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2006

Nguyen, N. T., & Huang, X. Y. (2006). Modeling, fabrication and characterization of a polymeric micromixer based on sequential segm. *Biomedical Microdevices*, 8(2), 133-139.

<https://hdl.handle.net/10356/93878>

<https://doi.org/10.1007/s10544-006-7708-4>

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Modelling, Fabrication and Characterization of a Polymeric Micromixer Based on Sequential Segmentation

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Abstract. Effective and fast mixing is important for many microfluidic applications. In many cases, mixing is limited by molecular diffusion due to constraints of the laminar flow in the microscale regime. According to scaling law, decreasing the mixing path can shorten the mixing time and enhance mixing quality. One of the techniques for reducing mixing path is sequential segmentation. This technique divides solvent and solute into segments in axial direction. The so-called Taylor-Aris dispersion can improve axial transport by three orders of magnitudes. The mixing path can be controlled by the switching frequency and the mean velocity of the flow. Mixing ratio can be controlled by pulse width modulation of the switching signal. This paper first presents a simple time-dependent one-dimensional analytical model for sequential segmentation. The model considers an arbitrary mixing ratio between solute and solvent as well as the axial Taylor-Aris dispersion. Next, a micromixer was designed and fabricated based on polymeric micromachining. The micromixer was formed by laminating four polymer layers. The layers are micro machined by a CO₂ laser. Switching of the fluid flows was realized by two piezoelectric valves. Mixing experiments were evaluated optically. The concentration profile along the mixing channel agrees qualitatively well with the analytical model. Furthermore, mixing results at different switching frequencies were investigated. Due to the dynamic behavior of the valves and the fluidic system, mixing quality decreases with increasing switching frequency.

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1. Introduction

While mixing at the macroscale relies mainly on chaotic advection and turbulence induced by stirrers, mixing at the microscale is constrained by laminar flow behavior inside microchannels. Because of their significance in many lab-on-a-chip applications, micromixers have been the focal point of microfluidic research in recent years (Nguyen, 2005). A number of passive and active techniques exist for improving mixing at the microscale. One of these techniques is reducing the mixing path between solvent and solute. The two established methods for reducing the mixing path are parallel lamination and sequential lamination (Nguyen, 2005). These lamination techniques keep solvent and solute in their particular streams. Mixing path is shortened perpendicularly to the axial flow direction. Another method, sequential segmentation, divides the solvent and solute into segments which occupy the whole channel width. Mixing occurs through dispersion in flow direction. Sequential segmentation can be achieved by alternate switching of the inlet flows, using controlled valves for instance, Fig. 1. The mixing ratio can be adjusted by the switching ratio. Because of the need of external actuators, micromixers based on sequential segmentation are categorized as active micromixers.

Deshmukh et al. (Deshmukh, 2000) described the concept of sequential segmentation numerically and observed the mixing results optically. The switching process was controlled by integrated micropumps. Similar concepts realized by external pumps were reported later by Fujii et al. (Fujii, 2003) and Okamoto et al. (Okamoto, 2004). Recently, Tan et al. (Tan, 2005) reported a glass/PDMS micromixer based on the same concept. All previous works only reported numerical simulation and experimental observation. In this paper, we first describe the mixing concept with an analytical model, which allows a better understanding of this mixing concept and the significance of Taylor-Aris dispersion. Next, the paper reports the low-cost fabrication process of a polymeric micromixer using laser micromachining and hot lamination. Our fabrication approach allows fast and low-cost prototyping of polymeric microfluidic devices. Mixing results are evaluated by an optical measurement setup, which can freeze and capture a relatively large transient concentration field with a relatively slow and lowly resolved CCD-camera. Finally, experimental results are discussed and compared to the analytical solution.

2. Device Concept and Modelling

The micromixer described in the paper is fabricated by lamination of several polymer sheets. Thus, the mixing channel has a relatively low aspect ratio ($H \ll W$), where W is the channel width and H is the channel height. Consequently, the mixing channel can be modelled by a parallel-plates model. The model assumes a uniform velocity profile in the channel width direction (z -axis) and a Poiseuille velocity profile in the channel height direction (y -axis). With non-slip boundary conditions, the velocity distribution

between two parallel plates is described as (Bird, 2002):

$$u(y) = 6U \left(1 - \frac{y}{H}\right) \frac{y}{H} \quad (1)$$

where U is the mean velocity in the flow direction along the x -axis, Fig. 2. The parabolic velocity profile described in (1) leads to an effect called the Taylor-Aris dispersion, which is characterized by the effective diffusion coefficient D^* . Using the Taylor-Aris approach (Aris, 1956) and the velocity profile (1), the effective diffusion coefficient in flow direction can be determined as (Brenner, 1993):

$$D^* = D + \frac{H^2 U^2}{210D} \quad (2)$$

where D is the molecular diffusion coefficient of the solute in the solvent and H is the channel height. In our case, (2) is only true if the position of x is long enough for completing diffusion across the channel height ($x \gg UH^2/D$). With $H = 100 \mu\text{m}$, a flow rate of 2 ml/hr or an average velocity of 5.6 mm/s, and a diffusion coefficient of $1.8 \times 10^{-9} \text{ m}^2/\text{s}$, the condition for (2) would be $x > 30.9 \text{ mm}$, which can not be met near the channel entrance. At this velocity, the ratio between the effective diffusion coefficient and the molecular diffusion coefficient is $D^*/D = 455$. In fact, three orders

Figure 1. Mixing concept based on serial segmentation.

Figure 2. Model of the mixing channel.

of magnitude in axial transport can be reached by Taylor-Aris dispersion. The second term in (2) is dominant in most practical cases. The higher the velocity or the smaller the channel height, the better is axial dispersion. Flat micro channels $H \ll W$, as in the case of devices made by hot lamination, suit best for this purpose. This unique effect can be well utilized for fast mixing at high flow rates.

Using this effective diffusion coefficient D^* , the problem of sequential segmentation is reduced to a simple one-dimensional macro transport model. In this model, the concentration distribution across the channel width and height is assumed to be uniform. Thus, only the concentration profile along the flow direction x needs to be considered. With a mean velocity U and a switching period T , the characteristic segment length is defined as $L = UT$. The general transport equation can be then reduced to its transient one-dimensional form:

$$\frac{\partial c}{\partial t} + U \frac{\partial c}{\partial x} = D^* \frac{\partial^2 c}{\partial x^2} \quad (3)$$

Figure 3(a) depicts this one-dimensional model with the boundary condition at the inlet. Figure 3(b) describes the periodic boundary condition for the inlet ($x = 0$):

$$c(t, 0) = \begin{cases} c_0 & 0 \leq t \leq \alpha T/2 \\ 0 & \alpha T/2 < t \leq T - \alpha T/2 \\ c_0 & T - \alpha T/2 < t \leq T \end{cases} \quad (4)$$

where c_0 , T , and α are the initial concentration of the solute, the switching period, and the pulse width ratio (or the mixing ratio), respectively.

Introducing the dimensionless variables $c^* = c/c_0$, $x^* = x/L$ and $t^* = t/T$, equation (3) has the dimensionless form:

$$\frac{\partial c^*}{\partial t^*} = \frac{1}{\text{Pe}} \frac{\partial^2 c^*}{\partial x^{*2}} - \frac{\partial c^*}{\partial x^*} \quad (5)$$

where the Peclet number is defined as $\text{Pe} = UL/D^*$. This Peclet number represents the ratio between convective transport and dispersion in flow direction. A large Peclet number indicates, that convection dominates over dispersion. Using the dimensionless variables defined above, the dimensionless boundary conditions of equation (4) is:

$$c^*(t^*, 0) = \begin{cases} 1 & 0 \leq t^* \leq \alpha/2 \\ 0 & \alpha/2 < t^* \leq 1/2 \end{cases} \quad (6)$$

Solving the dimensionless governing equation (5) using separation of variables and the boundary conditions at the inlet (6) as well as ($c^*(\infty) = \alpha$) results in the transient behavior of the dimensionless concentration profile along the mixing channel:

$$c^*(x^*, t^*) = \alpha + \sum_{n=1}^{\infty} \frac{2 \sin(\alpha \pi n)}{\pi n} \Re \left\{ \exp \left[\frac{1}{2} \left(\text{Pe} - \sqrt{\text{Pe}^2 + 8\pi n \text{Pe} j} \right) x^* \right] \right. \\ \left. \times \exp(2\pi n t^* j) \right\} \quad (7)$$

where j is the imaginary unit, and $\Re$ indicates the real component of a complex number. The concentration distributions along the mixing channel at different Peclet numbers and a mixing ratio of $\alpha = 1/2$ is depicted in Figure 4, which shows clearly that only a short mixing channel is required if the Peclet number is small. A small Peclet number

Figure 3. The transient one-dimensional model of a micromixer based on sequential segmentation: (a) Concentration profile as function of time t and axial position x , (b) time signal of the concentration at the inlet.

can be achieved either by a small mean velocity U or a short characteristic segment length L . A short segment length in turn is achieved by a short switching time T or a high switching frequency $f = 1/T$. Sequential segmentation can achieve different final concentrations simply by adjusting the switching ratio α .

3. Device Fabrication

The micromixer described in this paper was implemented by polymeric micromachining. The device was formed by lamination of four polymer sheets, which have a thickness of $100\ \mu\text{m}$. The polymer sheets are commercially available as lamination porches. The four device layers are depicted schematically in Fig. 5. The lamination process was carried out using a commercial hot laminator (Aurora LM-450HC, laminating temperature of $120\ ^\circ\text{C}$, laminating speed of $0.3\ \text{m/min}$, maximum laminating thickness of $600\ \mu\text{m}$). The channels and other structures was designed on a CAD program and transferred to the polymer sheets by a laser cutting machine. We used the Universal M-300 Laser Platform (Universal Laser Systems Inc.) with a maximum CO_2 -laser power of $25\ \text{Watt}$

Figure 4. Concentration profile along the mixing channel at different Peclet numbers ($\alpha = 0.5$).

Figure 5. The four layers of the polymeric micromixer (layer 1: bottom layer for optical access, layer 2: mixing channel layer, layer 3: valve chamber layer, layer 4: top layer for fluidic interconnects).

and a maximum beam speed of about 640 mm/s. This technique results in a mixing channel with a cross section of approximately $100 \mu\text{m} \times 1000 \mu\text{m}$.

After laminating the four device layers, fluidic interconnects are glued on the inlets and the outlet using two-components epoxy. Piezo discs are attached on top of the circular valve chambers. Commercially available piezo discs were selected as actuators. These piezo discs are normally used as buzzers in electronic gadgets. The piezo disc

Figure 6. The fabricated micromixer).

consisted of a piezoelectric ceramic layer and a brass disc. The ceramic layer is glued on the brass disc. The active valves used the brass disc directly as the valve seat. The brass disc had a diameter of 15 mm and a thickness of 95 μm . The piezoelectric layer had a diameter of 12 mm and a thickness of 175 μm . With a maximum drive voltage of 200 V, the maximum electrical field strength was 1.1 kV/mm which was lower than the break down field of most common piezoelectric materials (> 2 kV/mm). Figure 6 shows the completed device without the epoxy and the electrical interconnects to the piezo discs.

A switching circuit based on an electromagnetic relay was designed for controlling the piezo discs. The circuit uses square-wave signals as the control input. A constant DC voltage of 150 V is supplied to the circuit and switched between the two piezo discs. The two switches are provided by a miniature electromagnetic relay (Fujitsu, Takamisawa RY5W-K). The complete switching circuit is depicted in Fig. 7.

4. Experimental Results

Two identical syringes were filled with diluted dye and DI water and placed on a syringe pump (Cole-Parmer 74900-05, 0.2 $\mu\text{l/hr}$ to 500 ml/hr, accuracy of 0.5%). The identical syringes ensure the same flow rates for the two inlet flows. The switching ratio or the mixing ratio was kept constant at ($\alpha = 1/2$). The control signal (Fig. 7) is a square wave and comes from a signal generator. The high voltage is kept at 150 V. Thus, the supply voltages to the discs are switched between 0 V and 150 V, opening and closing the two mixer inlets.

A measurement setup with an inverted fluorescent microscope (Nikon, ECLIPSE TE2000-S) was used. The measured area was illuminated with a mercury lamp. An epi-

Figure 7. Switching circuit for controlling the inlet valves.

fluorescent attachment was used (Nikon B-2A, excitation filter for 450-490 nm, dichroic mirror for 505 nm and an emission filter for 520 nm). The solute in this experiment is a fluorescent dye (fluorescein disodium salt $C_{20}H_{10}Na_2O_5$ diluted in water). This dye is also called Acid Yellow 73 or C.I. 45350. The diffusion coefficient of this dye was determined in our previous works as $D = 1.8 \times 10^{-9} \text{ m}^2/\text{s}$ [9]. Two identical syringes were filled with the dye solution and the DI water and placed on a syringe pump. The measurement reported in this paper was carried out with a $2\times$ objective. An interline transfer CCD camera (Sony ICX 084) was used for recording the images. The resolution of the camera is $640 \text{ pixels} \times 480 \text{ pixels}$, with a depth of 12 bits gray scale. Since the CCD sensor size is $6.3 \text{ mm} \times 4.8 \text{ mm}$, the size of an image pixel is $4.95 \mu\text{m}$ and the size of the measured area is $3096 \mu\text{m} \times 2376 \mu\text{m}$. For good contrasts, the exposure time was kept at 3 ms.

The switching signal of the valves on the mixer was used to trigger the camera's shutter. Since the switching process is periodic, this triggering scheme is able to freeze the intensity images at an arbitrary time within the switching period. Thus, images at a fixed time can be taken along the mixing channel and joined together in a big mosaic image. Thus, the relatively low resolution of our camera still allows detailed measurement of the concentration field along the mixing channel. Figure 8 shows the intensity images at different switching frequencies. The rectangle indicates the actual captured image size. At a flow rate of 2 ml/hr, the Reynolds number is 0.622, which clearly indicates that the flow is laminar. The fact that images taken by the trigger signal are identical proves that no turbulence and chaotic advection are involved in this mixing concept. If both valves are opened ($f = 0 \text{ Hz}$), the two streams flow side by side. The high Peclet number based on channel width $Pe_W=6173$ at a flow rate of 2 ml/hr indicates that transversal mixing is not possible. The higher convective

transport dominates over transversal diffusion, Fig. 8(a). The Peclet number based on axial transport can be three order of magnitudes smaller than Pe_W , when the inlets are switched. The Peclet numbers decrease from 37.7 at $f = 1$ Hz to 7.54 at $f = 1$ Hz. These low Peclet numbers are caused by Taylor-Aris dispersion, which improves mixing significantly. From the results in Fig. 8, one can observe that the valves can not be fully closed at higher frequencies. This effect will be discussed later.

The measured intensity of the pixel I as shown in Fig. 8 is normalized against the maximum $I_{\max}$ and minimum $I_{\min}$ of the intensity at the inlets. The measured dimensionless intensity is then assumed to be equal to the dimensionless concentration of the fluorescent dye:

$$c^* = I^* = \frac{I - I_{\min}}{I_{\max} - I_{\min}} \quad (8)$$

Figures 9 and 10 compare the theoretical and measured concentration profile along the mixing channel. The profile is taken at the centerline along the mixing channel. The qualitative agreement confirms the low Peclet number and the contribution of Taylor dispersion.

The probability density function (PDF) of the captured images can be used for evaluating the mixing quality. The PDFs in Fig. 11 show the distribution of normalized concentration values at the channel end. If there are two peaks at $c^* = 0$ and $c^* = 1$, solvent and solute do not mix. A PDF with a single peak indicate good mixing due the existence of a single concentration value. The sharper the peak, the better is the mixing quality. Figure 11(a) shows clearly that the solvent and solute do not mix if both valves are opened ($f = 0$ Hz). Best mixing was achieved with a switching frequency of 2 Hz, where axial Peclet number is reduced to only $Pe=18.9$, Fig. 11(b). The PDF shows a peak, which represents the homogeneity of the captured area. However, this peak broadens at higher frequencies indicating worse mixing (Fig. 11(c) to 11(f)). The effect shown in Figures 11(c) to 11(f) can be explained by the dynamic behavior of the valves. At too high frequencies, the valves cannot be fully closed making serial segmentation ineffective.

5. Conclusions

This paper reports the theory, fabrication and characterization of a micromixer based on sequential segmentation. The analytical model presents the time-dependent one-dimensional concentration distribution along the mixing channel. The analytical model shows, that mixing in axial direction can be improved by three orders of magnitudes due to Taylor-Aris dispersion. The micromixer was fabricated by laminating four polymeric sheets, which are micromachined by CO_2 laser. Hot lamination bonds all four polymeric layers together and forms the mixing channel as well as the valve chamber. The active valves at the inlets were actuated by two piezo discs, which are driven by a switching circuit. Experiments were carried out with a diluted solution of fluorescent dye. Experimental results agreed qualitatively well with the prediction from the analytical

Figure 8. Intensity images at different switching frequencies (flow rate 2 ml/hr, $Re=0.622$, the rectangle indicates the actual captured image): (a) $f = 0$ Hz, $Pe_W=6173$; (b) $f = 1$ Hz, $Pe=37.7$; (c) $f = 2$ Hz, $Pe=18.8$; (d) $f = 3$ Hz, $Pe=12.6$; (e) $f = 4$ Hz, $Pe=9.43$; (f) $f = 5$ Hz, $Pe=7.54$.

Figure 9. Normalized concentration profile at the centerline of the channel as indicated in Fig. 8(b) ($f = 1$ Hz, $Re=0.622$, $Pe=37.7$).

Figure 10. Normalized concentration profile at the centerline of the channel as indicated in Fig. 8(c) ($f = 2$ Hz, $Re=0.622$, $Pe=18.8$).

model. However, the effective diffusion coefficient from Taylor's theory overestimates the actual coefficient near the channel's entrance. Due the dynamic characteristics of the valves, higher switching frequency does not mean a better mixing quality.

Acknowledgments

This work was supported by the academic research fund of the Ministry of Education Singapore, contract number RG11/02. N. T. Nguyen would like to thank Mr. Wang

Figure 11. Probability density functions (PDF) of the captured image at the channel end (flow rate 4ml/min, $Re=1.24$): (a) $f = 0$ Hz, $Pe_W=6173$; (b) $f = 2$ Hz, $Pe=18.9$; (c) $f = 4$ Hz, $Pe=9.44$; (d) $f = 6$ Hz, $Pe=6.30$; (e) $f = 8$ Hz, $Pe=4.72$; (f) $f = 10$ Hz, $Pe=3.78$.

Cheng for his assistance during the characterization experiments.

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