A stretchable micrometer with enhanced performance for intermediate Reynolds numbers

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Abstract

Chemical reactions in microscale require good mixing at a relatively low flowrate. However, mixing in microscale faces the major challenge of stable laminar flow associated with the low Reynolds number, the relative ratio between inertial force and viscous force. For low Reynolds numbers of less than unity, mixing occurs due to molecular diffusion. For high Reynolds number of more than several tens, chaotic advection enhances mixing. However, in the intermediate regime, mixing is not efficient. This paper reports a stretchable micromixer with dynamically tuneable channel dimensions. Periodically stretching the device changes the channel geometry and the curvature induced secondary Dean flows. The dynamically evolving secondary and main flows in the mixing channel result in chaotic advection and enhance mixing. The concept was demonstrated in a stretchable micromixer with a serpentine channel. We evaluated the performance of this stretchable micromixer both experimentally and numerically. At the intermediate range of Reynolds numbers from 4 to 17, the periodically stretched micromixer showed a better mixing efficiency than the non-stretched counterpart. Therefore, our stretchable micromixer is a potential candidate for applications where precious reagents need to be mixed at relatively low flow rate conditions.

1. Introduction

Microfluidics is the science and engineering of handling and analysing a tiny amount of liquid for a broad range of applications in chemistry, food science, and medicine [1]. The small size of microfluidic devices brings advantages such as controllable reaction dynamics, high reaction efficiency and low cost [2]. Many microfluidic components and devices have been developed, examples are micromixers [3], microdispensers [4], micropumps [5], microvalves [6], and microfluidic sorters [7].

Micromixer is an indispensable component in miniaturised platforms for chemical, biochemical and biomedical applications such as drug delivery [8], detection and synthesis of DNA and RNA [9], enzyme assay [10], disease diagnosis [11, 12], bioreactors[13], and microreactors [14, 15]. These applications require thorough mixing of the reagents. Thus, a reliable micromixer is essential. However, since viscosity dominates the flow characteristics in microchannels, fluid flow is in the laminar regime of low Reynolds numbers ($Re < 1$) [16–18]. In a laminar flow regime, the fluids move alongside each other in parallel streams. The molecules diffuse from one fluid stream into the other by Brownian motion from the higher concentration to the lower concentration regions[19]. The diffusivity depends on the fluid viscosity, molecular size, and temperature. One of the solutions to enhance mixing is enlarging the interfacial area. Both passive and active strategies have been employed to increase the interface between the two fluids [20]. Active mixing techniques require external forces to disturb the fluid flows through mechanical, magnetic, electrical, thermal, or ultrasonic means [20]. Passive micromixers, on the other hand, rely on intrinsic fluid dynamics to stretch, fold, split, and recombine fluids to increase the interface between fluids [21, 22]. The performance of passive mixing is determined by the dimension, configuration, and geometry of the mixing channels. Passive micromixers with both two-dimensional planar structures [23–26] and three-dimensional structures [27–31] have been studied extensively.

Curved microchannels have been used for developing passive micromixers [26–27]. Introducing curvature in the flow direction induces centrifugal force, leading to the formation of a pair of circulating vortices perpendicular to the mainstream direction [32, 33]. These vortices, also called Dean vortices, result in subsequent chaotic advection, improving the mixing efficiency by folding the fluids [34, 35]. Following this mechanism, fluid mixing has been investigated in serpentine [36], spiral [37], square wave [38], zigzag [39], and convergent-divergent [40] channels. However, in all these past works, micromixers are rigid, and their geometries and dimensions cannot be tailored or adjusted on-demand and in real time. Thus, mixing efficiencies were evaluated only for stationary conditions. The fluid flow was steady, and the channel shape and dimensions were fixed. Currently, no existing works in the literature reported on the effects of dynamic and real-time modification of channel curvature and dimensions for enhanced mixing.

We recently proposed the concept for flexible and stretchable microfluidics, which enables tuning the channel dimensions after their fabrication [41–43]. Stretchability allows for elongating the microfluidic channel in a desired manner and accordingly changing its dimensions. In the present paper, we expand the concept of flexible microfluidics further and propose a stretchable micromixer with dynamically tuneable dimensions and evaluate its mixing performance. The geometry of the stretchable micromixer can be dynamically tuned by adjusting the stretching length and the frequency. Periodic deformation alters the dimensions of a stretchable serpentine channel. The coupled effects of the dynamic geometry changes lead to periodic and rapid transformation of the flow field, particularly of the Dean vortices, resulting in improved mixing. Figure 1 illustrates the concept of stretchable mixing. We first experimentally investigated the mixing performance of the stretchable micromixer under periodic elongations. Next, we performed numerical simulation to gain insight into the dynamic changes in the flow field and the induced Dean vortices. The numerical results agreed well with the experimental data. Our stretchable micromixer provides an innovative way to modify fluid mixing in microchannels in the intermediate regime, where both molecular diffusion and chaotic advection do not work.

2. Materials And Methods

2.1. Device fabrication and design

Standard photo lithography and soft lithography were used to fabricate the silicon mould and poly(dimethylsiloxane) (PDMS) microfluidic devices, respectively [44]. To start with, a 4-inch silicon wafer was spin-coated with SU-8 3050 (MicroChem Corp.) photoresist which was followed by soft baking. Next, the photoresist was selectively exposed to UV light through a photomask. Subsequently, the unexposed photoresist was dissolved by the developer, leaving behind the mould with the channel patterns. Following the fabrication of the mould, soft lithography was employed to make the thin stretchable channel layers. Liquid PDMS (Sylgard 184, Dow Corning), prepared by mixing the base and curing agent at a 10: 1 ratio was spread on the mould to replicate the channels. The PDMS layer was subsequently cured for 2 hrs at 75°C. Next, two thick PDMS slabs were bonded to the inlet and outlet regions of the thin channel layer after plasma treatment (PDC-32G-2, Harrick Plasma). These slabs were used to hold the tubing connected to the chip. The inlet and outlet holes were then punched manually. Finally, the PDMS

layer comprising the channel patterns with the thick slabs on it was plasma bonded to a plain PDMS layer.

Using the above steps, we designed and fabricated a stretchable micromixer with a curved serpentine mixing channel. The serpentine channel with 20 mixing units has a uniform width of 250 μm , and a height of 113 μm . The radius of curvatures is 312 μm . The total thickness of the stretchable microdevice is 1 mm in the mixing channel region and 5 mm at the inlet/outlet region.

2.2. Stretching platform

Fig. 2a illustrates the entire experimental setup for the stretchable micromixer. A custom-made stretching platform was designed and manufactured for automatic, unidirectional, periodic elongation of the micromixer, Fig. 2b. This platform is assembled on top of a poly (methyl methacrylate) (PMMA) sheet that can be fitted onto the stage of an inverted microscope for observation. A translation stage, driven by a hybrid stepper motor (JKM NEMA17 Two Phase 42 mm), is mounted on the platform for applying automated single-axis motion to the micromixer, Fig. 2b. This motor is controlled by a microcontroller board (Arduino), and can apply a periodic translational motion to dynamically stretch the micromixer along its length. The inlet and outlet areas of the stretchable device are firmly clamped by the PMMA holders so that no slip will happen during the stretching process, and the mixing unit (the channel section) can be stretched freely, Fig. 2c. The platform can stretch the micromixer along its length up to 4 mm with different frequencies at constant stretching velocity (triangle-wave displacement).

2.3. Experimental setup

The stretchable micromixer has two inlets for the introduction of the two fluids to be mixed. The first fluid is deionised (DI) water (Milli-Q® Direct 8 water purification system). The second is a 0.1% (w/v) solution of fluorescein sodium salt (Sigma-Aldrich) in DI water. The fluorescent solution and DI water were loaded in glass syringes and delivered into the microdevice using a flow-rate-controlled syringe pump (neMESYS, Centoni GmbH). We used a fluorescence inverted microscope (Nikon Eclipse Ti2) coupled with an LED illumination source (pE-4000, CoolLED) to visualise of the mixing behaviour of the fluorescent stream in the stretchable micromixer. A digital camera (Nikon, DS-Qi2) was mounted on the microscope for recording fluorescent images of mixing channels. The mixing behaviour was evaluated at the outlet region where the device oscillation does not impact the focusing plane of the camera.

2.4. Data analysis

The recorded images from both the experimental setup and the COMSOL simulations were analysed using MATLAB to evaluate the mixing performance quantitatively. For this purpose, the mixing index (MI) of the fluorescent solution in water was calculated as [45]:

$$MI = 1 - \sqrt{\frac{1}{N} \sum_{i=1}^N (I_i^* - \bar{I}^*)^2} \quad (1)$$

Where N is the total number of discrete samples taken (total number of pixels); $\bar{I}^*$ is the average of the normalised concentration intensity field; and I_i^* is the normalised concentration intensity for the i th pixel. The value of mixing index varies from 0 for the unmixed condition (binary mixture) to 1 for complete uniform mixing. This formula was employed over all pixels of the selected frame. The reported mixing indexes in this paper have been measured based on one period of elongation.

2.5. Numerical modelling

We further studied the effect of periodic elongation on the fluid flow and mixing performance of the stretchable micromixer using numerical simulation. The three-dimensional (3D) numerical model of the stretchable serpentine microfluidic chip was formulated in COMSOL Multiphysics 5.6 software. The governing equations for the physics of solid deformation, Navier-Stokes equations for the incompressible fluid with no external forces, and the diffusion equation for mass transport were solved numerically.

The whole process of periodic elongation, fluid flow, and mixing were modelled with three different physics provided in COMSOL: *Solid Mechanics*, *Single-Phase Laminar Flow*, and *Transport of Diluted Species*. *Moving Mesh* was also applied to the model for the time-dependent simulation. Water, as the input flow, was considered as a Newtonian incompressible flow with the following properties: $\rho = 1,000 \text{ kg m}^{-3}$, $\mu = 10^{-3} \text{ kg s}^{-1} \text{ m}^{-1}$. The concentration of the dye was set at 0 mol/m^3 for the two side inlets and at 1 mol/m^3 for the middle inlet. The diffusion coefficient of the fluorescent dye was set at $10^{-9} \text{ m}^2 \text{ s}^{-1}$. The boundary conditions were no slip and no flux for the walls and zero pressure at the outlet.

3. Results And Discussion

3.1. Fluid mixing in the stretchable micromixer with serpentine channel

As mentioned previously, most micromixers perform well only at either very low or high Re . However, several micromixing applications are slow processes associated with Re ranging from 1 to several tens [46]. The dominant mixing mechanism at low Re is molecular diffusion, which requires a long residence time and correspondingly a long channel. By introducing curves, chaotic advection can be generated and dominate the molecular diffusion in the high Re range, leading to better mixing. As such, various micromixer designs have been proposed with serpentine channels with zigzag, sinusoidal, and square wave forms. These micromixers work well at relatively high Reynolds numbers ($Re \gg 10$) but are not very efficient at the intermediate Reynolds numbers (approximately $1 < Re < 10$) [13, 14, 46–48]. Here, we

demonstrate that dynamic elongation can improve the mixing efficiency of a stretchable micromixer with a serpentine channel operated at this critical intermediate range of Reynolds numbers.

We fabricated a simple curved serpentine micromixer with a uniform width and two flow-focusing inlets, Fig. 1. The micromixer has in total 20 curved mixing units. To evaluate the mixing performance of the stretchable serpentine micromixer, the fluorescent solution was delivered through the centre inlet, with DI water introduced through the two side inlets. The flowrates of the fluorescent solution and DI-water were adjusted with a constant ratio of 1:6 to vary the Reynolds number from 0.09 up to 36. This flow rate ratio remains fixed across all Reynolds numbers under investigation. We selected a non-equal flowrate ratio because it is more representative of real chemical mixing applications. Figure 3 shows the mixing index as a function of Reynolds number Re without oscillation and at three representative oscillation frequencies. As expected, the micromixer exhibited good mixing performance. For Reynolds number of less than ~ 4 , good mixing was achieved due to the long residence time of the fluids, allowing for molecular diffusion, while for Reynolds number greater than ~ 17 the good mixing can be attributed to chaotic advection. However, the performance in the intermediate range $4 < Re < 17$ is poor, since neither mixing mechanism has a significant effect. We demonstrated that periodic elongation improved the mixing index for this low Re range. The best improvement was observed for 1-Hz stretching, which increased the mixing index by as much as 0.2 as compared to without stretching. Increasing the frequency beyond 1 Hz slightly decreases the performance, but it is still much better than without oscillation. We hypothesize that this improvement in mixing performance is attributed to the disturbance caused by the oscillation, as well as the dynamic changes of the microchannel dimensions. Under elongation, not only does the channel length increase, but the cross section also shrinks [41]. These rapid changes in the geometry of the stretchable micromixer induce disturbance to the flow, resulting in an improved mixing. The dynamic changes affect the flow in different ways, including generating variations in the flow velocity at different locations of the microchannel. This could potentially lead to the continuous transformation of Dean vortices and create more chaotic advection in the flow field.

To test the above hypothesis, we investigated the changes in channel dimensions including the height, width, and the radius of the curvature both experimentally and numerically. Figure 4(a-b) show respectively the actual and simulated deformation of the mixing channel for different stretching lengths. Stretching the entire micromixer device along its length alters the width of the channel non-uniformly. The width parallel to the stretching direction, the horizontal width W_h , increases, Fig. 4c. In contrast, the width perpendicular to the stretching direction, the vertical width W_v , decreases, Fig. 4c. Figure 4c compares the simulated channel deformation to the experimental data, and shows a good agreement. The simulation results also showed that the height of the channel reduced slightly, Fig. 4d. An additional change to the geometry was that the radius of curvature altered significantly under stretching, Fig. 4e. Therefore, under oscillatory elongation, the channel dimensions and the cross-sectional area of the channel vary periodically. At a constant total flow rate, periodic change of the cross section disturbs the flow field, leading to chaotic advection and promoting mixing.

3.2. Numerical simulation of mixing in the stretchable micromixer with serpentine channel

In the above section, we demonstrated experimentally and numerically that stretching leads to change of cross section and curvature. Here, we hypothesise that the geometry change will affect the flow field as well as the Dean vortices at different locations of the mixing channel. To test this hypothesis, we carried out a numerical simulation to model the complex mixing process with a time-dependent 3D model. Figure 5 shows the velocity magnitude in the microchannel at the zero-stretch condition ($t = 1$ s, Fig. 5a) and maximal stretch condition ($t = 1.5$ s, Fig. 5b). Figures 5(c-h) show the close-up images of the channel curvatures at different times from zero-stretch ($t = 1$ s) to maximum stretch ($t = 1.5$ s). The results indicate that the velocity of the fluid increases during the stretching process. Thus, the axial velocity has varying values along the mixing channel, with the highest value while passing through W_v and the lowest value while passing through W_h . As the fluid passes through the device, it encounters continuous changes in flow velocity, which disrupts the stable flow and hence promotes mixing.

Another critical factor affecting mixing is the changes in Dean vortices generated in the curvatures. Dean vortices are secondary flows that form in a curved channel due to inertial forces. Dean vortices promote mixing by inducing transversal transport and chaotic advection. We studied the variation of Dean vortices numerically at different times during the periodic elongation. Figure 5c shows the simulation results. Dean vortices altered in shape and magnitude from the initial condition (zero-stretch, $t = 1$ s) to maximum stretch condition ($t = 1.5$ s). Under the periodic elongation, the deformation of the channel cross-section and the corresponding dynamic changes in the axial flow led to changes of the Dean vortices.

Finally, we compared the numerical mixing performance with experimental data. The flow rates were 10 $\mu\text{L}/\text{min}$ for the dye stream in the centre and 60 $\mu\text{L}/\text{min}$ for the side inlets, which correspond to a Re of 6.43. Figure 6a shows the experimental images of the outlet section of the serpentine channel. Figure 6b shows the simulated results at the same channel locations and flow conditions as the experimental results. We can see that the numerical results are consistent with the experimental data. In addition, the quantitative mixing index from both the experimental and numerical studies agree relatively well, Fig. 6c. These results demonstrate that the fluid mixing can be enhanced by oscillatory stretching.

4. Conclusion

This paper reports the concept of a stretchable micromixer with a serpentine channel design. By periodically stretching the device along its length, we demonstrated an improvement in the mixing performance at intermediate Reynolds number range. The improvement in mixing performance can be attributed to periodic changes in the geometry and cross section of the micromixer, affecting the main flow field and the secondary Dean vortices. These coupled effects lead to disruption of the stable fluid flow and enhance mixing. We verified that this is the mechanism of mixing improvement via both experiment and numerical studies. This proof-of-concept investigation demonstrates that tuneable

micromixers can be achieved. The results show a way of improving the efficiency of existing micromixers operating at intermediate regime, between diffusive mixing and chaotic advection, which is important for many practical applications.

Declarations

Declaration of competing interest

The authors declare no competing financial interests.

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Figures

Figure 1

The concept of the stretchable micromixer. (a) Micromixer with no elongation; (b) Micromixer at maximum elongation; Dean vortices at the AB plane under (c) the non-stretched condition; and under (d) the stretched condition.

Figure 2

(a) Experimental setup including the inverted microscope, syringe pump, and Arduino board; (b) Custom-made automated stretching platform with a translation stage driven by a hybrid stepper motor moving the PMMA holder. (c) The stretchable micromixer with its inlet and outlet areas firmly clamped by PMMA holder.

Figure 3

Mixing index as a function of Re for the stretchable micromixer at different frequency conditions. The inset shows an elongated micromixer with the window for evaluation of mixing index.

Figure 4

Changes in channel width and radius of curvature obtained from (a) experiments and (b) numerical simulation. (c) Changes in Wh (horizontal width) and Wv (vertical width) while being stretched, acquired from experiments and simulation. (d) Changes of the height obtained from simulation. (e) Changes in the radius of curvature of the serpentine channel measured using ImageJ.

Figure 5

Numerical simulation of the stretchable micromixer: (a) Velocity magnitude of the 3D dynamic simulation of the stretchable serpentine micromixer at (i) the initial condition (zero-stretch), $t = 1\text{ s}$ and (ii) maximum stretch condition, $t = 1.5\text{ s}$; (b) Close-ups of the curved channel sections at different times during the oscillation. (c) Cross-sectional streamlines of the secondary flow during the stretching process.

Figure 6

A comparison of (a) the experimental and (b) the numerical results of mixing performance of the stretchable serpentine micromixer for a flowrate ratio of 10/60 $\mu\text{L}/\text{min}$ equivalent to $\text{Re}=6.43$. (c) Mixing index values of the corresponding images in (a) and (b).