-strain response of PDMS is roughly independent of the strain rate in the range of 0.3 to 2.0 s⁻¹. In our experiment, we compressed and expanded our PDMS device at a constant rate of about 0.2 s⁻¹, which is fairly close to the frequency-independent range. For every strain increase of ≈ 7%, we let the material relax for 3 s. Although this dwell time is considerably shorter than the relaxation time for PDMS (which is of the order of 100 s), the decrease in the relaxation modulus over 100 s is only around 5%. This suggests that viscoelastic effects are not dominant in our experiment, and can be ignored. Even ignoring viscous effects, PDMS should be properly modeled as a non-linear elastic material. For the sake of obtaining a simple closed-form model, though, in this work, we ignored all material non-linearities and simplified the response to linear elasticity.

The channels were fabricated according to the following procedure. Two parallel polyacrylate (PLA) guiding fibers (purchased from Tenjin) of 300 μm in diameter, placed 4 mm apart, created electrolyte reservoirs. A single 20 μm fiber was attached perpendicularly to the guiding fibers and became the negative of the channel. We used 10:1 PDMS (Dow Corning) to fill the three-fiber mold. The PDMS curing was performed for an hour at 80°C. The guiding fibers were manually removed and the device was immersed into dioxane to dissolve the thin bridge fiber. Side and top views of the device together with an optical microscopy image of the cross-section of the channel is shown in Fig. 1.

Three clamps of 2 mm, 3 mm, and 4 mm width made out of translucent polycarbonate were used to apply compressive strains to the elastic channel. The notion of compressive strain is defined here in the engineering sense, i.e., as the change of thickness divided by the initial thickness of the device. The thickness was changed in steps of ≈ 37 ± 4 μm, imposed by a syringe pump (Kent Scientific). The initial thickness of the PDMS sample was 5.2 mm, and the range of applied strain varied from 0 to ≈0.3. The schematic of the mechanical setup is shown in Figs. 1(a) and 1(b). The deformation of the average cross-section of the channel under macroscopic compression applied in the y-direction was first observed by Nano-CT (see Fig. 1(d) for an applied compressive strain of 0.23). The Nano-CT scan is carried out using ZEISS Xradia 410 Versa, which combines high-performing X-ray microscopes and computed tomography (CT) systems. This allows for non-destructive 3D imaging. The microchannel was scanned using 20X magnification with a field of view of 948.6 μm × 948.6 μm, and a pixel size of 0.954 μm. During the scan process, the X-ray source of the Nano-CT is set at 50 kV and 12 W with a filter LE5, and the exposure time for each image is 40 s. Finally, the 3D image of the microchannel is reconstructed by 1601 2D images under a total 360° scan. We averaged and normalized the channel profile over the channel length of about 40 μm.

As expected, the circular cross-section becomes deformed into an elliptical shape under compression. The average channel area is estimated by attributing voxels with less than 50% normalized intensity to the channel in Fig. 1(d). The color code illustrates the normalized scan signal. At the strain level of 0.23, the estimated final cross-sectional area is about 27% of the cross-section of the initial channel.

Electrical conductivity in the channel as a function of compression was studied using 1 M KCl at pH 8. The experimental set-up is shown in Fig. 1(b). Two non-polarizable Ag/AgCl electrodes were used to apply voltage and measure the resulting ion current. The voltage was generated by a computer controlled DAQ device (NI USB-6009). In order to minimize charging effect, an alternating square voltage of ±5 V with 0.5 Hz frequency was used. The ion current was recorded with a multimeter (Keithley Series 2000). We tested the reproducibility of the channel properties by using the same device for over two hundred compression and expansion cycles. We observed no damage to the PDMS device after the completion of all 200 cycles. For any given cycle, difference between the electrical conductivity under compression versus expansion (except in the hysteretic regime) at any particular strain is less than 1.5%, and unless
noted, the symbols in all figures are larger than this error. Over the course of the 200 compressions, the electrical conductivity was reproducible from run to run, with a maximum random variation of 3\%, indicating a fully reversible material response. The device was cleaned by injecting the millipore water from one side of the guiding channel, allowing the clean water to flow through the center channel to remove the electrolyte. Then the device was sonicated in a clean vial with millipore water for 30 min. After cleaning, the device is stored in the millipore water and sealed in a clean vial.

III. RESULTS

Measurements of current-voltage curves for a single channel under different compressive strains ranging from 0 to 0.29 are shown in Fig. 2. As expected, an increase in strain leads to an increase in resistance, indicating a reduction in the cross-sectional area of the channel. At compressive strain close to 0.29, the channel conductance becomes unstable. We refer to this phenomenon as “shutting off” of the channel under compression.

The linear character of I-V curves for all strain magnitudes allowed us to characterize the channel by a well-defined ionic resistance. As an example of device behavior, Fig. 3(a) shows two consecutive loading-unloading cycles for different values of maximum applied external strain that were taken after approximately 180 cycles of the device. We illustrate the behavior in the strain range of 0 to 0.29. For compressive strains below 0.276, a completely reversible behavior was observed. However, when the maximum compression exceeded 0.29, the resistance-strain response become hysteretic as shown in Fig. 3(b). Above the critical strain, the resistance is not measurable due to the unstable nature of the current. After current shut off, no current flow was measured until the value of applied strain decreased to \( \approx 0.225 \), after which the microchannel recovered completely.

Aside from hysteresis, the resistance versus strain trend is attributed to the change in area of the microchannel as induced by the displacement field in the elastomeric matrix upon macroscopic compression. Assuming a linear elastic behavior for the PDMS matrix throughout the entire strain range, the deformation of the channel can be predicted by the well-known Eshelby solution for ellipsoidal holes embedded in linear elastic solids.\(^{39-41}\) Under uniform far-field compression in the y-direction, the initially circular channel deforms into an elliptical shape with aspect ratio \((a/b)\) increasing with strain. With any increment in strain \( \Delta \epsilon \) (assumed positive in compression), the vertical radius \( b \) of the ellipse decreases by an amount \( \Delta b = (1 + 2a/b)b\Delta \epsilon \), whereas the horizontal radius \( a \) increases by an amount \( \Delta a = a\Delta \epsilon \). As both \( a \) and \( b \) undergo significant variations.
over the strain range of interest, the area of the channel
$A/\Delta A_0 = 4ab/D^2$ (with $D$ the initial diameter of the channel) at a finite value of the applied strain $\epsilon$ should be obtained by integration, and is plotted as a solid curve in Fig. 4. As the strain approaches the value $\epsilon_c \approx 0.3442$, the area asymptotically approaches 0. An approximate close-form solution for the area can be obtained by ignoring the change in aspect ratio $(a/b)$ of the channel in the expression for $\Delta A$ above, resulting in

$$A/\Delta A_0 = (1 + \epsilon)(1 - 3\epsilon). \quad (1)$$

This equation (plotted as a red dashed curve in Fig. 4) becomes unphysical at strains in excess of 1/3, for which it predicts a negative channel area, but is a good approximation for the strain range of interest.

The resistance versus strain curves discussed above provide an indirect measurement for the cross-sectional area of the channel. The resistance of a channel filled with an electrolyte is calculated as $R = \rho \frac{L}{A}$, where $L$ and $A$ are the length and the cross-sectional area of the microchannel. The resistivity $\rho$ of 1 M KCl is 0.098 ($\Omega \cdot m$) at room temperature ($\approx 20^\circ C$). The system acts as two resistors in series, and we get

$$R = \rho \left( \frac{L_0 - l}{A_0} + \frac{1}{A} \right), \quad (2)$$

where $A_0$ and $A$ are the original cross-sectional area and the deformed cross-sectional area, and $L_0$ and $l$ are the total length of the channel and the length of the compressed part of the channel, respectively. For the experimental data plotted in Fig. 4, $L_0$ is equal to 4 mm and $l$ is equal to the clamp width. As the system resistance is measured from I-V curves, the variation of the cross-sectional area with strain can be obtained from Eq. (2). These measurements are plotted in Fig. 4 alongside the theoretical predictions discussed above. Obviously, the length of the compressed portion of the channel extends beyond the size of the clamp. Hence, the length $l$ in Eq. (2) should be interpreted as a lower-bound estimate. Edge effects are certainly present around the edge of the clamp but will be ignored here for the sake of simplicity.

Three key conclusions emerge: (i) the experimental trend is generally in good agreement with the analytical predictions, supporting the assumption that resistance changes with strain are primarily due to channel area reductions; (ii) the experimental data clearly show hysteresis behavior near the shut-off point, which might be due to stiction or elastic instabilities—clearly this phenomenon cannot be captured by the linear elastic model discussed above; (iii) the experimentally measured shut-off strain is smaller than predicted analytically, by $\approx 10\%$. Notice that the extraction of the cross-sectional area from resistance measurements assumes constant fluid conductivity and uniform compression along the channel length. The latter was verified by Nano-CT analysis: although the cross-sectional area fluctuated slightly along the channel (as can be observed by the broad distribution of the CT image), for an applied compressive strain of 0.23, the CT image shows an area that is $\approx 27\%$ of the original channel cross-section, in good agreement with resistance measurements (Fig. 4). The fact that the experimentally measured area is consistently lower than predicted (and hence the resistance constantly higher) is tentatively attributed to two possibilities: non-linear behavior in the response of the PDMS matrix and changes in the ionic flow (and hence the ionic resistivity, $\rho$) due to boundary effects nearby the edge of the clamp. Given that the Nano-CT measurements are in agreement with the conductivity based measurements at a strain of 0.23, we presume that any discrepancies for smaller strains are due to elastic effects and do not arise from the behavior of the ionic current. However, at higher strains, we expect some impact of the small channel diameter on the electrolyte flow, such as electro-osmotic effects or possible leaching of electrolyte into the PDMS. To better understand the behavior near shut-off, we convert the theoretical calculation for change in area to an effective resistance.

Combining Eqs. (2) and (1) provides an analytical estimate for the variation of ionic resistance with strain, i.e.

$$R = \frac{\rho L_0}{A_0} \left[ 1 + \frac{l}{L_0} \epsilon \left(1 + \epsilon(1 - 3\epsilon) \right) \right]. \quad (3)$$

This equation is plotted for three different values of $l/L_0$ in Fig. 5 alongside the experimental measurements. The curve of resistance versus compressive strain exhibits roughly two regimes excluding the shut-off behavior, as illustrated in Fig. 3(a). Initially, the response is relatively flat, but as the system approaches the shut off point, there is a sharp

![Normalized cross-sectional area of a single PDMS channel as a function of the compressive strain. The clamp width is 2 mm. The symbols in the plots indicate the compression (black open square) and expansion (blue solid triangle) up to the maximum strain of 0.276, and the compression (black open circle) and expansion (red solid inverse triangle) up to the maximum strain of 0.29, respectively. The cross-section was estimated based on Eq. (2), which describes a compressed channel as two resistances in series and normalized to the initial cross-sectional area of the microchannel. The area decreases almost linearly with the compressive strain. The blue solid curve is an Eshelby elasticity solution, obtained by applying small increments of strain until the area asymptotically approaches 0 (see text for details); the red dashed curve is an approximate linearized solution which is obtained from Eq. (1).](image-url)
increase in the slope of the resistance-strain plot. Therefore, the initial behavior of the resistance for small \( \epsilon \) is dominated by the \( \rho L_0/A_0 \) term, which is the resistance for the uncompressed channel. In this case, the resistance increases slowly with compression. As one approaches the singularity, the resistance as a function of strain diverges. This occurs around the shut-off strain. Therefore, the sensitivity of the resistance to the applied strain increases dramatically and allows for at least two distinct regimes of operation.

A surprising feature of the device is the dependence of the shut-off value on the spatial extent of the applied strain. Although most of the experimental trends and values are well captured by the model, the model predicts that the shut-off strain is equal to 1/3 and independent on the compressed length \( L \); whereas the experimental measurements consistently show a shut-off strain lower than 1/3 that decreases with increasing compressed length \( L \) \((\approx 0.3 \text{ for } L = 2 \text{ mm}, \approx 0.29 \text{ for } L = 3 \text{ mm}, \text{ and } \approx 0.27 \text{ for } L = 4 \text{ mm})\). The dependence of the shut-off strain with compressed length \( L \) indicates that there is non-trivial physics involved in the shut-off behavior. We are currently exploring a number of different candidates for the dependence of a critical shut-off strain on \( L \). One possibility is the role of boundary effects: as \( L \rightarrow L_0 \), the deformation extends to the channel boundaries, violating plane strain conditions. It is conceivable that the entrance and the exit of the channel deform more substantially than the center, adding a significant resistance term to Eq. (2). This would explain why the shut-off strain approaches the theoretical value as the compressed length \( L \) is reduced (Fig. 5). Another interesting possibility is the fluid-dynamics of the ionic solution in the channel. The difference of total channel length changes the spatial extent in which the fluid is highly confined, and therefore, impacts the role of any complexity of fluid dynamics arising in the highly confined geometry. Such effects are likely to be enhanced by the hydrophobic nature of the PDMS, so future work on changing the chemical composition of the channels will play an important role in understanding this system.

**IV. DISCUSSION AND CONCLUSIONS**

We have presented initial studies of a strain-controlled ionic device. The device has a number of interesting features. First, it is highly robust, exhibiting consistent electrical resistance over 200 cycles of compression-expansion. Second, despite exhibiting hysteretic behavior at high strain values, the electrical resistance is highly reproducible for low strain values as a function of compression and expansion. From an application point of view, the device exhibits at least three distinct regimes of electrical resistance: (i) a regime where the resistance is weakly dependent of the strain; (ii) a regime in which there is a strongly divergent dependence of electrical resistance on strain; (iii) a highly non-linear, hysteretic regime. The first two regimes can be understood in terms of a simple elastic model of channel area contraction upon far-field mold compression, whereas the hysteretic behavior requires further investigation.

In the hysteretic regime, there are several possible explanations for the resistance-strain relationship (Fig. 3). First, PDMS is known to be hydrophobic and thus it is possible that beyond the critical compressive strain the ionic solution is removed from the channel by the capillary force. Recovery of the ion current requires achieving a sufficient cross-sectional area under expansion that fluid can fill the channel again. Another possibility is that the channel mechanically adheres when its sides touch upon extreme compression, which requires expansion to re-open the channel. In fact, it is likely that both effects play a role to some degree. This potential interplay between fluid dynamics in nano-scale channels and mechanical properties of the surrounding matrix offer a wide-range of future design possibilities involving an interplay between mechanics, chemistry, and fluid dynamics.

The other intriguing feature of this device is the reproducible connection between the macroscopically applied strain and the detailed response at the local micro-channel scale, which allows accurate control of the channel size (and its ionic resistance) by means of deformation fields applied remotely (thousands of diameters away from the channel). Additionally, we have shown that varying the spatial extent of the applied strain has a direct impact on the features of the ionic resistance versus strain curves. This opens up the possibility for a wide range of applications, including the design of materials based on arrays of channels and spatially varying macroscopic strains.

It is important to note that there is a non-trivial relation between the spatial extent of the applied strain and the local deformation of the channel. In the simplest physical model, one would assume that the fraction of the channel which contracts, or more precisely the change in resistance as a function of the region which is strained, is directly proportional to the extent of the applied strain. We show that applying the simplest conductivity models as a means of computing area change and simple elastic modeling of the deformation are in disagreement. This points to the need for a better

**FIG. 5.** The measured resistance and the theoretical prediction (from Eq. (3)) of the device with three different clamp widths are shown. The symbols and curves in the plot indicate the experimental data and theoretical prediction for the clamp width of 2 mm (black square; black solid), 3 mm (red circle; red dashed), and 4 mm (blue triangle; blue dotted), respectively. The dependence of resistance on compressive strain is qualitatively similar for all three clamp widths, however, the shut off points shift to a higher value of strain for a smaller clamp width.
understanding the details of both the ionic flow in the small channels and the precise nature of the elastic deformations. We understand that the non-linear viscoelastic properties of PDMS may affect the characterization and operation of the device, especially when high-frequency response is desired. We will address this time-dependent material response in future studies. In addition, we will focus on simulations of the response of the material to applied strain and increased nano-CT studies of the channel deformation to better understand the impact of the spatial variation of applied strain on the resistance response as a function of strain. Such an understanding is critical for the design of more complex channel networks that can be selectively actuated by strategically located load patches on the device boundary.

In summary, the PDMS channel under compression is a highly tunable resistance element with three distinct response regimes. The resistance is essentially constant under a small amount of compressive strain; it increases steeply as the strain approaches the shut-off point, which can be tuned by controlling the dimension of the remote load patches; finally, upon further compression, the resistance behaves hysteretically under expansion. The non-trivial behavior of the resistance as a function of applied strain makes this system an ideal candidate for tunable circuit elements in ionic circuits, providing key elements for the ionic computing. In addition, the wide-range of tunable aspect ratios presents other interesting design possibilities. First, combining multiple channels with spatially varying applied strains could produce interesting hierarchal materials. And, because these are achieved with mechanical deformations of relatively large channels, this has a direct impact on the one of the major challenges of small-scale structure: resistance to fluid flow. With these materials, a system could be filled while the channels are relatively large, and then, the hierarchy of scale created by the applied strain once the system is filled. As discussed earlier, such application will require a better understanding of the hysteretic behavior, especially the relative importance of effects such as hydrophobicity and stiction.

ACKNOWLEDGMENTS

We acknowledge the support of UCI Seed Funding and Research Corporation. Professor Esser-Kahn acknowledges support from an AFOSR Young Investigator Grant under FA9550-12-10352, a 3 M Non-Tenured Faculty award, and an ACS PRF Award No 53493-DN for financial support. Professor Sun acknowledges NSF-CMMI-1229405. Professor Siwy acknowledges NSF-CHE-1306058. Professor Dennin acknowledges NSF-DMR-1309402.

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