



Combined improved mixing and reduced energy dissipation by combining convective effects and lamination

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HIGHLIGHTS

- ▶ Power dissipation combined with mixing quality allows for evaluation of a mixer.
- ▶ Mixer effectiveness encompasses good mixing at a lower power cost.
- ▶ Combining these mixing principles allows for improved mixing at a lower cost.
- ▶ Older designs were studied; new designs with better mixer effectiveness were made.

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ABSTRACT

The current paper describes an in situ mixing study of mixers that rely on both convective effects and lamination. The mixers were milled in polymethylmethacrylate and their performance was measured by fluorescence dilution experiments, monitoring the pressure drop as well. Thanks to the ability to examine the local mixing efficiency within each mixing element, improvements could be suggested, which were also supported by experimental data. The study aimed at improving the performance of planar passive micromixers with channel dimensions between 200 and 500 μm capable of operating under laminar conditions for flow rates ranging between a few 100 μl to several ml per minute. Two new mixer designs were developed and compared with three well known designs: the T-mixer, the micromixer with static mixing elements and the square-wave mixer. The first new design was the modified square-wave design with by-passes. In this design the square-wave mixer is modified by introducing by-passes leading to serial lamination. A simplified model that predicts flow splitting at the by-pass is proposed. The second design is the three dimensional variant of the modified square-wave mixer with by-passes. This 3D mixer allows for enhanced mixing when recombining the flows. The mixing performance is analyzed over a wide range of flow rates (ml/min range) and constitutes of both the degree of mixing as well as the power requirement. A number of design improvements resulted in improved mixing with a simultaneous reduction in energy dissipation, which contrasts with (off-chip) observations in the literature. Mixing is an energy-requiring process, but depending on the applied mixing mechanism the energy dissipation required for mixing can be reduced, resulting in higher mixer effectiveness.

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1. Introduction

Mixing processes are critical in a variety of applications, e.g. for fast chemical reactions and biochemical analysis, because it controls the extent of the reaction. In most microfluidic devices, the Reynolds number is low due to a viscosity dominated flow, as can be understood from $Re = \rho D/\mu$ (with ρ = fluid velocity, D = hydraulic diameter, μ = kinematic viscosity). Under laminar flow conditions the mixing of two flows in a microfluidic device

is purely diffusion based. The required diffusion mixing time (t) across a distance of x can be estimated by the formula $t \sim x^2/D_{mol}$, where x is the characteristic dimension and D_{mol} is the molecular diffusion coefficient [1,2]. Mixers with dimensions in the order of hundreds of microns have diffusion based mixing times of tens of seconds (D_{mol} is (for liquids) generally in the order of $10^{-9} \text{ m}^2/\text{s}$ or smaller), which is slow and ineffective as compared to convective motion [3]. To overcome this purely diffusion based mixing limit, numerous innovative designs have been proposed during the last decade [4–7]. Recently a number of interesting studies have appeared on mixers [8–13]. All these designs aim at enlarging the contact surface and reducing the diffusion distance, as this is the obvious way to enhance mixing.

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Passive type micromixers working in laminar conditions can be mainly categorized as lamination and chaotic-advection-types. Lamination micromixers (split-and-recombine mixers) redistribute the fluids in sub-channels to reduce the diffusion distance between the flows that are being contacted. Chaotic advection type micromixers introduce planar designs or 3D structures to generate transverse flows which can enlarge the contact interface between the liquid streams. Mixers that can be operated effectively in flow rate conditions from a few, to tens of milliliters per minute are of significant interest; as these are the flow rates that typically appear in for example microreactors and in analytical liquid chromatography (HPLC), where a mixer is needed for the mobile phase gradient generation.

A number of mixers that can be operated within this flow rate region have been already studied elaborately in the literature. In the T-mixer, two fluids are brought into contact at a 180° angle at the inlet section and flow alongside each other in a straight channel. The mixing quality depends on the flow rate. At low flow rates, diffusion is the sole mechanism to induce mixing. At higher flow rates, convective effects in the inlet section strongly enhance the mixing quality [14–16]. To improve its performance, so-called static mixing elements (SMEs) mixers have been developed. These are T-mixers with added mixing structures in the main channel. At low Reynolds numbers, the SME lead to an increased contact surface, in turn leading to shorter mixing times. At higher Reynolds numbers, these SME induce the formation of vortices and chaotic advection, which drastically improves mixing when comparing to plain diffusion based mixing [17–19]. In the square-waver mixer, fluids move in a lateral and axial fashion and mixing is expected to be accomplished by the convective motion in the bends of the channel. At high flow velocities, local deviations of the laminar behavior occur in the bends. These deviations can induce a transversal component of velocity, the so-called racetrack effect which improves mixing [20–25]. This racetrack effect may lead to a tendency to move outwards when having a velocity that is too elevated to curve smoothly around a bend.

In this work, two redesigns of the square wave mixer are proposed so that more efficient mixing can be effectuated under a fairly broad flow rate spectrum (roughly from 0.1 to 10 ml/min), yet at a lower power expense. The performance of these adapted mixers is compared using in situ fluorescence microscopy to the T-mixer, the static mixing element mixers and the square-wave mixer.

In situ measurements are valuable, as they provide a straightforward interpretation of the influence of design parameters and lead to suggestions as to where adjustments are needed. More importantly, the pressure drop has been taken into account as well. This allowed for the calculation of the power dissipation required to achieve a certain mixing quality. The method of fabrication – CNC milling of polymethylmethacrylate (PMMA) sheets – allowed for inexpensive experimental studies on more complex 3D designs.

2. Experimental section

2.1. Production of micron sized channels by milling of PMMA sheets

The microchannels were fabricated in a PMMA (Eriks–Baudoin, Hoboken, Belgium) substrate by milling (CNC milling engraving machine, DATRON, New Hampshire, USA). The rotation speed was 50,000 rpm and the milling speed was 200 mm/min for the X- and Y-axis and 50–100 mm/min for the Z-axis (depending on the size of the mills) (Datron Double Flute End Mill D1 0.2 mm, 0.3 mm, 3 mm, Datron, New Hampshire, USA). About 300 µm of material was removed during each repeating step. Afterwards, possible leftover debris was manually removed and subsequently

purged by pressurized air. The mixers consist of two thermally bonded sheets (130 °C, 30 min). The bottom sheet was structured with functional units, whereas the top sheet was a blank PMMA sheet.

2.2. Design of the micromixers

In this section the design of the micromixers is discussed, the motivation for several design aspects is clarified in the results section. An overview of the studied designs is given in Fig. 1.

2.2.1. Planar mixers with static mixing elements (SMEs)

A T-shaped channel was modified by introducing static mixing elements (SMEs). For the experiments, three different shapes for the elements were used: circular pillars (CPs), diamond shaped pillars (DPs) and stretched pillars (SPs). The height of the elements was chosen equal to the height of the channel. The elements were put symmetrically in the middle section of the channel. To avoid preferential flow at the sides of the mixing channel (where no elements are placed), a second set of mixers was made, which included structured walls. The best performing mixer of these two sets of mixers is compared with the new mixer designs in the results section. The internal dimensions of the mixers are given in Table 1 and indicated in Fig. 1A and B.

2.2.2. Square-wave type micromixers

The experiments in the square-wave mixers were conducted in a meandering channel with a Y-shaped inlet section. The width of the main channel was varied between 200 µm to 400 µm. The internal dimensions of the mixers are given in Table 2 and Fig. 1C.

2.2.3. Modified square-wave mixer with by-passes (MSWBPs)

The design of this mixer is based on the square-wave design, but with added by-pass channels (BP, see Fig. 1D) to improve the mixing performance. The width of the main channel (l) was kept at 300 µm and its height at 200 µm. The lateral width of the mixing structure (m) (i.e. the larger and smaller element encompassing the by-pass) was enlarged to 1900 µm to prevent channeling of the flow, because this renders the mixers ineffective. The by-pass channels had a width and depth of 200 µm and the axial and lateral width of the separate blocking element (n) was 400 µm.

2.2.4. Modified square-wave mixer with by-passes: 3D (MSWBPs-3D)

An extra layer was added to the planar design to allow for a flow perpendicular to the plane of the MSWBP. This addition enabled repeated countercurrent contacting over the by-passes (Fig. 1E). In the conducted experiments, the main channel width (o) was 300 µm and its height was 200 µm. The by-pass height was 200 µm and its width 200 µm, the length of the by-pass was 400 µm. The diameter of the hole to the 3D plane (p) was 300 µm. The length of the channel in the third dimension was 3 mm. While this length does not significantly influence the mixing performance (the mixing is essentially determined by the by-passes and bends), this length was imposed by the thickness of the employed PMMA plates (3 mm thick). Removing a fraction of this plate would have resulted in poor optical quality for the fluorescence signal; hence it was preferred to leave the thickness of the 3D-mixer as it was.

2.3. Mixing experiments

To study the performance of the mixers, dilution experiments were performed. A Fluorescein Isothiocyanate (FITC, 10^{-5} M, Sigma Aldrich, Bornem, Belgium) stock solution was prepared by dissolution in ultrapure water (Synergy UV Water Purification System, Millipore, Massachusetts, USA). Two syringe pumps (260D Syringe

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