Laminar Mixing in Different Interdigital Micromixers: I. Experimental Characterization

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Interdigital-type glass micromixers with alternating feed channels to periodically create liquid multilamellae, were fabricated for basic investigations of hydrodynamics for liquid mixing of two streams. Three interdigital designs developed, rectangular, triangular, and slit-type, differed in their flow-through mixing chamber. These designs are based on simple polygon geometries and their combinations. The flow patterns of an aqueous solution dyed with blue and uncolored water were investigated for different interdigital mixers. Since mixing in a nonfocused device, such as the rectangular mixer, was not completed, focusing techniques were applied. Geometric focusing was used to reduce lamellae width and to speed up mixing. In the special version of the triangular mixer, SuperFocus mixer, liquid mixing times are reduced to about 10 ms, as determined by iron-rhodanide reaction imaging. In the latter case, the lamellae are compressed by a factor of 40, from a width of 160 to 4 μm. For both the triangular and slit-type micromixer, flow patterns differed from regular multilamination ones. At high flow rates, lamellae tilted. When the flow cross-sectional area was expanded, jet and associated eddies formed further improved mixing. Mixing investigations also helped develop imaging techniques. The rhodanide imaging complemented water blue contrasting.

Introduction

Micromixers recently attracted attention for various applications in chemistry (Löwe et al., 2000). The well-known kind of mixer among devices are for multilamination of fluid layers (Branebjerg et al., 1996a,b; Ehrfeld et al., 1999; Löwe et al., 2000; Schubert et al., 2001; Zech et al., 2000). Among this class of mixers, so-called interdigital mixers (Ehrfeld et al., 1999) were tested in regard to widespread applications. A number of these investigations demonstrated benefits concerning process intensification (in terms of an increase in selectivity, yield, or space-time yield) (Krummradt et al., 2000), processing time (Löwe et al., 2000), energy consumption (Bayer et al., 2000b), and process maintenance (Bayer et al., 2000a). Moreover, the fast construction and disassembly of plants based on such devices accompanied by the high process safety (Ehrfeld et al., 2000) and low consumption of chemicals (de Bellefon et al., 2000) provides large flexibility when facing new challenges in process engineering.

In this context, interdigital micromixers were used for fast and efficient process development regarding a metallo-organic reaction (Krummradt et al., 2000) and to prevent polymer fouling of acrylic resins (Bayer et al., 2000a). Moreover, these devices allowed to safely gather process information in otherwise explosive regimes (Ehrfeld et al., 2000). Apart from advantages concerning reaction engineering, interdigital micromixers turned out to be efficient tools for extraction in the framework of miniplant technology (Benz et al., 2001) and for emulsification (Haverkamp et al., 1999; Hessel et al., 2001; Schiewe et al., 2000). Finally, recent research is directed towards screening of liquid/liquid reactions using homogeneous catalysts (de Bellefon et al., 2000).

Although more and more information on process performance is gained, a fundamental knowledge on the underlying mixing processes in interdigital mixers was so far not broadly accessible. At first sight, one could raise the objection that it is known from other investigations that the multilamination process provides simple geometric patterns of fluid layers (Branebjerg et al., 1996b; Floyd et al., 2000), termed lamellae

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(Branebjerg et al., 1996a). However, it is not so evident how these patterns may change if mixing chamber geometries which are very different from the existing ones, often rectangular, are employed. For instance, this refers to a multilamellae flow, which is abruptly compressed by a large change in channel cross-section over a short distance.

There are a number of reasons to use mixing chambers with distinctly different geometry from simply rectangular (see below). This also holds for combinations with conventional equipment. A practical argument may refer to fabrication issues, which is more severe when series fabrication is demanded. For instance, due to the price-performance ratio, it sometimes is adviseable to connect the micromixer to conventional equipment such as double-mantled tubes. For some process conditions, mixing then will inevitably take place in the conventional tube. Due to the fluidic interfaces between the micromixer and the tube, the fluid domain may change in the geometry of the channel cross-section (such as from rectangular to circular) and compressing and stretching of lamellae will be unavoidable.

With regard to the mixing process itself, there are also reasons to modify the cross-section of the mixing chamber. A directly cogent argument refers to the speed of mixing. Knowing that typical lamellae widths in today's multilamination mixers are often not smaller than 100 μm, it is likely to assume typical mixing times for liquids to be in the order of some seconds (by applying the diffusion equation as, for example, given in Branebjerg et al. (1996a), which is needed for quench-flow techniques or mixing of reactions that are completed in the second range or even faster. Therefore, focusing techniques by geometric (Branebjerg et al., 1996b; Floyd et al., 2000) and hydrodynamic (Knight et al., 1998) constraints have been identified as a proper means to speed up mixing in multilamination mixers.

Moreover, keeping in mind that defined setting of mixing in its own may be used to control reaction selectivity, ratio of product isomers and regioselectivity for organic reactions (Roessler and Rys, 2001), it is evident that there is a request for control over spatial and temporal evolution of reactant and product concentration. This, in turn, will demand for specialized micromixers differing in the geometry of their mixing chamber.

In this context, this article focuses on a structure-property analysis being dedicated to show the interplay between mixer chamber geometry and flow patterns. Not only is this information given, but, additionally, the methods are provided to obtain it, both on the experimental and theoretical side. Consequently, this article was split into two parts.

The first part of the article will rely on the description of experimental work that is in line with the discussion above. A base kit of three micromixers with different mixing chambers is introduced. Experimental findings will be presented which are not consistent with simple multilamination patterns. By establishing two suitable flow visualization techniques, it was possible for selected cases to get information of a complementary nature. The base kit was supplemented in a later stage by a fourth optimized micromixer, termed *SuperFocus*.

In the Part II of the article, computational fluid dynamics will be used to understand the experimental results, with some being unexpected. The existence of distinct flow patterns, different from simple multilaminated lamellae, is shown. Using the information gained from mathematical modeling, a rational design of the new focusing micromixer *SuperFocus* was developed, with the goal of achieving minimal mixing times.

Micromixer Design

For the purposes mentioned above, micromixers completely made of glass were developed, allowing the visualization of the flow within the entire device (see Figure 1).

All glass micromixers were made by means of an etching technique using a special photostructurable glass termed FOTURAN (Freitag and Dietrich, 2000; Dietrich et al., 1996). Foturan is a commercial speciality glass made by Schott Desag AG (Grünenplan, Germany) and licensed to mgt mikroglas technik Mainz (Mainz, Germany). The composition of FOTURAN is $SiO_2 = 75-85\%$, $Li_2O = 7-11\%$, $K_2O = 3-6\%$, $Al_2O_3 = 3-6\%$, $Na_2O = 1-2\%$, $ZnO = 0-2\%$, $Sb_2O_3 = 0.3\%$, $Ag_2O = 0.1\%$, $CeO_2 = 0.015\%$.

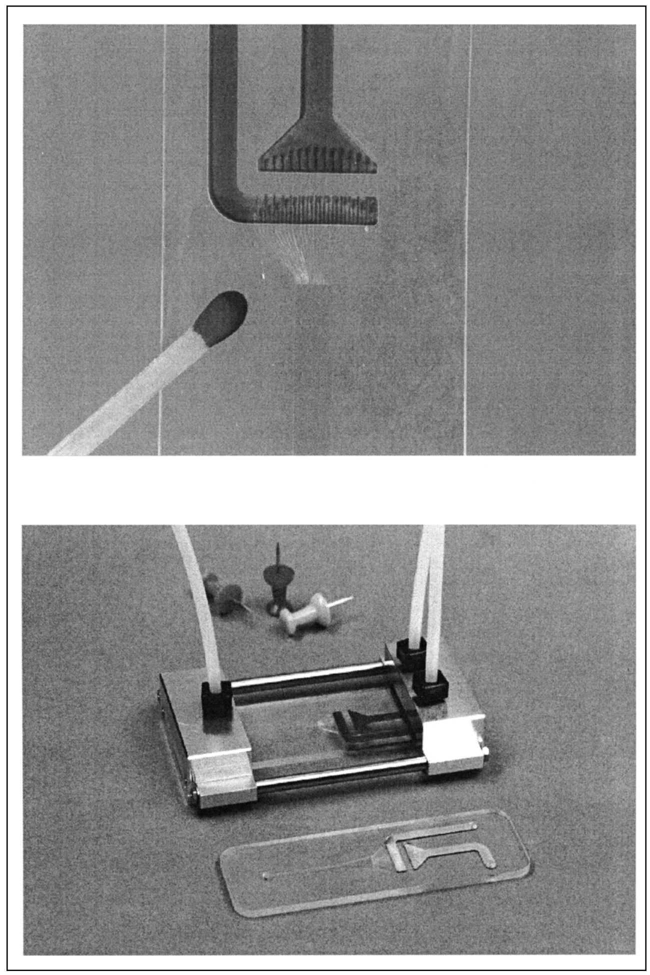

Figure 1. Interdigital glass mixer made by an etching process of a photostructurable glass, Foturan.

Top: bonded glass layer stack. The large black structures are the two main feed channels. The residual feed structures and the mixing chamber are hardly visible for reasons of missing contrast. Bottom: Bonded glass layer stack inserted into two aluminum end caps comprising polymer fluid connectors.

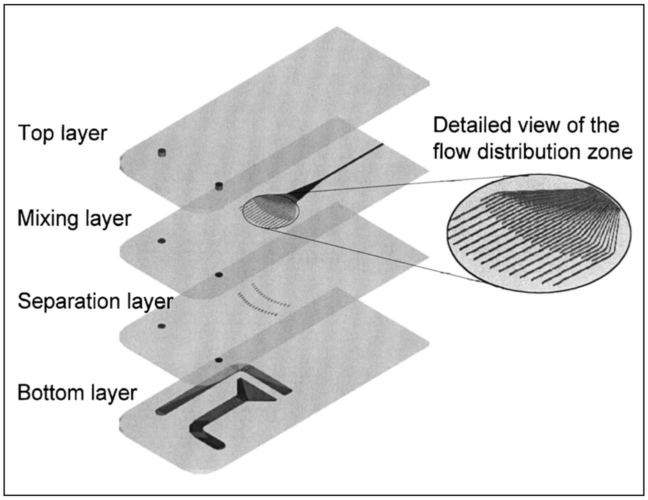

Figure 2. Explosion view comprising four microstructured glass layers, which form the triangular interdigital micromixer.

Individual layers can be structured, either leading to rectangular channel systems on top of the layer or to conduits, for example, to interconnect channels. Each micromixer is composed of layer architecture, bonded irreversibly (see Figure 2). In particular, the two feed sections have to be segregated and integrated in separate glass layers. These two glass layers are connected via a so-called separation layer. A top layer serves for interconnection to conventional fluidic peripherals.

Three glass micromixers were initially employed during the course of the investigations. Their 2×15 feed microchannels have a width of 60 μm, and the walls separating them are 50 μm thick. The depth of these channels, as well as for the whole microstructured area, amounts to 150 μm. A further fourth micromixer, termed *SuperFocus*, was developed at a later stage of the investigations, when the characterization of the three former devices was completed.

The three initial devices contain the same feeding and contacting principle based on interpenetrated, but separated, channels, termed interdigital elements. They, in turn, differ in the fluid passage following the start of contacting, simply named "mixing chamber." These mixing chambers have either a rectangular, triangular or so-called slit-type shape (see Figure 3). The shape termed "triangular" actually consists of a triangular and a rectangular section.

While the first two shapes belong to the pool of simple polygon geometries, chosen for the reasons explained in the Discussion section, the latter shape represents a more complex assembly composed of polygon or other elementary shapes. This type of shape largely corresponds to a design previously realized in stainless steel.

Figure 4 reveals a cut through the top part of the housing of such a steel mixer. A contacting zone shaped like an arc of a circle (termed a slit due to its appearance when observed from the front side of the housing) is connected to a small bore hole, followed by a large bore hole for fittings. Using the etching technique in glass, similar circular structures cannot be produced, since this type of microfabrication is only

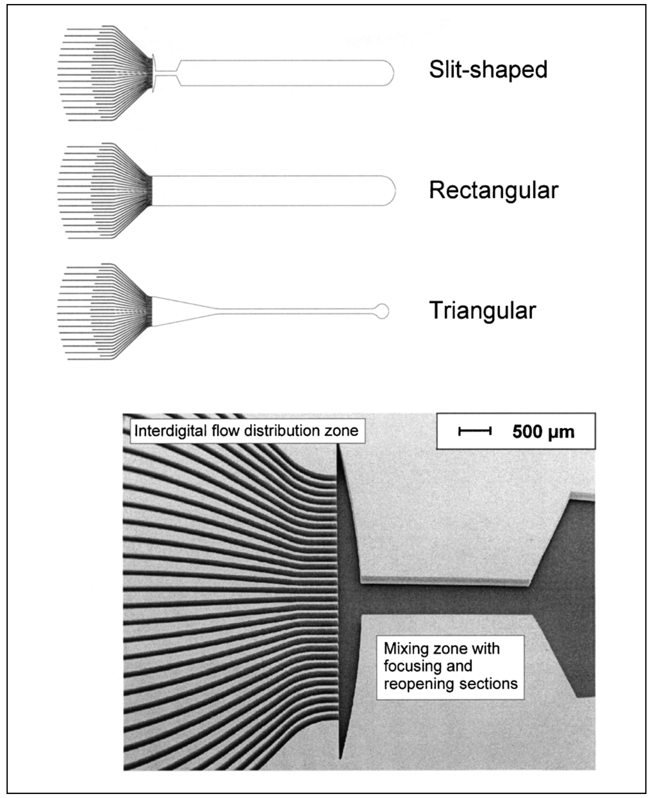

Figure 3. Top, design of three interdigital micromixers with different mixing chamber geometries slit-shaped, rectangular, and triangular; bottom, SEM image of one microstructured layer building the slit-shaped interdigital micromixer.

suited to produce planar architectures. Rather, the corresponding glass structure has a set of three channel zones with rectangular walls (see Figure 3).

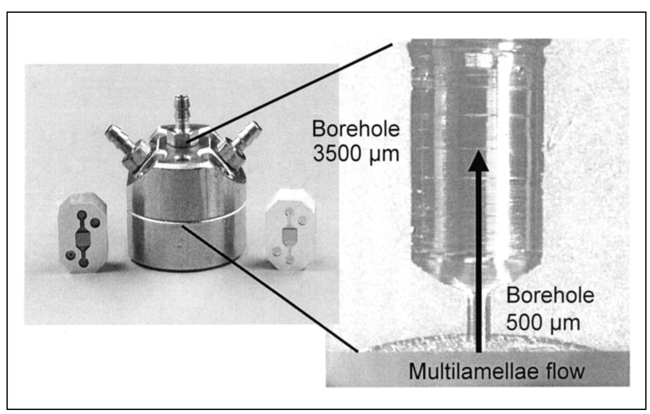

Figure 4. Assembled stainless steel interdigital micromixer and a cut through the steel top housing part.

The position of this detail in the assembled mixer is indicated. The cut reveals a slit-shaped mixing chamber.

Experimental Studies

Flow rate determination

The flow rates were determined by programming piston pumps used (KP 2000, Desaga Sarstedt-Gruppe, Wiesloch, Germany) and the corresponding accuracy of this setting was checked by occasional volumetric analysis control, that is, by gauging the capacity by liters.

Pressure drop determination

For functional control, the pressure drop is determined using the differential pressure meter 2003P (amenable up to 7 bar) of Digitron Instrumentation Ltd. (Hertford, U.K.) that was placed between the pump and micromixer. For instance, at a total flow rate of 50 mL/h, a pressure loss of 84 and 48 mbar was found, respectively, for the triangular and slit mixers. For a total flow rate of 1,000 mL/h, the respective pressure loss values were 1,647 and 1,059 mbar.

Recording of the mixing processes

Mixing was recorded by using digital recording with an optical microscope Stemi 2000-C, Zeiss Göttingen, Germany (magnification 13× . . . 100×) equipped with a video camera 3CCD Color Video Camera Power HAD, DX-950P, Sony, Tokyo, Japan (resolution 768×576×24).

Illumination

A defined illumination was established by using the light guide system KL 1500 with two goose necks, Schott, Wiesbaden, Germany. Due to problems with achieving a constant fixed illuminaton with the existing equipment, a new calibration was made for each experimental run (typically lasting a day) with each mixer by using standard solutions of the dye with a known concentration. The images of both calibration and experimental solutions were converted to greyscale format. Therefore, the concentration distribution was gathered when analyzing the images taken under fixed illumination with images of the calibration solutions. Using the AnalySIS software (Soft Imaging System GmbH, Münster, Germany), concentration profiles along the channel cross-section were obtained.

Flow pattern visualization

Accompanying the development of transparent micromixers, suitable imaging techniques are needed. The main issues were to achieve a simple and fast analysis rich in contrast and complex in information based on visual inspection or microscopic imaging. Two approaches were used.

The first approach relies on aqueous solutions with high concentrations of water blue (5 g/L, $6.25 \cdot 10^{-3}$M, Fluka) and pure water, which were fed through the interdigital micromixers. These blue-colored solutions provide excellent contrast due to the high solubility and large extinction coefficient of the dye. They are, in particular, suitable to image multilaminated systems arranged parallel to the direction of observation and multiphase systems such as gas/liquid and liquid/liquid (results not shown here, see, for instance, Hardt et al. (2001)). However, when assuming a layer structure which is horizontal or tilted with respect to the observer, it stands to reason that one cannot distinguish between a real mixed system and a layered fluid structure.

In the latter case, a second visualization approach was applied. Mixing of uncolored iron ion (Fe^{3+}) and rhodanide (SCN^{-}) solutions (81.3 g/L, 0.5 M $FeCl_3$, Merck p.a., and 40.5 g/L, 0.5 M NaSCN, Riedel-de-Haen, 99% purity), resulting in the formation of the respective brownish complex, turned out to provide a reasonable contrast and is free from any plugging phenomena. Differing from the water blue solutions, potentially providing information both on the fluid layer formation at the very beginning and during the course of mixing, the iron rhodanide system displays only the completion of mixing, linked to the color reaction. Thereby, mixing even in complex fluid systems, such as tilted lamellae, can be visualized.

Finally, one disadvantage of using the water blue imaging must be mentioned. Due to the large molecular weight of this dye, diffusion is certainly different from low-molecular weight species such as the iron ions. However, when using organic dyes, this feature is inherent to most molecules thereof, belonging to condensed extended aromatic systems.

Results

Flow patterns in the rectangular interdigital mixer

Typical flow patterns, obtained at individual flow rates equal or above 5 mL/h, in the rectangular mixer and their corresponding concentration profiles after residence times ranging from 4.3 to 1,140 ms, are displayed in Figure 5. First, the formation of lamellae, as a consequence of injecting the two water streams into the interdigital structure, can be observed via the corresponding dark and light sections. Figure 5

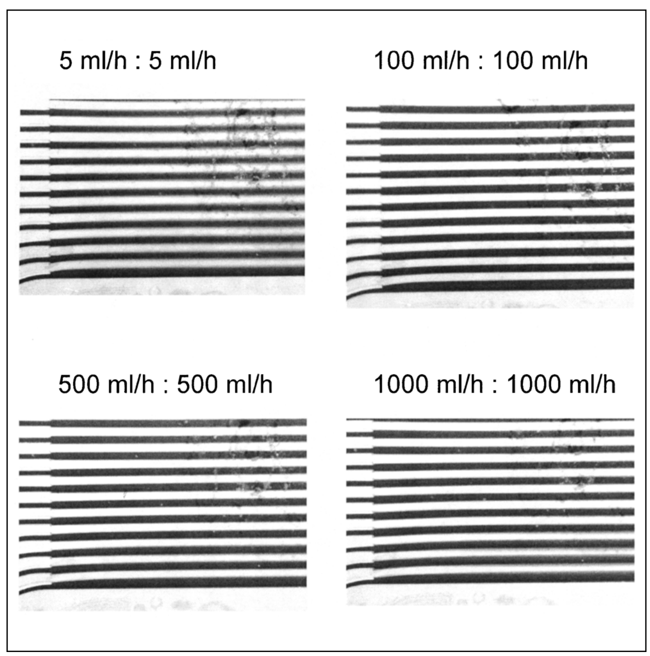

Figure 5. Multilamination flow patterns in the rectangular interdigital micromixer visualized by water blue imaging.

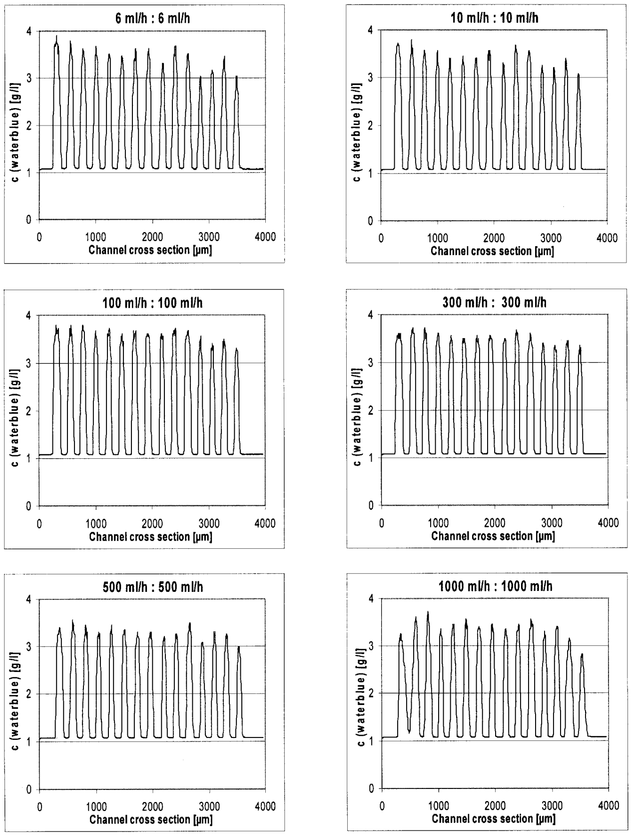

Figure 6. Cross-sectional concentration profiles for various total volume flows in the rectangular interdigital micromixer, taken at a distance of 4.86 mm (corresponding residence time interval: 1.14 s to 4.3 ms for flow rates ranging from 12 mL/h up to 2000 mL/h) from the inlet section.

The concentration profiles do not level at zero, since it was not possible to distinguish between solutions with a concentration lower than about 1 g/L due to insufficient contrast.

also displays that this flow pattern is independent of the flow rate in the investigated range. The distribution of color over the observed area, corresponding to approximately half of the mixing chamber, remains widely unchanged. A more thorough analysis of the concentration profiles at various total volume flows from 12 to 2,000 mL/h (4.3 to 1,140 ms) confirms this finding and gives further details on the mixing process (see Figure 6). Periodical profiles, stemming from the alternating feed of dye and water, are obtained even at the largest residence time. Within one concentration profile (at one volume flow), differences in concentration exist for the various lamellae. This may result from statistical fluctuations, such as, pulsating pumps (see the inner lamellae), or systematic deviations, such as by surface or flow distribution effects (see the three outer lamellae at large values of channel cross-section).

In order to determine the Reynolds and Péclet number regime corresponding to the prescribed volume flows, the diffusion constant of water blue was estimated based on the Wilke and Chang correlation for diffusion in liquids (Reid et al., 1987). The diffusion constant of water blue in water as a

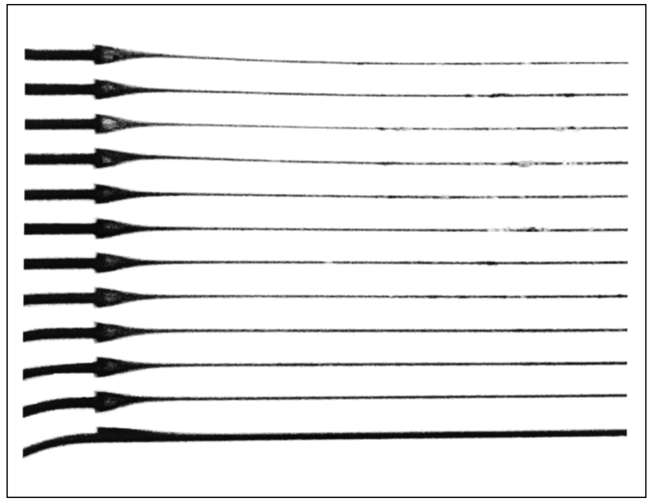

Figure 7. Flow pattern in the rectangular interdigital micromixers when using different individual flows visualized by water blue imaging.

By means of hydrodynamic focusing, lamellae thinning and thickening occurs.

solvent was determined as $D = 3.2 \times 10^{-10}$ m^2/s. The values of the liquid density and viscosity were assumed as those of water at 20°C and a pressure of one atmosphere. On this basis, the range of volume flow between 12 mL/h and 2,000 mL/h corresponds to a Reynolds number range between 2 and 341. The corresponding Péclet numbers lie between 6.41×10^3 and 1.07×10^6. All the Péclet numbers reported in this article are based on the diffusivity of water blue.

Due to the periodical profiles, it is evident that this process of mixing, which is solely based on diffusion, is not completed in the given mixing chamber, especially at high flow rates. Similar findings were also made by other authors (Branebjerg et al., 1996a). Therefore, it suggests itself to search for possibilities to speed up mixing in the interdigital mixers. In this context, Figure 7 shows a flow pattern observed in the rectangular mixer when choosing different flow rates of the single water streams (20 and 250 mL/h, respectively). The consequence is a narrowing down of the lamellae width of one fluid (from 110 μm to about 30 μm lamellae width) at the expense of increasing the width of the other. Although, by this procedure, the system as an entirety is not mixed faster, regions of particular interest, namely the thin lamellae, experience a faster change of their composition, that is, a penetration of molecules of the thicker lamellae. In the Discussion section, a possible application for this is identified, referring to carrying out chemical reactions with an excess of one reactant.

Flow patterns in the triangular interdigital mixer

In Figures 8 and 9, the hydrodynamics of fluid flow in the triangular mixer are demonstrated when performing similar experiments, as shown in Figure 5. For a total volume flow of 20 mL/h, separated light and dark sections become increasingly smaller when passing through the triangular zone and reveal a more homogeneous in color profile after entering the rectangular outlet zone. This is confirmed by a detailed analysis of the concentration profile (see Figure 9). For total

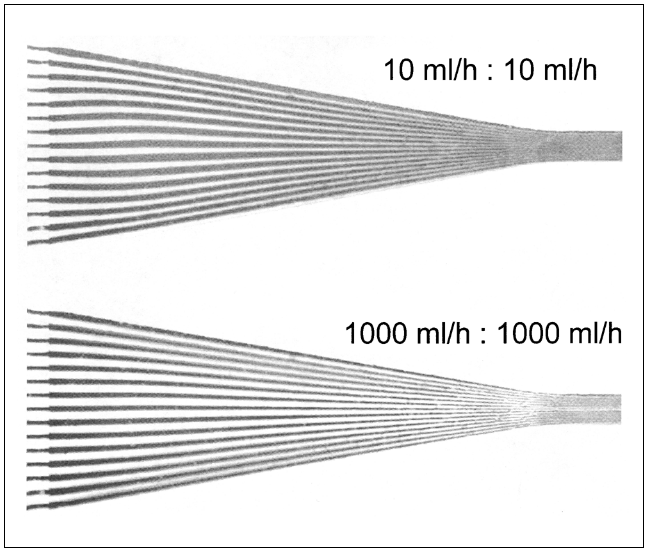

Figure 8. Multilamination flow patterns superposed by focusing in the triangular interdigital micromixer visualized by water blue imaging.

volume flows of 12 and 20 mL/h, more flattened profiles are obtained in comparison to those of the rectangular mixer. The profile at 12 mL/h indicates a higher degree of mixing in comparison to that at 20 mL/h due to the longer mixing time. Moreover, both profiles comprise strong deviations in concentration between the outer and inner lamellae (curved profile), in particular given for one of the outer lamella next to the mixing chamber wall. The origin for the curved profile is not totally clear, but may be correlated to a small bending of the cover of the mixing chamber, thereby slightly changing the optical path over the cross-section. For total volume flows equal to 200 mL/h or larger, even more complex concentration profiles are found that have too many peaks, that is, more than the number of generated lamellae.

Moreover, Figure 8 shows that not all lamellae have absolutely equal thickness as opposed to the observations of Figure 5. The thickness is decreasing from the interior towards the exterior. Figure 10 shows the individual thickness of the lamellae, which is derived by precise measurement of the respective high-contrast image at large magnification. This deviation from average probably is caused by the parallel orientation of the inlets, that is, the various lamellae flows have different angles with respect to the channels' direction, rather than being guided in the same direction. Thereby, lamellae width becomes slightly dependent on the channel position.

As depicted in Figure 11, choosing different flow rates of the single water streams in the triangular mixer leads to a further reduction of lamellae width which results in flow patterns characterized by thin and wide lamellae, becoming smaller towards the end of the triangular zone (see also Figure 7). The volume flow range between 20 mL/h and 200 mL/h translates to Reynolds numbers in the mixing channel between 17 and 170, and corresponding Péclet numbers between $5.34 \cdot 10^4$ and $5.34 \cdot 10^5$.

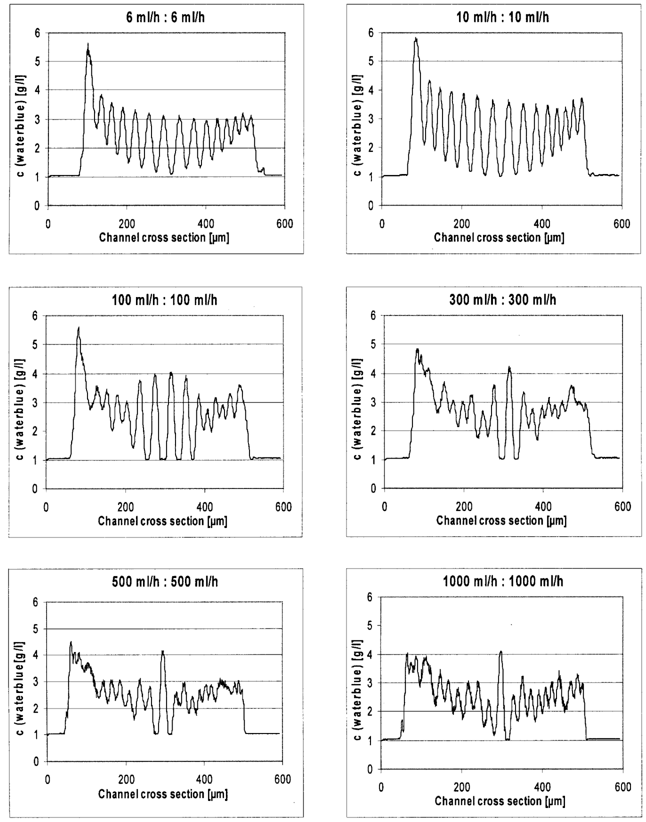

Figure 9. Cross-sectional concentration profiles for various total volume flows in the triangular interdigital micromixer, taken at a distance of 2.0 mm (corresponding residence time interval: 1.14 s to 4.3 ms for flow rates ranging from 12 mL/h up to 2,000 mL/h) from the beginning of the rectangular channel.

The concentration profiles do not level at zero, since it was not possible to distinguish between solutions with a concentration lower than about 1 g/L due to insufficient contrast.

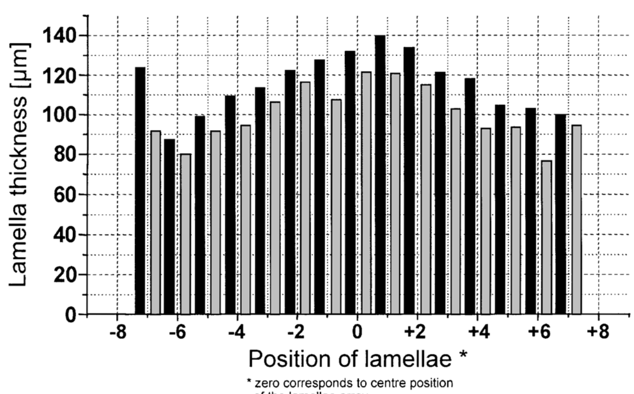

Figure 10. Thickness of light and dark lamellae for a 10 mL/h:10 mL/h flow in the triangular mixer, as defined in Figure 7.

The thickness was derived at a position of 1.5 mm distance from the feed inlet. It is believed that the difference between light and dark lamellae thickness is not due to experimental facts, but rather relies on the difficulty to correctly set the border between both.

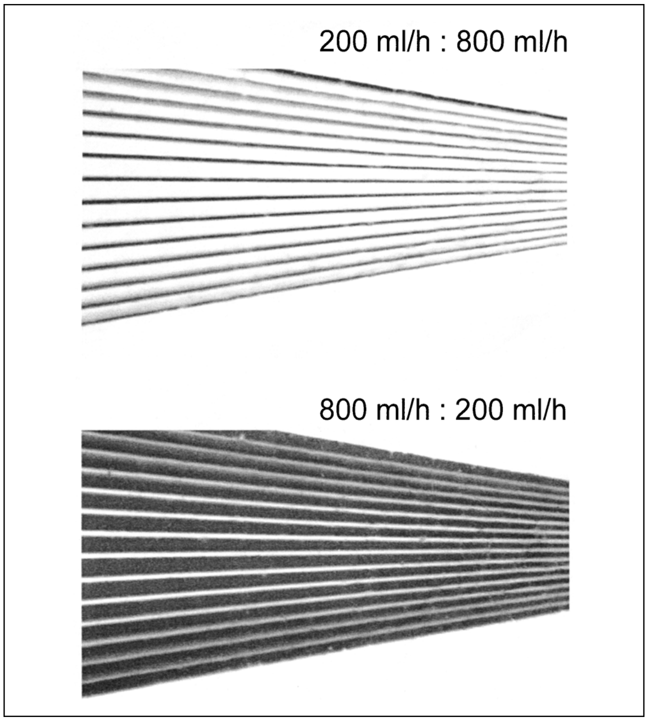

Figure 11. Flow pattern in the triangular interdigital micromixers when using different individual flows visualized by water blue imaging.

With hydrodynamic and geometric focusing, lamellae thinning and thickening occurs.

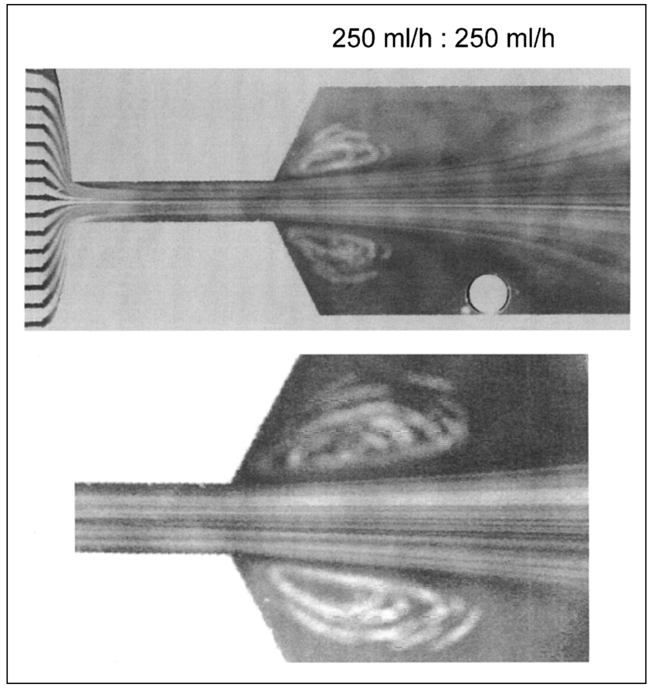

Figure 13. Multilamination flow pattern superposed by focusing and reopening/jet formation in the slit-shaped interdigital micromixer visualized by water blue imaging.

The total mixing chamber including focusing zone, small, and large rectangular channel attached is depicted (top). The total flow rate was set to a medium value. Additionally, the detailed view of the jet formation is given (bottom).

Flow patterns in the slit-type interdigital mixer

As can be seen in Figures 12–14, hydrodynamics tend to be more complex when using a slit-type interdigital mixer resulting in different flow patterns in the respective three zones of the mixing chamber—the slit area, small and large rectangular channel.

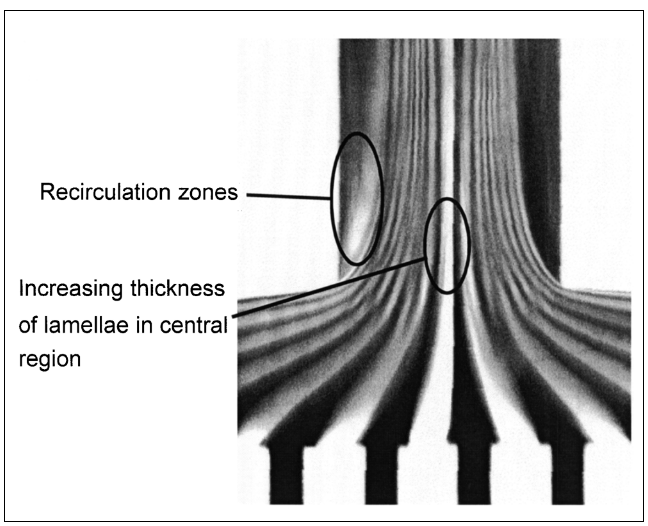

Figure 12. Multilamination flow pattern superposed by focusing in the slit-shaped interdigital micromixer visualized by water blue imaging.

A detail of the focusing zone and the small rectangular channel attached is given.

In the slit area, that is, the contacting section, focusing occurs; all lamellae are thinned when entering the following rectangular channel (see Figure 12; pattern at a flow rate of 500 mL/h:500 mL/h). However, the individual widths of the lamellae are not equal when entering the small rectangular channel, with the central lamella being much thicker as the others. A similar finding, but at a lesser extent, was made for the triangular mixer (see Figures 8 and 10).

In the small rectangular channel, a closer look reveals blurred shapes, which are different from the flow patterns of the rectangular and triangular mixers (see Figures 5 and 8). At high flow rates, some of the lamellae even seem to be broken. In addition, wakes are found on both sides of the entrance area to the mixing element.

In the wider rectangular channel, at medium or high flow rates, a jet is formed emerging from the narrow channel (see Figure 13; pattern at a flow rate of 250 mL/h:250 mL/h). This jet induces eddies at both sides of the mixing chamber. The shape of these eddies is well predicted by CFD simulations (not shown here). Although still differences in color are visible in the mixing chamber, the fluid system is clearly more dispersed as in the case of the rectangular or triangular mixers.

In contrast to that, at low flow rates, multilamination patterns are observed in all three zones—including the wide

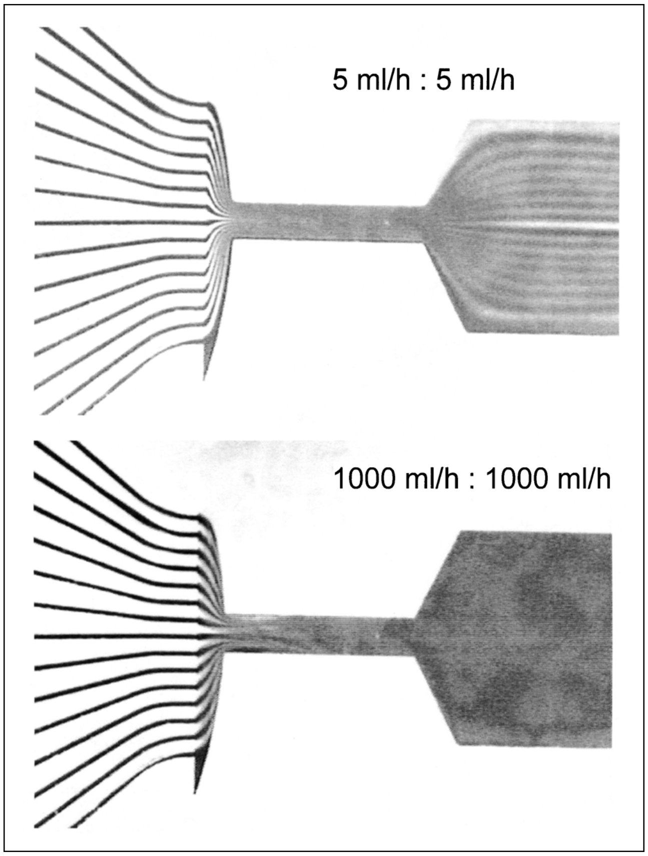

Figure 14. Multilamination flow pattern superposed by focusing and reopening/jet formation in the slit-shaped interdigital micromixer visualized by acid blue imaging.

The total flow rate was set to low (top) and high (bottom) values.

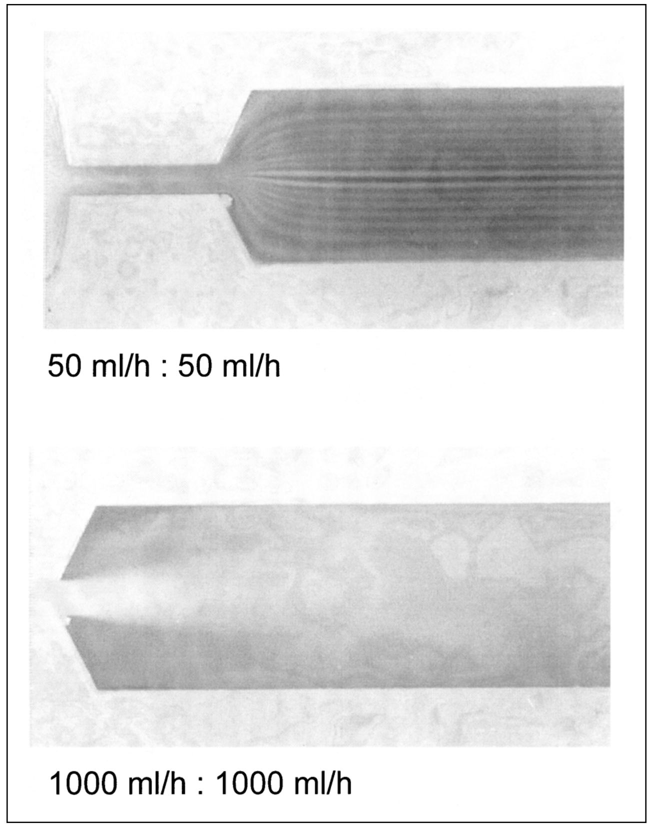

Figure 15. Multilamination flow pattern superposed by focusing and reopening/jet formation in the slit-shaped interdigital micromixer visualized by rhodanide imaging.

The total flow rate was set to low (top) and high (bottom) values.

rectangular channel (see Figure 14a; pattern at a flow rate of 5 mL/h:5 mL/h). At high flow rates, a nearly uniform color develops in this zone (see Figure 14b; pattern at a flow rate of 1,000 mL/h:1,000 mL/h).

Since the patterns of Figure 12–14 are more complex, and not totally understandable at first sight (see Discussion), it was decided to use the rhodanide reaction as a second means of flow and mixing visualization. Carrying out this process at low flow rates results in a multilaminated fluid architecture (see Figure 15a). This information is equivalent to that of Figure 15a. Hence, at low flow rates, the same information is revealed using either water blue coloring or the rhodanide reaction.

In contrast to that, at high flow rates, the corresponding images are considerably different (see Figures 14b and 15b; patterns at flows rate of 50 mL/h:50 mL/h and 1,000 mL/h:1,000 mL/h, respectively). Rhodanide imaging shows no color formation in the small rectangular channel. The corresponding flow even enters the wide rectangular channel as an uncolored jet flowing in an otherwise more or less homogeneously colored chamber. Imaging by water blue, in turn, resulted in blur-shaped flow at the entrance of the narrow rectangular channel becoming more homogeneous to the channel's end. The wide rectangular channel is homogeneously colored.

In all of the experiments described above, the total flow rate was in a range between 100 mL/h and 2,000 mL/h. This range corresponds to Reynolds numbers in the small rectangular channel between 85.15 and 1,702.94 and Péclet numbers between 2.67×10^5 and 5.34×10^6.

Apparently, the information obtained does not coincide, but, rather, appears to be complementary. On a first sight, the water blue results seem to indicate mixing already in the small rectangular channel, while the rhodanide results seemingly show this only deep inside the large rectangular channel. This apparent contraction is further addressed in the Discussion section and resolved in Part II of this article, which shows simulation results for the slit-type interdigital mixer.

Flow patterns in an optimized triangular interdigital mixer

The results of the triangular mixer demonstrated the success of focusing, although the reduction of lamellae width was not extreme, being a factor of about 6. The focusing in the slit mixer, having a similar focusing ratio, showed limits, that is, lamellae of different thickness, if the length of the focusing zone is not correctly chosen (see Figure 10). Hence, it is clear that, for a proper design of a focusing mixer, the ratio

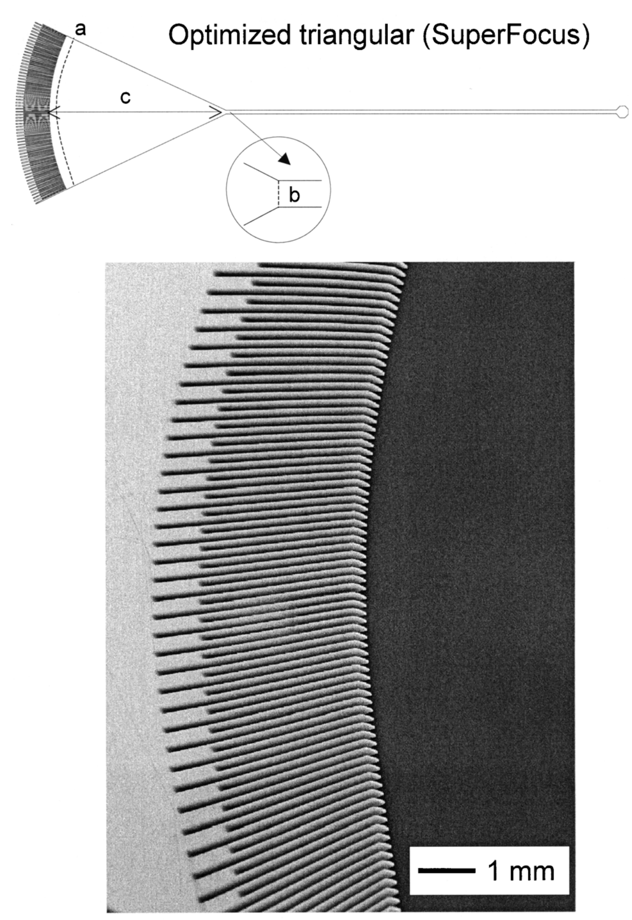

Figure 16. Design of an optimized triangular interdigital micromixer, termed SuperFocus, and SEM image showing detail of the corresponding feeding zone.

of the widths of the triangular (the width at the beginning) to the rectangular channel (termed *a* and *b*), and the length between the outlets of the feed zone and the beginning of the rectangular channel *c*, are of crucial importance.

In connection with that, an optimized mixer, termed SuperFocus, was developed (see Figure 16). The inlet section comprises 124 microchannels of a width of 100 μm. The length, width, and depth of the rectangular channel amount to 70, 0.5, and 0.5 mm, respectively. The ratio *a/b* was set to 40 which is as large as possible due to current pressure and fabrication issues. The latter arise from adaptation of the SuperFocus design to a special format for the outer dimensions, which the mikroglas company established for reasons of modularity and compatibility. This provides a maximum size for the parameter *a*. The format is similar to the slice size typically used for optical microscopy. Pressure loss issues demand not to reduce the other parameter much more beyond the value chosen for the present design (500 μm). The length *c* was chosen equal to that of the current triangular mixer. A curved arrangement of the inlets was chosen in order to avoid the deviations of lamellae thickness shown in Figure 8 (the triangular mixer had to a planar array of inlets).

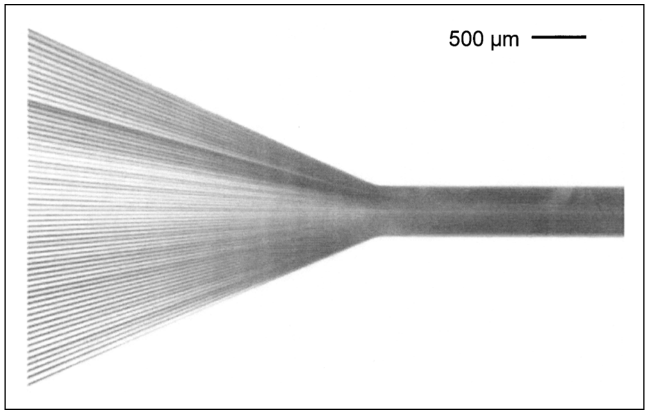

Figure 17. Multilamination flow pattern superposed by focusing in the SuperFocus interdigital micromixer visualized by rhodanide imaging.

The image demonstrates fast mixing at large total volume flow (5 L/h), and is completed within the micromixer.

For reasons of better imaging of the individual lamellae, different individual flow rates of the two aqueous reactant solutions were chosen (4 L/h:1 L/h).

Simulation results predict that mixing is completed within less than 10 ms for the SuperFocus mixer which is a considerable improvement in performance as compared to the triangular mixer (see Part II of this article). For instance, at a total volume flow of 1,000 mL/h, only fluid passages of less than 10 mm are needed, hence, assuring that mixing should also be completed within the device at high flows.

Figure 17 confirms that indeed this predicted new quality of focusing is obtained when using the SuperFocus mixer. At a large volume flow of 8,000 mL/h (at a pressure drop of about 2.5 bar), a clear solution is observed near the feeding zone when using the rhodanide visualization. The closer the streams approach the rectangular channel, that is, the more they are being focused, the more intense color is formed at the interface of the lamellae. Shortly after entering the rectangular channel, a homogeneous color is observed, indicating completion of mixing. In Part II of this article it is shown that the corresponding experimental mixing time amounts to a few ms which corresponds well to the simulation results. The maximum flow rate of 8,000 mL/h corresponds to a Reynolds number in the mixing channel of 4,427 and a Peclet number of 1.39×10^7.

Discussion

Micromixer design requires knowledge about the underlying hydrodynamics which is at best gained via direct observation of the underlying flow pattern. Authors focusing on industrial mixing processes especially highlight the benefits of using laboratory glass or other transparent vessels for hydrodynamic studies (Tatterson, 1994). The mixer devices reported here are completely made of glass, hence, allowing imaging of flow from basically all directions (different, for example, from only glass-caped devices). In combination with the imaging techniques of high resolution and contrast presented here, even subtle phenomena are detectable.

As a result, the experiments described in this article reveal a scenario of flow patterns, ranging from simple architectures of interpenetrating lamellae to more complex patterns with jets or eddies, and, so far, unidentified patterns characterized by blurred shape textures. While the first pattern relies only on diffusion mixing, jets and eddies induce convective phenomena, which are typical for turbulent mixing, where multilamellae structure are superposed and modified.

Applicability to visualization of a well-known flow pattern, multilamination, and modifications thereof

The investigation of a well-known flow pattern, namely multilamination, was first chosen to demonstrate the applicability of the visualization via water blue. Moreover, some modifications by posing geometric and hydrodynamic constraints on the laminated flow for distinct interdigital mixers, as given in Figure 3, could be identified.

The separated fluid layers of Figure 5 and the periodical concentration profiles of Figure 6 show that the residence time provided is too low to result in a notable mixing. This existence of multilamination flow patterns is in accordance with findings reported by other authors (Branebjerg et al., 1996b; Floyd et al., 2000). In addition, the absence of detectable mixing effects of reasonably large throughputs corresponds to theoretical predictions of mixing in similar multilamellae structures (see, for example, Branebjerg et al. (1996a)).

Consequently, mixing can only be completed within many previously described mixers at very low flow rates, typically below 5 mL/h, and correspondingly needs large residence times, typically several seconds. This is not necessarily faster than mixing achieved by intense stirring in small laboratory vessels, but is, as to be expected, faster than mixing in industrially used reactors, such as, stirred tanks (Tatterson, 1994). Moreover, it is naturally faster than mixing in laminar-flow microstructures with only a mixing tee-configuration, that is, without any further mixing element.

Decreasing mixing time for pure diffusion-based mixing of multilamellae can be achieved by generating thinner lamellae via narrower channels, which is restricted by the capabilities of today's manufacturing techniques and by pressure loss limits. Hence, one has to rely on focusing techniques (Floyd et al., 2000; Knight et al., 1998).

Focusing as a means to intensify mixing

Focusing of fluid streams, that is, narrowing of the lamellae width, is a proper and simple method to intensify mixing. Special micromixers have been described employing this phenomenon (Veenstra et al., 1998). Hydrodynamic focusing refers to the reduction of the layer width of one fluid at the expense of the other. Practically, this is achieved by means of increasing the ratio between the two individual liquid flow rates.

Figure 7 shows this hydrodynamically induced reduction of lamellae width. This facilitates a fast penetration of molecules dissolved within the thinner lamellae with those of the larger lamellae. By this method, only the early stages of the mixing process are speeded up, whereas nothing is gained in the final stages. This situation, hence, is desired in the case of mixing using an excess of one reactant, which actually is applied in a number of chemical processes. This provides one route for the fast carrying out of such reactions by using one liquid (that contains a reactant) in excess; the corresponding lamellae compress the deficient liquid/reactant lamellae. Consequently, hydrodynamic focusing in micromixers provides a simple means to speed up such processes, if the increase in waste production due to a larger amount of nonreacted species is not prohibitive (such as when avoided by recycling).

Focusing by means of geometry restriction, that is, the narrowing of the fluid passage (Floyd et al., 2000; Veenstra et al., 1998; Branebjerg et al., 1996b), is another, more generally applicable, means of reducing lamellae width. This variant, hence, may be termed geometric focusing, in analogy to the term used above. The flow patterns and concentration profiles of Figures 9 and 17 clearly show the onset of mixing as a result of focusing. However, geometric focusing may also cause lamellae tilting (see Figure 9 and Part II of this article). Figure 9 shows the superposition of several periodical profiles, which are typically known for multilamination patterns, giving interference profiles. This implies that several lamellae stacks are observed along the optical path as a result from their tilting. Tilting causes a more undefined flow pattern in the micromixers and, hence, its impact on the mixing process is more difficult to predict.

More detailed conclusions on the impact of focusing will be drawn in Part II based on fluid dynamic simulations. It will be shown that optimized focusing can result in a huge decrease of mixing time down to only a few ms. In connection with that, liquid mixing inside a triangular mixer can be completed easily.

Naturally, geometric and hydrodynamic focusing can be combined, such as, when operating the triangular mixer with largely different flow rates of the two liquids (see Figure 11).

Jet mixing

The use of the water-blue imaging solution is in particular beneficial when observing mixing phenomena with fine details. This is required, for example, for imaging the rotational flow phenomena in the slit-type mixer depicted in Figures 13 to 15. Correspondingly, generation of new and optimization of existing microdevices is significantly facilitated using a glass mixer design. By these means, a number of flow phenomena in interdigital mixers, being desired or undesired, were identified recently.

Undesired phenomena, for instance, refer to the existence of wakes and lamellae of unequal thickness, as shown in Figures 12 and 13. Before the development of the glass devices, that is, when using the formerly developed stainless steel devices, all these phenomena were unknown. Only in one case —a result that could not be explained by simple diffusion mixing (Ehrfeld, 1999))—it was speculated that a flow phenomenon in addition of multilamination exists, termed parasitic mixing in absence of a more precise definition (Ehrfeld et al., 1999).

Using glass mixers in combination with applying the imaging solutions now allows for a proper judgment of such assumptions. In connection with that, jet mixing has been identified to be one suitable explanation for the origin of this second, parasitic effect (see Figure 13 in this article and compare it to the findings of Ehrfeld et al. (1999)). In large-scale mixers such a coexistence of many mixing processes is common and well-known, usually with the dominance of one (Tatterson, 1994).

Lamellae tilting

Lamellae tilting were observed even for the liquid flow in the triangular mixer (see Figure 9), but are much more pronounced for the slit-shaped mixer. Mixing in the latter device, at flow rates ranging from about 250 to 1,000 mL/h, results in the formation of a nearly homogeneous color already in the first narrow rectangular channel next to the slit zone. However, simple estimations of mixing time based on diffusion (for example, given in Branebjerg et al., 1996a), as well as the results of the rhodanide reaction (see Figure 15), suggest that mixing cannot be completed and that, therefore, this experiment cannot be explained by the sole action of multilamination mixing. Hence, an additional phenomenon has to be present. However, this cannot be identified solely on the basis of the experimental results, but demands for flow simulation. The results, in connection with this, predict lamellae tilting and winding to occur. A much more detailed description of this hydrodynamic feature is given in Part II of this article.

It has to be pointed out here that a clear experimental verification of complex flow patterns, such as tilted and wound lamellae, could only be made using the two-fold visualization approach. The water blue coloring, relying on solutions colored already before mixing, and the rhodanide reaction, yielding a colored mixture, give complementary information.

Conclusions and Outlook

With the development of the glass mixers, provided by the mgt mikroglas AG company (Freitag and Dietrich, 2000), an easily accessible way has been paved towards gathering hydrodynamic information for micromixing devices with a large design variety, with the four designs presented here being only a first and not the most complicated choice thereof. The simple injection of liquid streams, either colored or colorless, but reacting to a colored product, requires little technical expenditure and is performed quickly.

The results presented have extended the know-how base with regard to chemical processing in interdigital mixers. For instance, in the framework of an industrial project, it was very obvious that the triangular interdigital mixer was the adequate choice for lab investigations of a quick, industrially applied reaction (Clariant process), as evidenced by the final success, which was an increase in yield by about 25% (Hessel et al., 2002).

Moreover, a much faster interplay between design development and realization of new mixing devices meanwhile has been achieved. The SuperFocus glass mixer was optimized with regard to mixing speed and throughput. Mixing time is considerably improved compared to the former triangular mixer (see also Part II of this article). In addition, the mixing chamber of the interdigital steel mixers was changed, which is now similar to the shape of the triangular mixer, that is, the former slit geometry was changed to trapezoid. Several other mixers are under development.

The use of the glass microsystems as ideal diagnostic tools for process monitoring has been extended to the observation of contacting immiscible media, which yields dispersed systems. This has been described elsewhere recently and has theoretically transferred currently to much more realistic model systems and even to reacting media which enables a much deeper process understanding (Herweck et al., 2001).

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