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Evaluation of Micro Mixers using Competing Reactions

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Abstract

In this study, two micro mixers were fabricated and tested using a system of parallel competing reactions involving the Dushman reaction between iodide and iodate, coupled with neutralization. Using competing reactions allowed the influence of the operating conditions and microstructure on mixing performance to be investigated. The results of this study revealed that the degree of mixing was improved as the flow rate through the mixer increased, which is consistent with existing experimental data obtained using conventional optical techniques. This study also demonstrated that the mixing performance could be enhanced by changing the inlet shape of the mixing chamber. Numerical simulations conducted to determine why the change in inlet shape enhanced the mixing performance indicated that mixing performance is dependent on the degree of impingement and the circulation flow, which are dominated by the flow rate and the inlet shape of the mixing chamber, respectively.

Keywords: Mixing performance evaluation, Micro mixer, Competing reactions, Numerical simulation

Introduction

Mixing of the fluid flowing through the microchannels of microfluidic systems is important, therefore, many mixing methods have been proposed. Despite the large amount of research that has recently been conducted on micro mixers, their evaluation still remains a challenge, and it is particularly important to quantify the extent of mixing in order to evaluate the performance and enable the optimization of micro mixers.

Optical techniques that visualize the boundary layer between fluid layers have traditionally been used to determine the extent of mixing[1]. However, these techniques depend on the orientation of mixing relative to the imaging direction, therefore, if the imaging direction is perpendicular to the interface between fluid layers it is difficult to determine the degree of mixing using captured images. Competing reactions, which act as a molecular probe, have been used to assess micromixing efficiency since the early 1980s[2]. The aforementioned imaging problem is not an issue when the chemical method of assessment is used, and this method has been used to evaluate the mixing performance of industrial reactors[3-5]. However, to the authors' knowledge, no studies have been conducted to determine if competing reactions can be used to evaluate the efficiency of micro mixers. Therefore, in this study a system of parallel competing reactions involving the Dushman reaction between iodide and iodate, coupled with neutralization, were used to evaluate two micro mixers fabricated for this study. The use of competing reactions also allowed the influence of operating conditions and microstructure on mixing performance to be evaluated. In addition, numerical simulations of the tests were conducted to clarify the reasons for which the experimental results were obtained.

Results and Discussion

Figure 1 shows the structure of the two micro mixers used in this study. As seen in Figure 1(a), design 1 is similar to that of the conventional micro T-mixers. For Design 2, shown in Figure 1(b), the inlet of the mixing chamber was designed to laminate the fluid layers in a different way than in design 1. The micro mixers were fabricated using standard microelectromechanical systems (MEMS) technology, as shown in Figure 2. Both micro mixers were tested using the experimental setup shown in Figure 3.

The degree of mixing is proportional to the absorbance at 353 nm, therefore, it can be quantified by measuring the absorbance of the product at this wavelenght. To quantify the degree of mixing using absorbance, a mixing index I_M was defined as follows:

$$I_M=\frac{Abs._{\text{least mixed}}-Abs.}{Abs._{\text{least mixed}}-Abs._{\text{most mixed}}} \qquad (1)$$

where Abs. represents the absorbance of the product obtained from the micro mixer at 353 nm. $Abs._{\text{least mixed}}$ and $Abs._{\text{most mixed}}$ are the absorbance values obtained when the least mixing occurred and

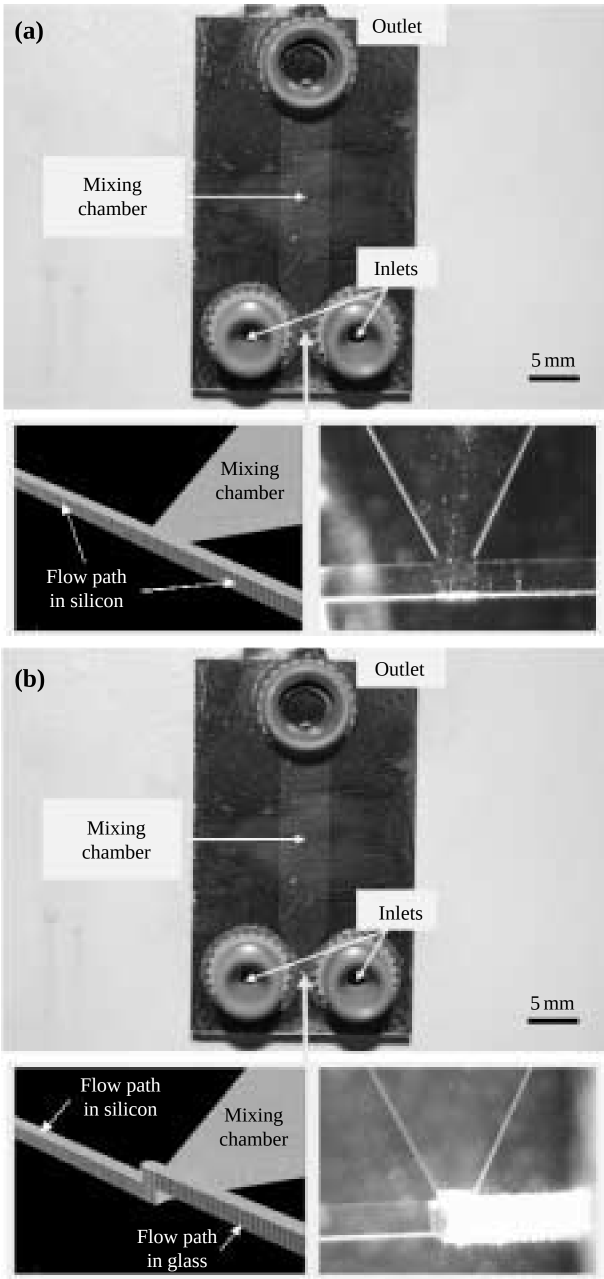

Figure 1. Tested micro mixers: (a) Design 1; (b) Design 2.

when the most mixing occurred, respectively. Since an I_m of 0 indicates that no mixing occurred and an I_m of 1 indicates complete mixing, an I_m closer to 1 can be obtained by improving the degree of mixing.

The absorbance measurements are summarized in Figure 4. The absorbance was found to decrease as the flow rate increased, and the absorbance obtained using Design 1 was found to be greater than that obtained when Design 2 was used at the same flow

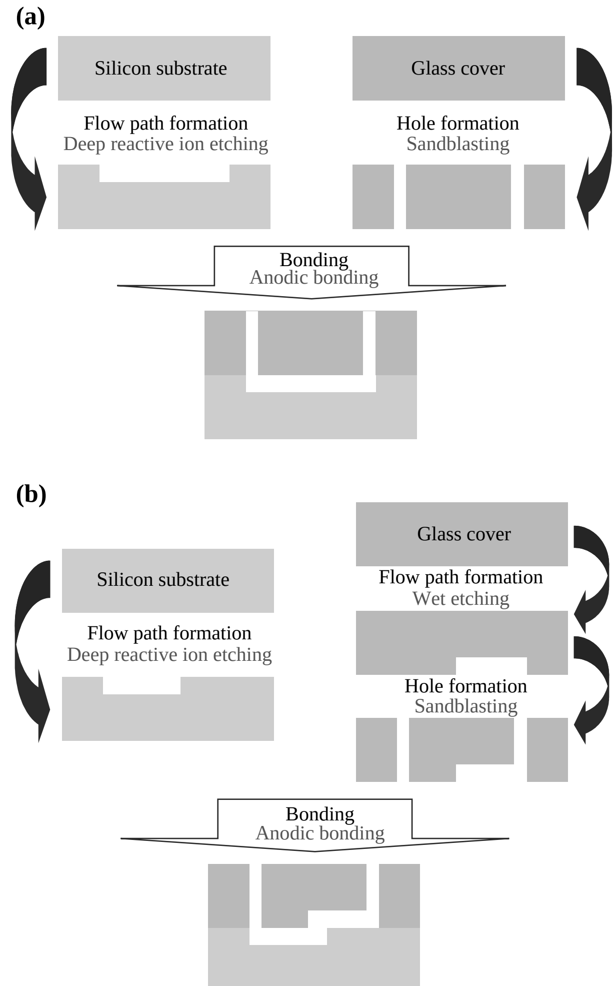

Figure 2. Fabrication processes: (a) Design 1; (b) Design 2.

rate. When the absorbance values were converted to I_M values, the degree of mixing was found to improve as the flow rate through the mixer is increased as shown in Figure 5. This is consistent with the findings of studies conducted on micro mixers using conventional optical techniques[6]. Figure 5 also shows that the degree of mixing obtained using Design 2 was greater than that obtained using Design 1 when the same flow rate was used.

To determine why the degree of mixing increased as the flow rate increased, the tested cases were numerically simulated using the commercial simulation tool CoventorWare. Figure 6 shows the constructed 3D numerical models. In these models, Fluid 1 flows into the mixer through Inlet 1 and Fluid 2 flows into the mixer through Inlet 2. Next, fluid 1 and 2 are mixed in the mixer, and the mixture of Fluid 1 and 2 then passes through Outlet. The standard deviation of

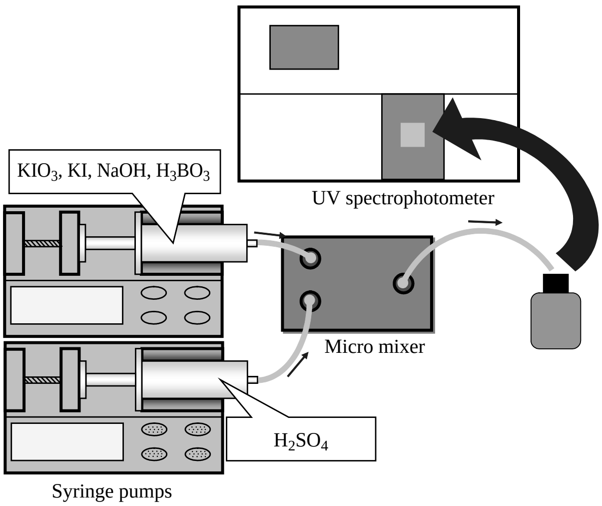

Figure 3. Experimental setup.

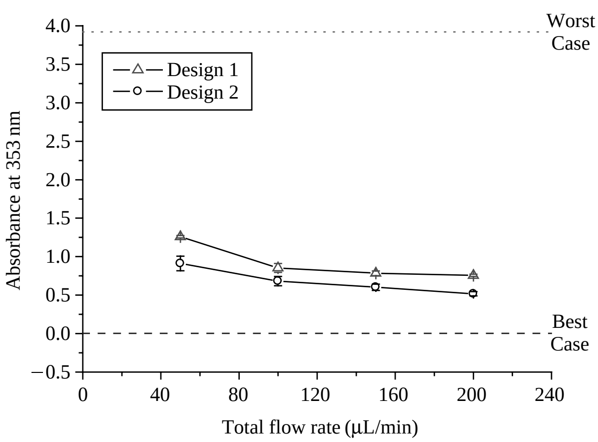

Figure 4. Absorbance measurement results.

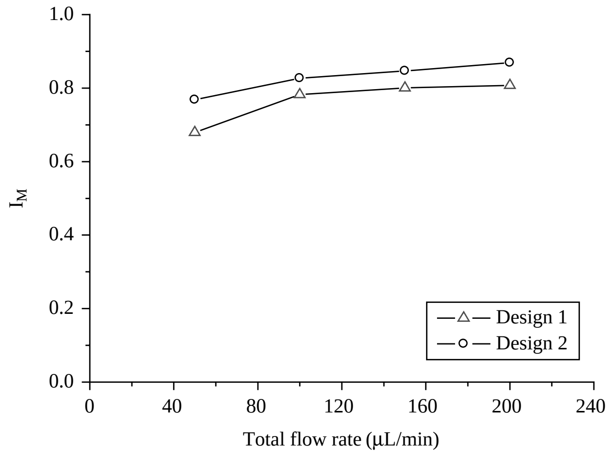

Figure 5. Test results on mixing index of micro mixers.

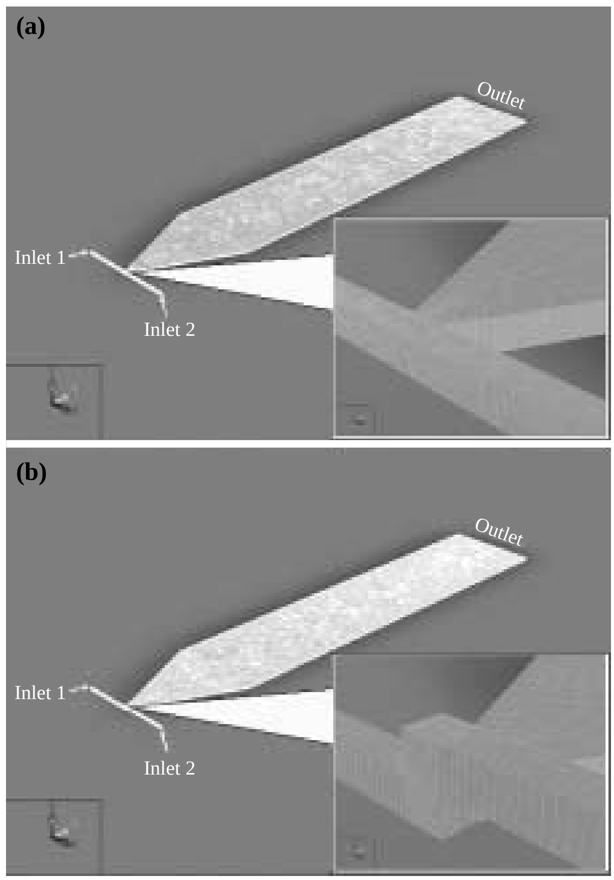

Figure 6. Numerical models: (a) Design 1; (b) Design 2.

Fluid 1 concentration at the Outlet was calculated and is summarized as shown in Figure 7. Since the standard deviation decreases as the mixture becomes more uniform, the standard deviation can be used to evaluate the degree of unmixedness. As shown in Figure 7, the degree of mixing obtained using Design 2 was greater than that obtained using Design 1, which indicates that the mixing performance of Design 2 was better than that of Design 1, which is in agreement with the experimental findings shown in Figures 4 and 5. Additionally, as was shown by the experimental results, the numerical results show that the degree of unmixedness decreased as the flow rate increased. The agreement between the experimental and numerical results validates the numerical simulations used in this study; therefore the validated numerical results were used to conduct a detailed analysis of the effect of the microstructure and the total flow rate on the mixing phenomenon. Figure 8 shows the effect of the total flow rate on the mixing phenomenon in Design 1. The velocity vector profiles shown

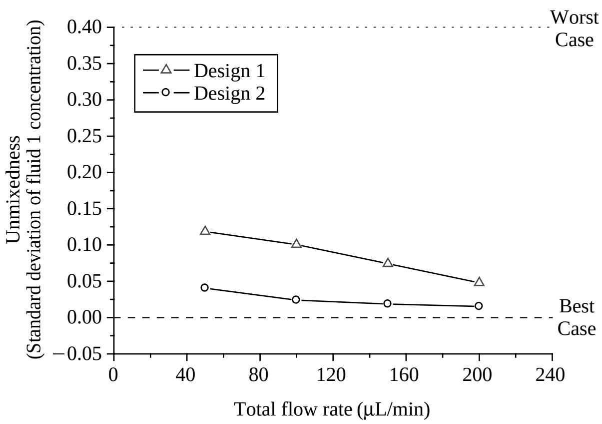

Figure 7. Results of numerical simulation to determine unmixedness.

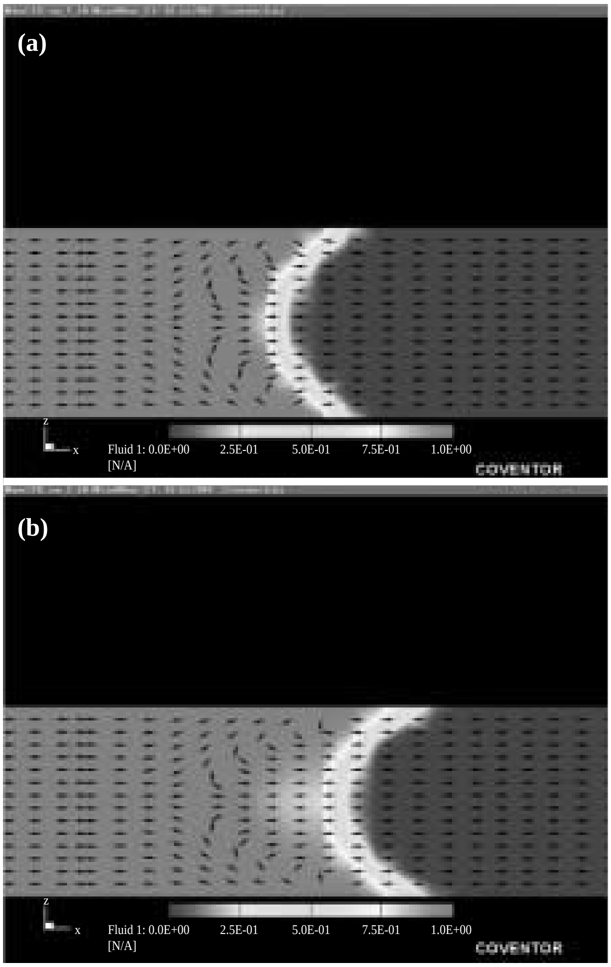

Figure 8. Effect of total flow rate on mixing phenomenon (Design 1): (a) 50 μL/min; (b) 250 μL/min.

in Figure 8 indicate that the mixed zone at the interface between two fluids was initially generated by the impingement of the two fluids. Because the impingement was dominated by the momentum of fluids, the mixed zone became broader as the total flow rate increased, as shown in Figure 8, this phenomenon can be ascribed to the enhanced mixing that occurred as a result of the increased flow rate. As seen in Figure 9, the vector profiles for design 2 indicate that both impingement and circulation flow occurred when Design 2 was used, and the circulation flow stretched the mixed zone, thereby facilitating the mixing phenomenon. Therefore, it can be inferred that the better mixing performance obtained when Design 2 was used occurred as a result of the circulation flow generated by the microstructure. However, the velocity vector profiles shown in Figure 9 show that the effect of the flow rate on the degree of circulation is not important, therefore, it can also be inferred that the mixing performance enhancement that occurred as a result of the increased flow rate when Design 2 was used was primarily due to more active impingement. The effect of the circulation flow on mixing generated when Design 2 was used became greatest when a low flow rate was used, therefore the greater mixing performance by Design 2 becomes evident as the flow rate decreases, as shown in Figures 4 and 7.

Conclusions

In this study, two micro mixers were fabricated using MEMS technology, and then tested successfully using a system of parallel competing reactions involving the Dushman reaction between iodide and iodate, coupled with neutralization. Using the competing reactions allowed the influence of operating conditions and microstructure on mixing performance to be investigated, and revealed that the degree of mixing was improved as the flow rate through the mixer increased. This is consistent with findings regarding micro mixers that have been obtained using conventional optical techniques. This study also demonstrated that mixing performance can be enhanced by changing the inlet shape of the mixing chamber. To clarify the reason these results were obtained, the experimentally tested cases were simulated numerically. The numerical simulation indicated that mixing performance was dependent on the degree of impingement generating the mixed zone and the circulation flow stretching the mixed zone. Because impingement and circulation are dominated by flow rate and the inlet shape of the mixing chamber, respectively, it should be possible to determine the geometry and operating conditions that would provide the optimum

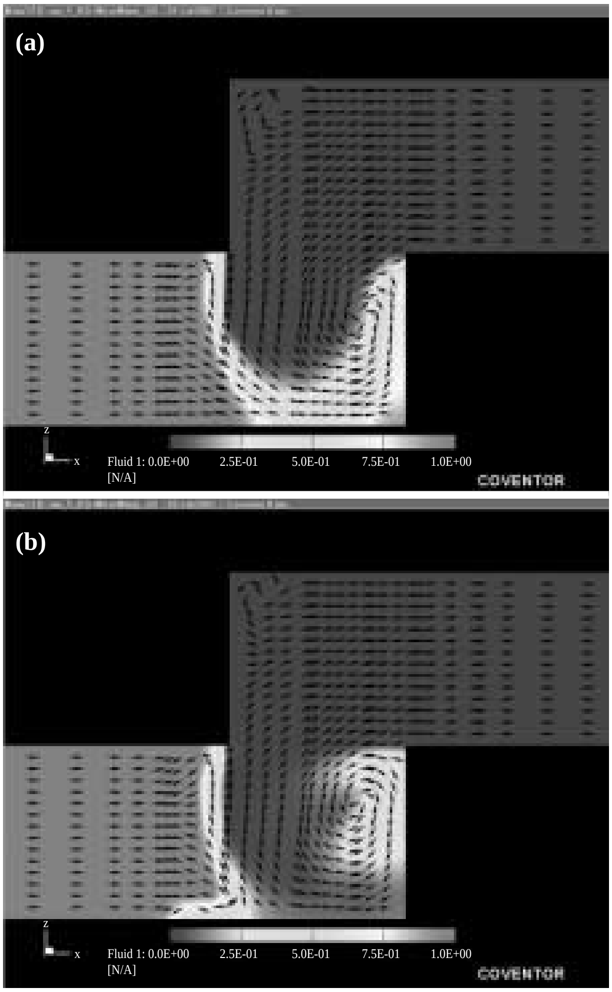

Figure 9. Effect of total flow rate on mixing phenomenon (Design 2): (a) 50 μL/min; (b) 250 μL/min.

mixing performance.

Materials and Methods

When a basic mixture of iodate, iodide, sodium hydroxide and boric acid is mixed with sulfuric acid, an acid-base neutralization and an oxidation reaction, called the Dushman reaction, occurs according to the following steps[2].

$$H_2BO_3^- + H^+ \rightarrow H_3BO_3 \quad (2)$$

$$\left(\frac{5}{3}I^- + \frac{1}{3}IO_3^-\right) + 2H^+ \rightarrow I_2 + H_2O \quad (3)$$

$$I_2 + I^- \Leftrightarrow I_3^- \quad (4)$$

The first reaction, which is the neutralization reaction, occurs instantaneously with respect to the second reaction. In the last reaction, iodine produced by the second reaction reacts with iodide in excess to form I_3^-. If the micromixing is complete, the acid is totally and instantaneously neutralized by the first reaction and no I_3^- appears. However, if micromixing is incomplete, a local excess of acid induces iodine formation, yielding I_3^-. Therefore, the concentration of I_3^- in the final product indicates the degree of unmixedness, which can be determined by measuring the absorbance of the final product.

By using the competing reactions described above, the mixing performance of two micro mixers was evaluated. The micro mixers were fabricated using standard MEMS technology, as shown in Figure 2. Briefly, for both Design 1 and 2, a 100 μm deep flow path, including a mixing chamber (width: 5 mm, length: 20 mm), was formed on a silicon substrate by deep reactive ion etching. For Design 2, a 100 μm deep microchannel was formed on the cover glass by wet etching for Design 2, as shown in Figure 2(b). Holes for the flow connection were then fabricated by sandblasting for both Design 1 and 2. Finally, the silicon substrate and the glass cover were aligned and bonded together.

The fabricated micro mixers were then tested using the experimental setup shown in Figure 3. A basic mixture of iodate (2.33×10^{-3} mol/L), iodide (1.16×10^{-2} mol/L), sodium hydroxide (0.0909 mol/L), boric acid (0.0909 mol/L) and sulfuric acid (0.02 mol/L) was infused into the micro mixer by syringe pumps (Harvard PHD22/2000) at a flow rate ranging from 40 μL/min to 160 μL/min, which is four times as large as the acid. Next, the product obtained using the micro mixer was collected for 40 minutes, and the absorbance at 353 nm was then measured using a UV spectrophotometer (Varian Cary300Conc). To determine the mixing index (I_M) using the absorbance, the $Abs._{least\ mixed}$ and $Abs._{most\ mixed}$ must be measured in advance. To determine the least mixed value, sulfuric acid was mixed with iodate and iodide first, followed by infusion of sodium hydroxide and boric acid into the mixture. The absorbance of the mixture at 353 nm was then measured. Conversely, the most mixed value was determined by completely neutralizing the sulfuric acid with the mixture of sodium hydroxide and boric acid, after which iodate and iodide were infused into the mixture, and the absorbance at 353 nm was measured.

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