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2004

Wu, Z. G., & Nguyen, N. T. (2004). Convective-diffusive transport in parallel lamination micromixers. *Microfluidics and Nanofluidics*, 1(3), 208-217.

<https://hdl.handle.net/10356/79641>

<https://doi.org/10.1007/s10404-004-0011-x>

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Convective–diffusive transport in parallel lamination micromixers

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Abstract

Effective mixing and a controllable concentration gradient are important in microfluidic applications. From the scaling law, decreasing the mixing length can shorten the mixing time and enhance the mixing quality. The small sizes lead to small Reynolds numbers and a laminar flow in microfluidic devices. Under these conditions, molecular diffusion is the main transport effect during the mixing process. In this paper, we present complete 2D analytical models of convective–diffusive transport in parallel lamination micromixers for a binary system. An arbitrary mixing ratio between solute and solvent is considered. The analytical solution indicates the two important parameters for convective–diffusive transport in microchannels: the Peclet number and the dimensionless mixing length. Furthermore, the model can also be extended to the mixing of multiple streams—a common and effective concept of parallel mixing in microchannels. Using laser machining and adhesive bonding, polymeric micromixers were fabricated and tested to verify the analytical results. The experimental results agree well with the analytical models.

Keywords

Micromixer · Mass transfer · Diffusion · Parallel lamination · Fluorescent measurement

1 Introduction

Over the past few years, numerous microfluidic devices and systems have been developed for chemical and biochemical applications. Micromixing, as an indispensable process in microfluidic systems, becomes a challenge for the microfluidic community. In most microfluidic devices, the small Reynolds numbers lead to the laminar flow. Thus, conventional methods in the macroscale for enhancing mixing become less effective in the microscale.

A number of works have been carried out on micromixers, which can be categorised as active mixers and passive mixers [1]. The parallel lamination mixer belongs to the passive type. Parallel lamination splits the inlet streams into n substreams, and then joins them into one stream as laminae. The basic design is a long microchannel with two inlets ($n=2$). According to its geometry, this design is often called the T-mixer [2, 3] or the Y-mixer [4]. These basic devices lead to the designs of other different passive micromixers [5–8], the discrete droplet mixer [9] and the injection mixer [10]. Mixing can be improved either through increasing the number of contact areas or reducing the mixing path. Furthermore, passive mixers such as the packed-bed mixer [11, 12], the serpentine-shape mixer [13], the grooved pattern mixer [14, 15] and the stereo mixer [16, 17] utilise chaotic advection to improve mixing.

In active mixers, external energy is used to affect the flow field and to improve mixing. The external energy can be mechanical [18], acoustic/ultrasonic [19, 20], electrokinetic [21], magnetic [22, 23] or thermal [24]. In general, active mixers have a higher mixing efficiency. However, the requirement of extra energy makes them difficult to be integrated into a microfluidic chip. Furthermore, the relatively high power consumption and cost make active mixers less attractive for disposable applications. Often, passive mixers are preferred.

As mentioned above, many experimental works on micromixers have been done, but little has been reported about mixing theory in micromixers. Ismagilov et al. [25] modelled mixing in microchannels without considering diffusion effect along the flow direction. No analytical solution was given in this work. Later, a similar approach was presented by Kamholz et al. [26]. By introducing an effective dispersion coefficient [27], Beard [28], a later comment [29] and the response [30] simplified the 3D model to a 2D analytical model with the assumption that the channel's width is much larger than its height. However, the analytical form of this

solution is very complicated. Further analysis based on this solution seems to be rigorous. This model is not really suitable for the parametric optimisation and evaluation of micro-mixers. Recently, Holden et al. [31] presented the analytical solution for the 2D mixing model in microchannels without considering the diffusion effect along the flow direction. Our recent work considers the non-linear behaviour of the diffusion process [33]. All of the above works assumed a 1:1 mixing ratio. In practice, other mixing ratios are often needed in microfluidics to achieve a given concentration. To the best of the authors' knowledge, no analytical model exists for diffusive mixing with an arbitrary ratio. Furthermore, no theoretical work was done on diffusive mixing with multiple streams, as reported experimentally in [6] and [8].

In this paper, we present a complete 2D analytical model for diluted convective–diffusive transport in a parallel lamination micromixer with an arbitrary mixing ratio. Based on the simple analytical expression, we found that the dimensionless mixing length is very important for convective–diffusive mixing in micro-channels. Furthermore, the solution can be extended to parallel lamination mixing with multiple streams. The theoretical results are validated by experiments. Polymeric micromixers were fabricated using laser micromachining and adhesive bonding. The mixers were characterised using an optical measurement setup.

2 Analytical modeling

The ratio between mass transport due to convection and diffusion is represented by the Peclet number:

$$\text{Pe} = \frac{UW}{D} \quad (1)$$

where U is the average velocity of the stream, W is the channel width and D is the diffusion coefficient. From the definition in Eq. 1, the two influencing factors for convective–diffusive mixing are the velocity distribution across the mixing channel and the diffusion coefficient D . Non-linearities in the concentration distribution may be caused by these two factors. The following analytical models are established for investigating this problem.

2.1 Velocity distribution inside the mixing channel

A simple model for the velocity distribution is based on a parallel mixer with two inlets. The purpose of this model is to investigate the impact of the different viscosities of the fluids. In order to simplify the problem in the microchannel, the two streams are considered as immiscible. This case is acceptable for miscible streams at high Peclet numbers. Previously, Galambos et al. [34] and Stile et al. [35] established a similar model, but no extensive solutions were given. For the model of the channel cross-section depicted in Fig. 1, the fully developed flow in the microchannel is governed by the Navier–Stokes equations:

$$\begin{aligned}\frac{\partial^2 u_1}{\partial y^2} + \frac{\partial^2 u_1}{\partial z^2} &= \frac{1}{\eta_1} \frac{dp}{dx} \\ \frac{\partial^2 u_2}{\partial y^2} + \frac{\partial^2 u_2}{\partial z^2} &= \frac{1}{\eta_2} \frac{dp}{dx}\end{aligned}\quad (2)$$

where η_1 and η_2 are the viscosities of the two streams and dp/dx is the pressure gradient along the x axis. By non-dimensionlising the coordinate system by the channel using W , the dimensionless Navier–Stokes equations for region 1 and 2 in Fig. 1 are:

$$\begin{aligned}\frac{\partial^2 u_1^*}{\partial y^{*2}} + \frac{\partial^2 u_1^*}{\partial z^{*2}} &= P' \\ \frac{\partial^2 u_2^*}{\partial y^{*2}} + \frac{\partial^2 u_2^*}{\partial z^{*2}} &= \frac{P'}{\beta}\end{aligned}\quad (3)$$

where $P' = W^2 / (\eta_1 u_0) dp/dx$, representing the constant pressure gradient along the channel with reference velocity u_0 and ratio of viscosities $\beta = \eta_2 / \eta_1$. The non-slip conditions at the wall results in:

$$\begin{aligned}u_2^*(1, z^*) &= 0 \\ u_1^*(0, z^*) &= 0\end{aligned}\quad (4)$$

At the interface position r between the two streams, the velocity and the shear stress are

continuous:

$$u_2^*(r, z^*) = u_1^*(r, z^*)$$

$$\left. \frac{\partial u_1^*}{\partial y^*} \right|_{y^*=r} = \beta \left. \frac{\partial u_2^*}{\partial y^*} \right|_{y^*=r} \quad (5)$$

For a flat channel ($h^* \ll 1$) and a constant fluid density, the interface position of the two streams can be estimated based on the mass conservation as:

$$r = \frac{1}{1 + \beta\gamma} \quad (6)$$

where $\gamma = Q_2/Q_1$ is the ratio of flow rates. Galambos et al. [34] only discussed the special case of $\gamma = 1$. The solutions of Eq. 3 have the Fourier forms ($0 < y^* < 1$, $0 < z^* < h/2$):

$$u_1^*(y^*, z^*)$$

$$= P' \left[\frac{z^{*2} - h^2/4}{2} + \sum_{n=1}^{\infty} \cos \theta z^* (A_1 \cosh \theta y^* + B_1 \sinh \theta y^*) \right]$$

$$u_2^*(y^*, z^*)$$

$$= \frac{P'}{\beta} \left[\frac{z^{*2} - h^2/4}{2} + \sum_{n=1}^{\infty} \cos \theta z^* (A_2 \cosh \theta y^* + B_2 \sinh \theta y^*) \right] \quad (7)$$

The coefficients A_1 , A_2 , B_1 and B_2 can be obtained by solving the Fourier series with the above boundary conditions:

$$A_1 = (-1)^{n+1} \frac{4h^2}{(2n-1)^3 \pi^3}$$

$$B_1 = A_1 \frac{(1-\beta)(1-\cosh \theta) \sinh^2 r\theta + (1-\beta) \cosh r\theta \cosh \theta + \beta \sinh^2 \theta}{(1-\beta) \sinh r\theta \cosh r\theta \cosh \theta - \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta)}$$

$$- A_1 \frac{(1-\cosh \theta) \sinh r\theta \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta) + \cosh^2 r\theta}{(1-\beta) \sinh r\theta \cosh r\theta \cosh \theta - \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta)}$$

$$\begin{aligned}
A_2 = & A_1 \frac{(1-\beta) \sinh r\theta \cosh r\theta \cosh \theta - \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta)}{(1-\beta) \sinh r\theta \cosh r\theta \cosh \theta - \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta)} \\
& + A_1 \frac{-\beta \cosh \theta + (1-\beta) \cosh r\theta \cosh \theta - \beta \sinh^2 r\theta + \cosh^2 r\theta}{(1-\beta) \sinh r\theta \cosh r\theta \cosh \theta - \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta)} \\
B_2 = & A_1 \frac{\beta \cosh \theta + (\beta-1) \cosh r\theta \cosh \theta + \beta \sinh^2 r\theta - \cosh^2 r\theta}{(1-\beta) \sinh r\theta \cosh r\theta \cosh \theta - \sinh \theta (\cosh^2 r\theta - \beta \sinh^2 r\theta)}
\end{aligned} \tag{8}$$

Figure 2 shows the typical velocity distribution in a rectangular channel for streams with different flow rates. For streams with the same viscosity, the velocity distribution is flat, as shown in Fig. 2a. With the low aspect ratio of the microchannel discussed in this paper ($h^* \ll 1$), a uniform velocity U can be assumed across the channel width.

2.2 Concentration distribution inside the mixing channel

2.2.1 Mixing with two streams

As mentioned in the previous section, for a flat micro-channel and mixing streams of the same viscosity, a uniform velocity U across the microchannel can be assumed. Furthermore, no diffusion flux exists in the direction normal to the plane determined by the flow direction and the channel width. This situation appears in a Hele–Shaw-flow or in an electrokinetically driven flow. Thus, the 3D model can be simplified to a 2D model as depicted in Fig. 3. The mixer model consists of a long channel of width W , two inlets and one outlet. One inlet stream is the solute with a concentration of $c = C_0$, the other inlet stream is the solvent with a concentration of $c = 0$. The mass conservation equation including both diffusion and convection can be formulated as [32]:

$$U \frac{\partial c}{\partial x} = D \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right) \tag{9}$$

where D is the diffusion coefficient of the solute. With the same viscosity ($\beta=1$) and fluid density, the dimensionless interface location r (Eq. 6) is equal to the mass fraction of the solvent in the final mixture $\alpha = Q_1 / (Q_1 + Q_2) = 1/(1 + \gamma)$ ($0 \leq \alpha \leq 1$). Thus, the mixing ratio of the solute and the solvent is $1: \gamma$ or $\alpha:(1 - \alpha)$. By introducing the dimensionless variables for the

coordinates system $x^*=x/W$, $y^*=y/W$, the dimensionless concentration $c^*=c/C_0$ and the Peclet number (from Eq. 1), Eq. 9 has the dimensionless form:

$$\text{Pe} \frac{\partial c^*}{\partial x^*} = \frac{\partial^2 c^*}{\partial x^{*2}} + \frac{\partial^2 c^*}{\partial y^{*2}} \quad (10)$$

The corresponding boundary conditions for the inlets are:

$$f(y^*) = \begin{cases} c^*|_{(x^*=0, 0 \leq y^* < r)} = 1 \\ c^*|_{(x^*=0, y^*=r)} = 0 \\ c^*|_{(x^*=0, r < y^* \leq 1)} = 0 \end{cases} \quad (11)$$

Full mixing is assumed for the outlet, which means:

$$\left. \frac{\partial c^*}{\partial x^*} \right|_{(x^*=\infty, 0 \leq y^* \leq 1)} = 0 \quad (12)$$

The channel wall is impermeable. Thus, the flux at the wall should be zero:

$$\left. \frac{\partial c^*}{\partial y^*} \right|_{y^*=0, 1} = 0 \quad (13)$$

Equation 13 is also the symmetry condition for mixing with multiple streams, which is discussed later in this section. Equation 10 is a second-order linear partial differential equation. Separating the variables in Eq. 10 and applying the corresponding boundary conditions in Eqs. 11, 12 and 13, the concentration distribution in the channel is:

$$c^*(x^*, y^*) = \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{\sin n\alpha\pi}{n} \cos(n\pi y^*) \exp\left(-\frac{2n^2\pi^2}{\text{Pe} + \sqrt{\text{Pe}^2 + 4n^2\pi^2}} x^*\right) + \alpha \quad (14)$$

$n = 1, 2, 3, \dots$

Obviously, when $\alpha = 1/2$, resulting in the trivial solution $[0, W]$, the model is the common symmetric (1:1) mixing, as reported previously [33].

2.2.2 Mixing with multiple streams

The above solution can be extended to the case of multiple mixing streams. The trivial solution in Eq. 14 can be extended periodically along the channel width direction. With the distance between two neighbouring concentration extrema $W_{\min, \max}$, the same solution can be extended for micromixers with multiple streams. In this case, the mixing ratio can also be arbitrary. For example, if a ratio of a is given and we let the trivial solution be $[-W_{\min, \max}, W_{\min, \max}]$, an analytical solution for a cross-mixer with three inlets can be obtained using Eq. 14. In this case, the Peclet number is evaluated as $Pe = U W_{\min, \max} / D$.

Equation 14 consists of three parts: the modulating coefficients, the cosine function and the exponential function. Corresponding to their effects in the expression, the exponential function determines the whole trend of the concentration distribution along flow direction and the cosine function determines the variation in the channel width. If rapid mixing is needed, the absolute value of the exponent should be large. This means that a small Peclet number causes better mixing. Therefore, the Peclet number (Pe) is the most important parameter for the mixing process in parallel lamination micromixers. Figure 4 shows the results of the above analytical model for some typical cases.

3 Experiments

3.1 Materials and methods

Fully polymeric micromixers made of polymethylmethacrylate (PMMA) and acrylic double-layer adhesive tape were used for verifying the theory. The fabrication is based on the laser machining and adhesive lamination technique. The size of the device is 25×75 mm. Basically, the micromixers consist of three layers: the top layer (PMMA) for optical access, the intermediate adhesive layer (Adhesives Research, Inc., Arclad 8102 transfer adhesive) for channel structure and the bottom layer (PMMA) for fluidic access and alignment. First, the three

layers were cut using a CO₂ laser with a characteristic wavelength of 10.6 μm. Alignment holes are fabricated in the three layers. On the bottom layer, position marks are ablated for the convenience of later measurements. The three layers are laminated using alignment holes. The adhesive layer thickness of 50 μm defines the channel height. Using this technique, a mixing channel of 900 μm in width and 50 μm in height was fabricated for the experiments described in the next sections. This setup is shown in Fig. 5. The resulting aspect ratio of the channel is $h^* \approx 0.056$.

The same image acquisition system as reported in [33] (Fig. 6) was used to measure the velocity distribution and concentration distribution in the mixer. The system consists of three main components: an illumination system (a Mercury lamp and a laser system), an optical system (inverted microscope and CCD camera) and an image acquisition system together with its corresponding software. The laser was used for micro particle image velocimetry (micro-PIV) to verify the uniform velocity distribution in the channels. The two light sources are selected by an optical switch consisting of a reflection mirror. When the mirror is parallel to the input light axis, the laser is active. When the mirror is turned 45 ° to the inlet axis, the light from the Mercury lamp is active.

Both the micro-PIV and the concentration measurements are based on the fluorescent technique. In the micro-PIV measurement, 3-μm red particles (Duke Scientific Co.) were used to trace the movement of the fluid in the channel. In the concentration measurement, a fluorescent dye was used (C₂₀H₁₀Na₂O₅). To enhance the emission light of the fluorescein, 10× TBE buffer (for 1 l 108 g Tris base, Tris (hydroxymethyl)-aminomethane, 55 g Boric acid, and 40 ml 0.5 M ethylene diamine tetraacetic acid (pH 8.0) autoclave for 20 min) was added to the dye solution. The separated fluorescent bands of the microspheres (540/610 nm) and of the dye (490/520 nm) allow the measurements of both the velocity field and the concentration field using the same prepared liquids.

3.2 Results and discussion

3.2.1 Velocity distribution

The PIV measurement uses an epi-fluorescent attachment of type Nikon G-2E/C (excitation

filter for 540 nm, dichroic mirror for 565 nm and an emission filter for 605 nm). Both filters in the attachment have a bandwidth of 25 nm.

The measurement reported in this paper was carried out with a 4× objective. With a CCD sensor size of 6.3×4.8 mm and a resolution of 640×480 pixels, the size of an image pixel is $2.475 \mu\text{m}$ and the size of the measured area is $1,584 \times 1,188 \mu\text{m}$. Two 30-mJ laser pulses with a delay time of 3.5 ms were used as the illumination sources. The integration area is 32×32 pixels.

The velocity measurements were carried out for two cases; water/water and water/diluted glycerol. Diluted glycerol with a volume ratio of 1:4 (glycerol to DI-water at 25 °C) was used as the fluid with the high viscosity. Measurement of the two liquids with a viscometer (Brookfield LVDV-I) results in a ratio of $\beta = 1.7 \pm 0.2$. Only two inlets of the device were in use. Syringe pumps with identical syringes were used so that the same volumetric flow rate can be assumed for each inlet stream. Figure 7 shows the typical results of the micro-PIV measurement. The influence of the viscosity on the velocity distribution can be seen clearly. The measurement results also validates the assumption of a uniform velocity profile in the case of mixing water with dyed water.

3.2.2 Concentration distribution

In our experiments, a single mixer (Fig. 5) and the same mixing ratio 2:1 were used for both mixing cases. Three identical syringes (Hamintol, 500 μl , gas tight) were used in the experiments: one was filled with deionised (DI-)water and the other two were filled with fluorescent dye diluted in water. For the case of two streams mixing, the syringe with DI-water was connected to inlet 1 and the other two syringes were connected to inlet 2 and inlet 3. For three streams mixing, the syringe with DI-water was connected to inlet 2 and the other two syringes were connected to inlet 1 and inlet 3. All three syringes were driven by a single syringe pump (Cole-Parmer 74900-05, 0.2 $\mu\text{l/h}$ –500 ml/h, accuracy of 0.5%).

With the same mean velocity in the mixing channel, the Peclet number of the latter case was half of the former, due to the smaller mixing length of $W/2$. After successfully finding good agreement between our theory and the experimental results using the two streams case, the same parameters with half of the original Peclet number were applied to fit the results of the

three streams mixing.

After recording the images on the computer, the concentration profiles were evaluated using a customised program written in Matlab. First, the program removes the noise in the measured image with an adaptive noise-removal filter. For each pixel, a local mean value is calculated for a window of 5×5 pixels. The noise is assumed to have a Gaussian distribution. Subsequently, a path with the known position across the channel is evaluated. The position across the channel was normalised against the channel width $y^* = y/W$, while the measured pixel intensity I was normalised against the maximum $I_{\max}$ and minimum $I_{\min}$ of the intensity at the inlet:

$$I^* = \frac{I - I_{\min}}{I_{\max} - I_{\min}} \quad (15)$$

The measured dimensionless intensity is assumed to be equal to the dimensionless concentration of the fluorescent dye ($I^* = c^*$). Figure 8 shows the typical 3D intensity distribution in the mixing channel.

During the evaluation of the results, we observed non-linearity in the concentration distribution. Since the velocity is uniform, the non-linearity can only be caused by the diffusion coefficient D . This effect has already been reported in [36, 37, 38] and analysed in our previous work [33]. The concentration-dependent diffusion coefficient can assume the model:

$$D(c) = D_0 \left[(1 - a) \frac{c}{c_0} + a \right] \quad (16)$$

where D_0 is the coefficient at the maximum concentration c_0 and a is the factor describing the interaction between the solute and solvent. Since Eq. 16 makes the governing equation in Eq. 10 non-linear, and no pure analytical solution exists for this case, thus, an approximation with the linear solution (Eq. 14) was used to fit the measurement results. The fitting parameters are D_0 and a [33].

Because of both the unknown diffusion coefficient D_0 and the factor a , the non-linear mixing theory was used for fitting the measurement results. While the solute side was used for finding D_0 , the solvent side was used for determining the factor a . Using several measurements at different average velocities, the fitting parameters $a = 0.5$ and $D_0 = 1.8 \times 10^{-9} \text{ m}^2/\text{s}$ could be found for the experiments presented in this paper. Due to the added 10× TBE buffer, these parameters are slightly different to our previous results in [33].

Figures 9 and 10 depict the results of two streams mixing, while Figs. 11 and 12 show the results of three streams mixing. All these figures compare the measured concentration profiles and concentration gradient profiles with the theoretical results using the fitting parameters mentioned above. It can be seen clearly that the non-linear approximation describes relatively well the diffusive mixing process in the microchannel. The broadening band can be observed with the gradient profile. The band is thinner at a higher Peclet number.

During the course of the evaluation, another important parameter was found for the Y-mixer. Ignoring the diffusion in the flow direction, the analytical solution becomes:

$$c^*(x^*, y^*) = \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{\sin n\alpha\pi}{n} \cos(n\pi y^*) \exp\left(-\frac{n^2\pi^2}{\text{Pe}} x^*\right) + \alpha$$

$$n = 1, 2, 3, \dots \quad (17)$$

In [31], from the expression of the exponent (similar to the exponent in Eq. 17), Holden et al. introduced a dimensionless parameter $\kappa = x^*/\text{Pe}$. However, they did not give any further explanation on it. In our opinion, this parameter can be regarded as a dimensionless mixing length in microchannels. If the channel length is L , the residence time is $t = L/U$. According to Einstein [39], t is proportional to W^2/D . Thus, L is proportional to UW^2/D . Dividing x by UW^2/D results in the proportionality factor $DL/UW^2 = x^*/\text{Pe}$. In system theory, the response function $\exp(-t/\tau)$ is considered complete when $t \approx 3\tau$. Similar to this approach, κ can be used for judging whether or not good mixing is obtained.

3.3 Conclusions

Two-dimensional analytical models for the velocity distribution and diluted convective–diffusive mixing in parallel lamination micromixers with an arbitrary mixing ratio were presented in this paper. All the previously reported models are special cases of this new analytical model. Furthermore, the solution can be applied for parallel lamination micromixers with multiple streams because of the similarity of the boundary conditions at the wall and the symmetry condition in the middle of a single stream. We demonstrated that the Peclet number and the dimensionless mixing length κ are the most important parameters for a passive T-mixer. All these conclusions were verified by the experimental results. The model presented in the paper can be used as an analytical tool for the parametric optimisation of passive micromixers.

Acknowledgements This work was supported by the academic research fund of the Ministry of Education Singapore, contract number RG11/02. The first author wishes to gratefully acknowledge the PhD scholarship from Nanyang Technological University.

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Fig.1

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