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## Modeling of Mixing Process in Microreactors Used for Biofuel Production.

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# Modeling of Mixing Process in Microreactors Used for Biofuel Production

## نمذجة عملية الخلط بالمفاعلات الدقيقة المستخدمة في إنتاج الوقود الحيوي

Salah H. El-Emam, Mohamed Nabil Sabry, Mohamed H. Mansour and Mahmoud A. Shouman

### KEYWORDS:

*Biodiesel production* –  
*Passive micromixer* –  
*Mixing* –  
*Multilamination – Ribs*

*المخلص العربي:* - هناك اهتمام متزايد باستخدام تقنية المفاعلات الدقيقة لإنتاج وقود الديزل الحيوي نظرا لكفاءتها العالية. العنصر الأهم في المفاعل الدقيق هو الخلط الدقيق والذي يتحكم بشكل مباشر في كفاءة المفاعل. تم تطوير خلط دقيق مبني على نظرية تعدد الصفائح ليتناسب مع متطلبات خلط الزيت والكحول المستخدم في إنتاج وقود الديزل الحيوي بالمفاعلات الدقيقة. تم عمل نمذجة لعملية الخلط والسريران المستقر بالخلط الدقيق وذلك بهدف دراسة وتحسين أداء الخلط ليتناسب مع متطلبات التطبيق. تم تحليل أداء التصميم الابتدائي للخلط. كما تمت دراسة تأثير استخدام أشكال مختلفة لوحدة الخلط الدقيق. في النهاية تم اختبار الشكل المطور باستخدام مجموعة مختلفة من مخاليط السوائل. وقد أظهرت النتائج فاعلية كبيرة للشكل المطور للخلط بما في ذلك للمخاليط منخفضة الانتشارية كما هو الحال بالنسبة لخليط الزيت والكحول

**Abstract**—There is a growing interest in microreactor technology for biodiesel production due to its high conversion efficiency. The most important part of the microreactor is the micromixer, which directly controls reactor efficiency. In this paper, an efficient unflow passive micromixer configuration based on multilamination principle is developed, which is suitable for the low Reynolds numbers encountered in this application. The micromixer is modelled for the purpose of its shape optimization to develop a micromixer that is applicable for biodiesel production. A preliminary design of the micromixer is initially analyzed over a range of Reynolds numbers from 0.1 to 10. Effects of different modifications of the preliminary design are investigated. Finally, the developed micromixer is modeled using different fluid mixtures. Results show satisfactory mixing quality with relatively low pressure drop even with low diffusivity fluid mixtures such as oil and alcohol mixture used in biodiesel production. The configuration also helps enhancing

mass transfer by diffusion, which is the dominating mixing mechanism at low Reynolds numbers.

### I. INTRODUCTION

**B**IOFUELS are promising energy sources that can replace fossil fuels. One of the most viable liquid transportation fuels is biodiesel fuel which is considered an alternative for diesel fuel. Biodiesel is commonly produced by the transesterification process, which is a reaction between oil and an alcohol of short chain in the presence of a catalyst. The product of such reaction is a mixture of biodiesel and glycerol. This process is usually held in a batch reactor with a residence time ranging from several minutes to hours in order to reach a suitable yield. It has been reported that the use of microreactors in biodiesel production could be more efficient with a reasonable time of few minutes [1].

Since biodiesel production requires mixing of the two main liquids; oil and alcohol before the reaction takes place efficiently, a micromixer is usually used before entering the microreactor. In general, micromixers can be classified as active and passive. Active type can be achieved using an external energy source that produces turbulence to enhance the mixing process. On the other hand, passive type needs no

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external energy source and basically, depends on different geometrical parameters to guide the flow during the mixing process. Over the years, both types of micromixers have been frequently reviewed [2-5]. Different proposed configurations of passive micromixers have been introduced. They depend on either multilamination principle (i.e. creation of multiple interfaces) to increase molecular diffusivity or shape optimization to increase convective mixing.

The influence of using different types of micromixers in the production of biodiesel has been investigated both experimentally and numerically. Sun et al. [6] have experimentally investigated the influence of using four different commercially available micromixers; T-shape, J-shape, and two other commercial multilamination micromixers in transesterification of cottonseed oil and methanol with KOH as the catalyst. These available micromixers are T-shape, J-shape, and two other commercial multilamination micromixers; a rectangular interdigital micromixer (RIMM) and a slit interdigital micromixer (SIMM-V2). The yield obtained using multilamination micromixers was about double of that obtained with T and J shapes. This high yield was reached on the expense of a relatively high pressure drop across the micromixer. SIMM-V2 was also used by Sun et al. [7] for the production of biodiesel from high acid value oils. A high FAME yield was reached but no pressure drops were reported. Santana et al. [8] held a numerical comparison of *Jatropha curcas* oil and ethanol mixing over a range of Reynolds numbers (Re) from 1 to 160 in three different micromixers; T-channel, and two T-channel with different circular obstructions arrangements. They reported a mixing quality of 0.75 at low Re number and reached 0.99 at high Re number across the T-channel with alternate circular obstructions micromixer. Such results were obtained at a very high pressure drop ranging from 2 to 178 bar, respectively. Alam et al. [9] also investigated the effect of circular obstructions but in a curved microchannel instead of a straight one. The mixing performance was improved compared with the smooth curved microchannel mixer and T-micromixer with circular obstructions. The et al. [10] investigated a novel planar micromixer with cross inlets and special shaped mixing units to create vortices located along the channel length at low Re numbers. Satisfactory mixing quality with relatively low pressure drops were reached. Xia et al. [11] also investigated a novel planar micromixer with similar inlets and gaps and baffles over a range of Re number from 0.1 to 60. High mixing efficiency and relatively low pressure drop due to short mixing length were achieved.

Ait Mouheb et al. [12] numerically investigated the mixing of two fluids A and B having the properties of water in T-shaped and cross-shaped micromixers, and Re number ranging from 5 to 600 calculated at inlets (same flow rate). Cross-shaped micromixer showed significantly higher mixing quality at Re number greater than 75. For T-micromixer, it has been noticed that the mixing quality changed with Re number with three different mixing regimes. At low Re number (lower than ~ 50), the flow is considered to be stratified and the mixing is entirely due to molecular diffusion resulting in a decrease of

mixing quality with the increase in Re number, due to shorter residence time. For moderate Re number (lower than ~ 150), vortices are created, which causes a slight increase in mixing quality considering that diffusion is still the main mixing principle. For higher values of Re number, engulfment flow is observed resulting in a rapid increase of the mixing quality as the convection-dominated mixing mechanisms are observed. Similar results have been reported by Soleymani et al. [13]. A novel planar scaled-up passive micromixer with uneven interdigital inlets considering a teardrop obstructions arrangement along the mixing channel was experimentally investigated by Cook et al. [14]. Considerable mixing efficiencies were achieved for all tested Re numbers but no pressure drops were mentioned. Re number of 25 was the critical point, which separate the stratified flow, mixing zone and the vortex flow mixing zone.

The objective of this paper is to introduce and develop an efficient passive micromixer configuration based on multilamination principle that is applicable for biodiesel production. A numerical study is held to investigate the influence of different parameters on mixing performance. The study is conducted for Re number usually encountered in biodiesel production, which ranges from 0.1 to 10, where the flow is considered stratified.

## II. COMPUTATIONAL MODEL

Figure 1 shows the fluid domain of the proposed preliminary configuration of a micromixer based on multilamination principle. As the flow is usually assumed laminar in microchannels due to low-pressure requirements, creating multi-mixing zones is required to increase mass transfer by diffusion. For present work, the micromixer is divided into two zones, induction zone and mixing zone. Induction zone comprises a main entering channel and distribution runners for each fluid. Firstly, both fluids are divided into multi-streams flow using the separators at the end of the induction zone. Then, each stream flows through its runner. Finally, they enter the mixing zone to create multiple interfaces between both fluids for the purpose of enhancing diffusion. For mixing study, only the fluid domain after section A-A is considered. Oil and alcohol enter the mixer from inlets of fluid 1 and inlets of fluid 2, respectively. A low diffusivity mixture of soybean oil [15] and methanol with the physical properties listed in Table 1 was used for flow simulation. A methanol to oil molar ratio of 6:1 was assumed. Both diffusion coefficients were calculated using Wilke-Chang correlation for diffusion of non-associated solvents [16]. The temperature was assumed constant and equal 60 °C. According to Vignes [17], The activity-corrected coefficient of interdiffusion was calculated to be  $8.13 \times 10^{-10} \text{ m}^2/\text{s}$ , which doesn't differ from the diffusivity of oil in methanol mentioned in Table 1. So for further discussion, the activity-corrected coefficient of interdiffusion is assumed to be equal the diffusivity of oil in methanol.

Computational fluid dynamics (CFD) simulations are carried out to analyze the flow and mixing behavior within the

micromixer. The flow is assumed multicomponent, steady, laminar, isothermal, Newtonian and incompressible. The commercial CFD software ANSYS Fluent 15.0 is used to solve the three dimensional mass, momentum, and species conservation equations using finite volume method. Dilute approximation is used to model species transport due to diffusion. The SIMPLE algorithm is used to solve the pressure-velocity coupling where the spatial discretization QUICK scheme method is used to solve both momentum and species transport. Scaled residuals of  $10^{-5}$  are used as a convergence criterion.

For all cases, the mass flow rate has been calculated for the mixture based on Re number, oil density, and an area equal to  $240 \mu\text{m} \times 240 \mu\text{m}$ . Reynolds number was calculated using oil properties and a hydraulic diameter of  $240 \mu\text{m}$ . Mass flow inlet conditions are used for both inlets assuming normal flow, zero-gauge pressure outlet for exit face, and no-slip wall condition for the rest. Slip wall condition would have been important to consider for smaller channel spacing (S) [18].

The mixing quality (M) is calculated based on the standard deviation of mass fraction of oil ( $\sigma$ ) in exit section, as illustrated in the following equations:

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (c_i - \bar{c})^2}{N}} \quad (1)$$

$$M = \left(1 - \frac{\sigma}{\sigma_{max}}\right) \times 100\% \quad (2)$$

Where  $N$  is the number of nodes inside cross section,  $c_i$  is oil mass fraction at node  $i$ ,  $\bar{c}$  is oil average mass fraction, and  $\sigma_{max}$  is the maximum standard deviation, calculated using the following equation:

$$\sigma_{max} = \sqrt{\bar{c}(c_{max} - \bar{c})} \quad (3)$$

Where  $c_{max}$  is the maximum oil mass fraction that equals unity.

TABLE 1  
PHYSICAL PROPERTIES OF FLUIDS

Component	Molecular weight (kg/kmole)	Density (kg/m <sup>3</sup> )	kinematic viscosity (mm <sup>2</sup> /s)	Diffusivity (m <sup>2</sup> /s)
Soybean oil [15]	872	894.1	17.47	$8.843 \times 10^{-10}$
Methanol	32.04	755	0.464	$4.918 \times 10^{-10}$
Oil used by Santana et al. [8]	879.23	911.2	32.5944	$1.2 \times 10^{-9}$
Ethanol used by Santana et al. [8]	46.07	789	1.5	$1.2 \times 10^{-9}$
Water used by Alam and Kim [19]	18.02	999.8	0.9	$1.2 \times 10^{-9}$
Ethanol used by Alam and Kim [19]	46.07	789	1.52	$1.2 \times 10^{-9}$

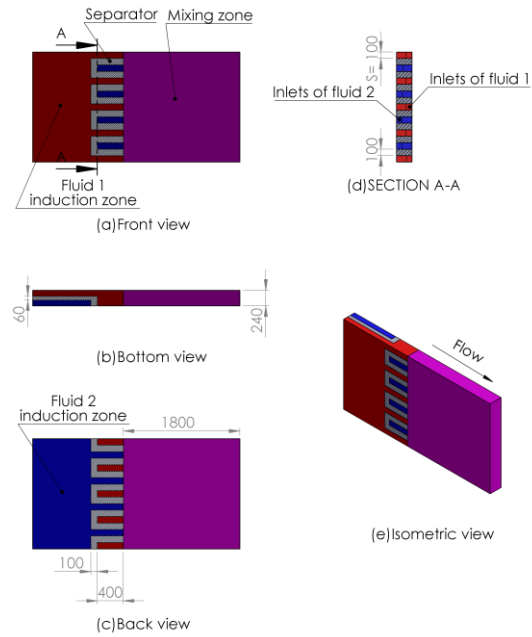


Figure 1. Description of induction zone and mixing zone of fluid domain. (Dimensions in microns)

ANSYS Meshing 15.0 software was used to discretize the domain into hex cells using CutCell algorithm. The domain shown in Figure 2.a was extracted from the original domain for preliminary mesh dependency study at which both sides of the domain were assumed to be a symmetry plane. Four different maximum cell size were investigated and the mixing quality was calculated at Re number of 0.1. Table 2 shows the standard deviation, mixing quality, and percentage deviation of mixing quality at exit plane. It is found that mixing quality slightly changes with mesh size. The case of maximum mesh size of  $5 \mu\text{m}/\text{cell}$  was selected as a reference case in calculating the percentage error. As indicated in Table 2, the error for all cases ranges from 0% for reference case to 0.87% for a maximum cell size of  $10 \mu\text{m}/\text{cell}$ . Therefore, a maximum mesh size of  $8 \mu\text{m}/\text{cell}$  was selected for simulation, which means 1,694,584 nodes for the whole domain.

The computational model was validated by comparing the results given by Soleymani et al. [13] For regular T-micromixer with the present model results. Figure 3 shows comparison between the two models for three different Re numbers. The maximum variation was found at higher Re numbers and reached 2.01% at Re number of 80.

TABLE 2  
EFFECT OF MESH SIZE

Maximum mesh size ( $\mu\text{m}/\text{cell}$ )	$\sigma$	M	% error
10	0.18497166	51.92%	0.87%
8	0.185772198	51.71%	0.47%
6	0.187048798	51.38%	-0.18%
5	0.18669921	51.47%	0.00%

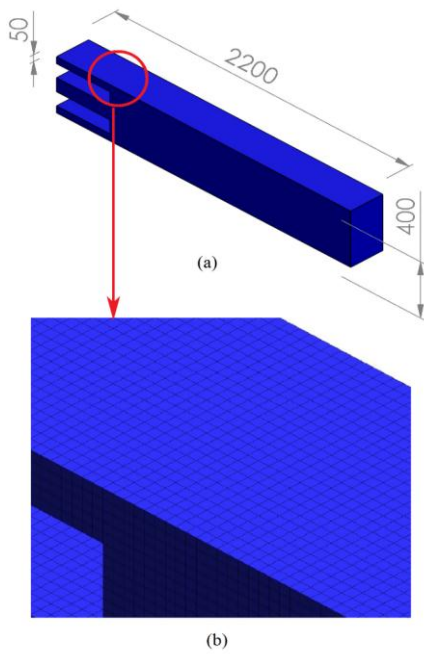


Figure 2. (a) Extracted domain for mesh dependency test, (b) mesh sample. (Dimensions in microns)

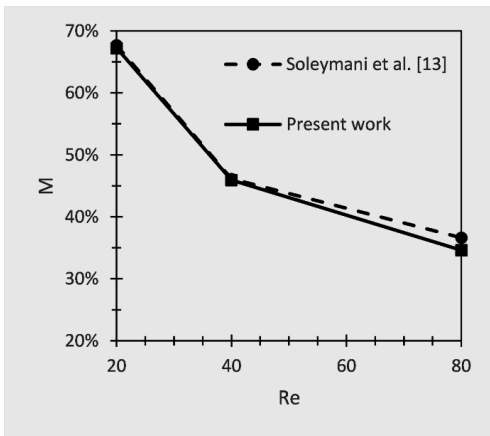
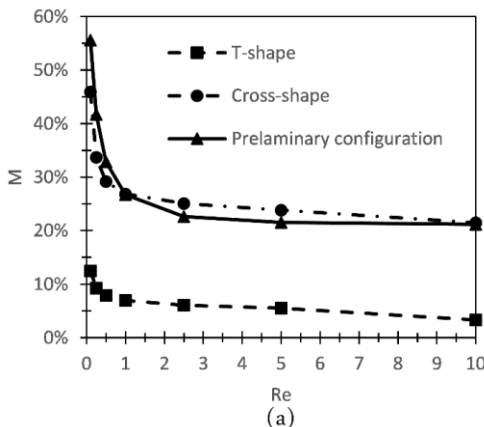


Figure 3. Comparison between Soleymani et al. [13] and present work.



### III. RESULTS AND DISCUSSION

#### 3.1 Comparison with T-shape and Cross-shape micromixers

Mixing quality and pressure drop at the exit of the preliminary micromixer were investigated for Re number ranging from 0.1 to 10 and compared with the T-shape and Cross-shape micromixers shown in Figure 4. Seven different Re number values were chosen for comparison; (0.1, 0.25, 0.5, 1, 2.5, 5, and 10). Figure 5.a shows that creating multi-mixing zones in the present work increases mixing quality compared with that of the T-shape and Cross-shape micromixers over the whole Re number range. Meanwhile, as the cross sectional area is increased, pressure drop across the mixer is decreased as shown in Figure 5.b.

It is noteworthy that the mixing quality for the preliminary configuration is 4 to 6 times that of the T-shape micromixer and almost the same compared to the Cross-shape micromixer. It is also noticed that with the increment of the Re number, the mixing quality decreases until it reaches a minimal value. This is probably because the flow is still in the stratified zone where the mixing is mainly due to mass transfer by diffusion. As Re number increases, the time available for mixing decreases resulting in a decrease in mixing quality

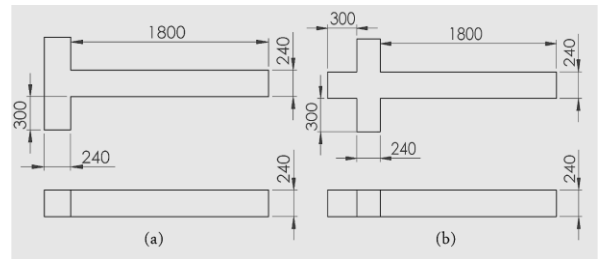


Figure 4. Front and top views of (a) T-micromixer and (b) Cross-micromixer used for comparison. (Dimensions in microns)

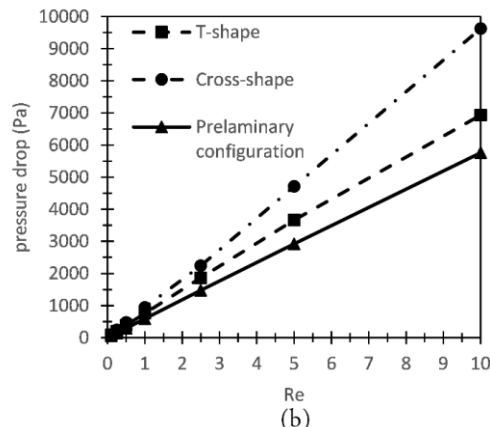


Figure 5. (a) Comparison of mixing quality with Re number between T- shape, Cross-shape, and preliminary micromixer configuration, (b) Comparison of pressure drop with Re number between T-shape, Cross-shape, and preliminary micromixer configuration

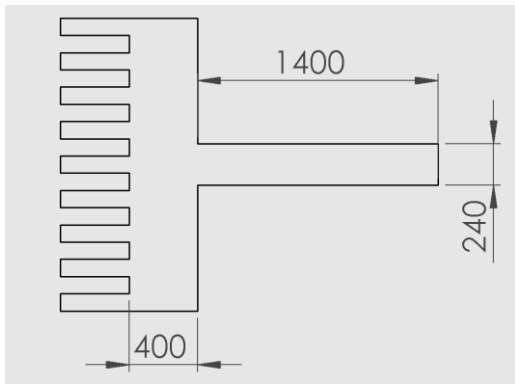


Figure 6. Micromixer with abrupt contraction. (Dimensions in microns)

### 3.2 Effect of abrupt contraction

An enhanced design by introducing an abrupt contraction section was investigated. It is expected that contraction would enhance mixing quality by thinning streamlines. Such effect can be shown in Figure 8, which illustrates the change in oil mass fraction at a mid-plane parallel to the streamlines and at exit plane at Re number of 5 for the case with and without

abrupt contraction. Figure 6 shows a schematic of a micromixer with abrupt contraction. After contraction, a square cross sectional area was assumed.

Figure 7 shows the effect of abrupt contraction on mixing quality and pressure drop with Re number as compared with the case without contraction. Figure 8 explains this result by showing concentration contours with and without abrupt contraction. It is noticed that the mixing quality almost doubled by contraction for Re numbers greater than 0.5.

### 3.3 Effect of spacing between separators

Figure 9 shows the effect of increasing the spacing between separators (S) illustrated in Figure 1.d at Re number of 5 and an abrupt contraction on both mixing quality and pressure drop. It is noticed that for a spacing of 150  $\mu\text{m}$ , the pressure drop decreased by 26% compared to the case of 100  $\mu\text{m}$ , while the mixing quality was reduced by only 6.5%. For greater increment in spacing, mixing quality was affected much more than the pressure drop.

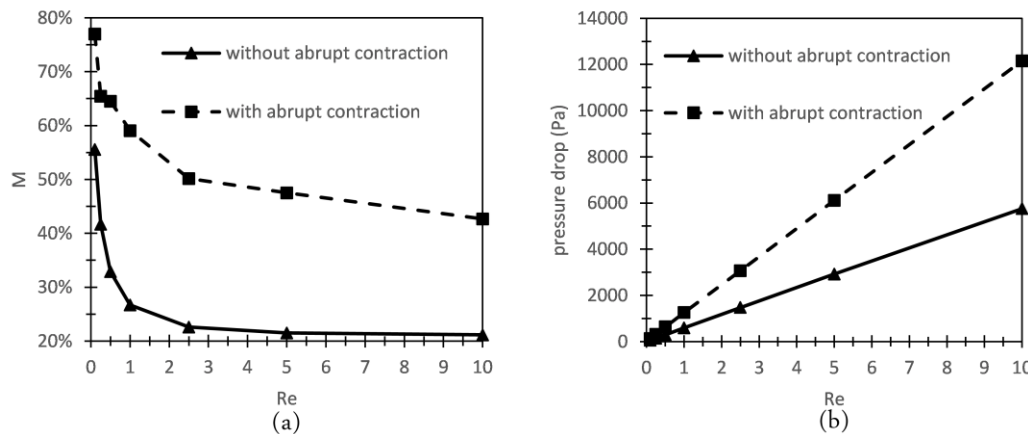


Figure 7. Effect of abrupt contraction on (a) mixing quality and (b) pressure drop

### 3.4 Effect of ribs after contraction

Ribs are usually used to create turbulence in flow direction, which help increasing mixing performance. Figure 10 shows a configuration of a micromixer with 3 ribs of equal height of H  $\mu\text{m}$ , width of 120  $\mu\text{m}$ , and pitch of P  $\mu\text{m}$ . Figure 11 shows the effect of a single rib height and 3 identical ribs with different heights and pitches in mixing quality and pressure drop at Re number of 5. It is found that for a single rib, an increment of only 19% in mixing quality is reached while the increment in pressure drop is 37% at a height of 120  $\mu\text{m}$  (half of the channel width). For greater heights, the pressure drop increment is much higher than the increment in mixing quality. As an example, for 160  $\mu\text{m}$ , the increment in pressure drop is 110% while the increment in mixing quality is 28%. For 3 ribs, a pitch of 240  $\mu\text{m}$  and a height of 120  $\mu\text{m}$  are

more than enough for a satisfactory increment in mixing quality with a reasonable increment in pressure drop. An increment of 36% in mixing quality is achieved with an increment of 131% in pressure drop.

Figure 12 shows the effect of number of ribs with P=240  $\mu\text{m}$  and H=120  $\mu\text{m}$  in mixing quality and pressure drop at Re number of 5. It is found that for a single rib, an increment of only 19% in mixing quality is reached while the increment in pressure drop is 37%. For 3 ribs, an increment of 36% in mixing quality is achieved with an increment of 131% in pressure drop. With more than 3 ribs, pressure drop is found to increase almost in a linear behavior, while the mixing quality slightly increases. So, a 3 ribs system is selected for further investigations

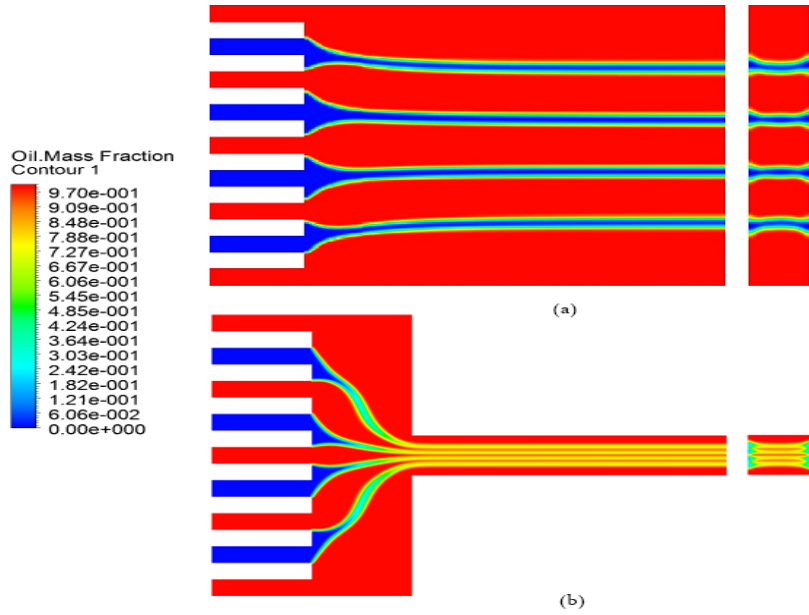


Figure 8. Filled contours of oil mass fraction at a mid-plane of the micromixer parallel to the stream lines and at exit plan for (a) micromixer without contraction, and (b) micromixer with abrupt contraction, both at  $Re = 5$ .

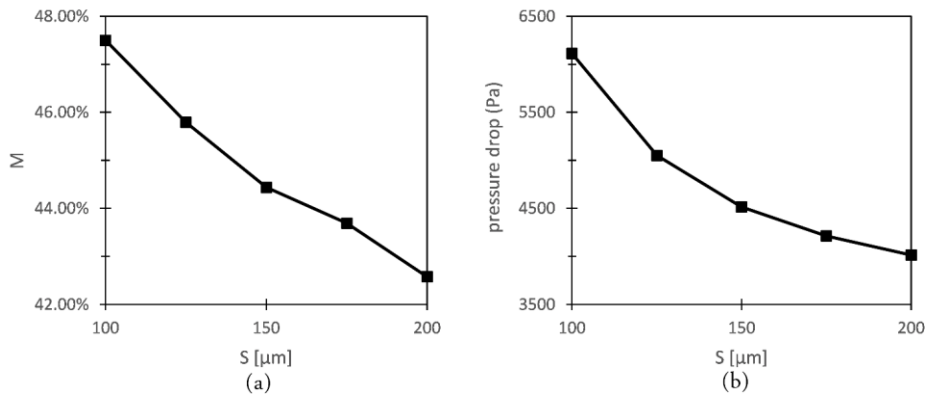


Figure 9. Effect of spacing between obstructions ( $S$ ) on (a) mixing quality and (b) pressure drop, at  $Re = 5$  and contraction angle  $\theta = 90^\circ$

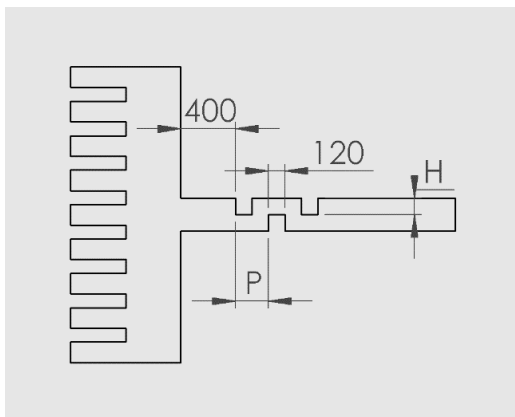


Figure 10. Micromixer with ribs. (Dimensions in micron)

### 3.5 Testing developed micromixer

The developed micromixer configuration shown in **Error! Reference source not found.** was numerically tested using different fluids mixture. Firstly, the presented low diffusivity oil-methanol mixture was used in simulation at  $Re$  numbers ranging from 0.1 to 60. Results are plotted in **Error! Reference source not found.**. Then, the mixture of oil and ethanol given by Santana et al. [8] was used at 3 different values of  $Re$  number as shown in Table 3 to investigate micromixer effectiveness for different flow regimes. Finally, the mixture of water and ethanol given by Alam and Kim [19] was used at 3 different values of  $Re$  number as shown in Table 4.

The configuration gave a satisfactory mixing quality with a relatively small pressure drops for all cases compared to Santana et al. [8] and Alam and Kim [19]. It was also effective for all flow regimes exhibited during the simulation even at

low Re number, where mixing was mainly due to diffusion.

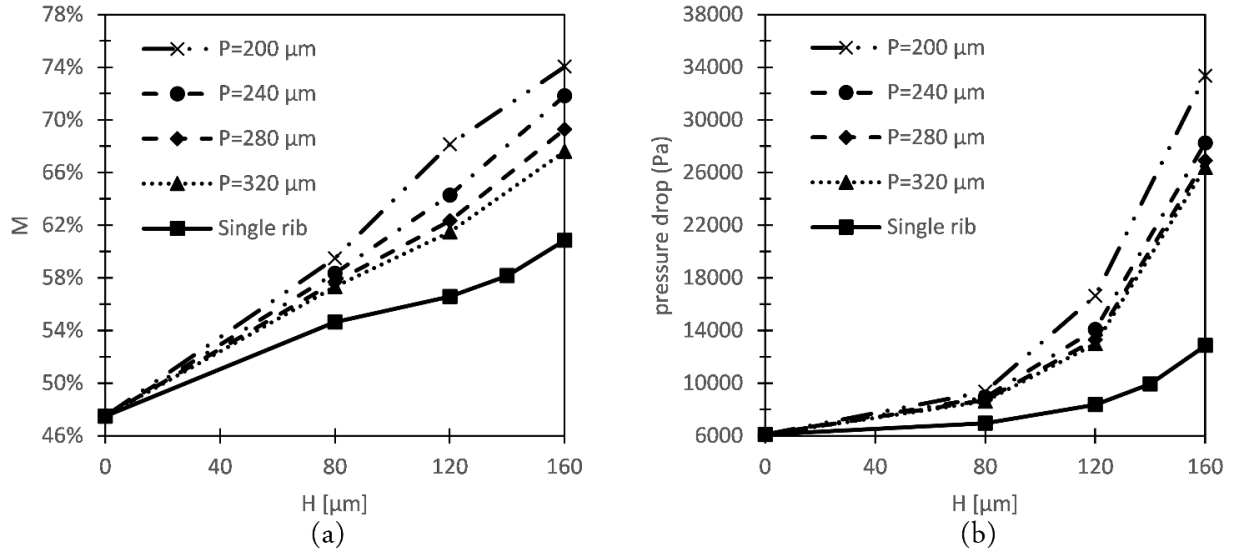


Figure 11. Effect of a single rib height and 3 identical ribs height at different pitches on (a) mixing quality and (b) pressure drop, at Re = 5.

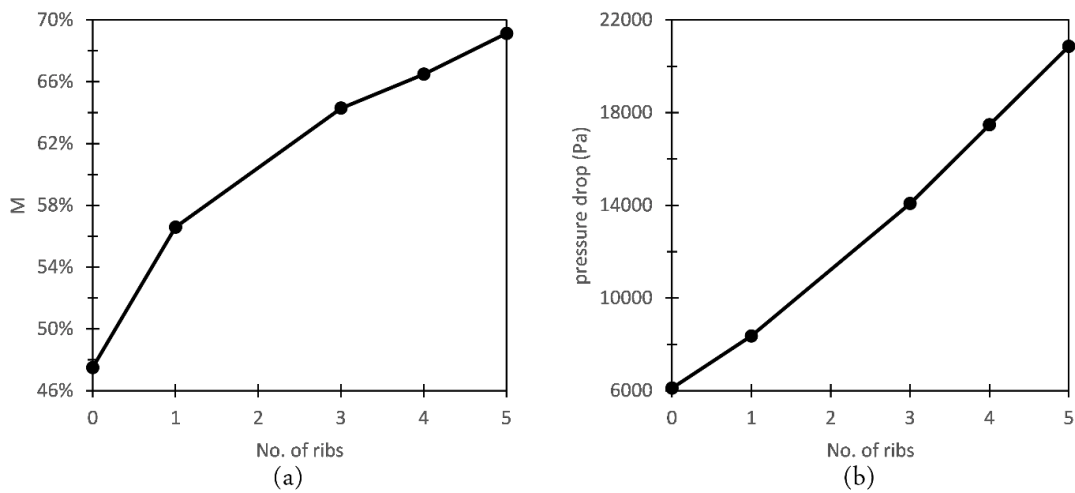


Figure 12. Effect of number of ribs on (a) mixing quality and (b) pressure drop, at Re = 5

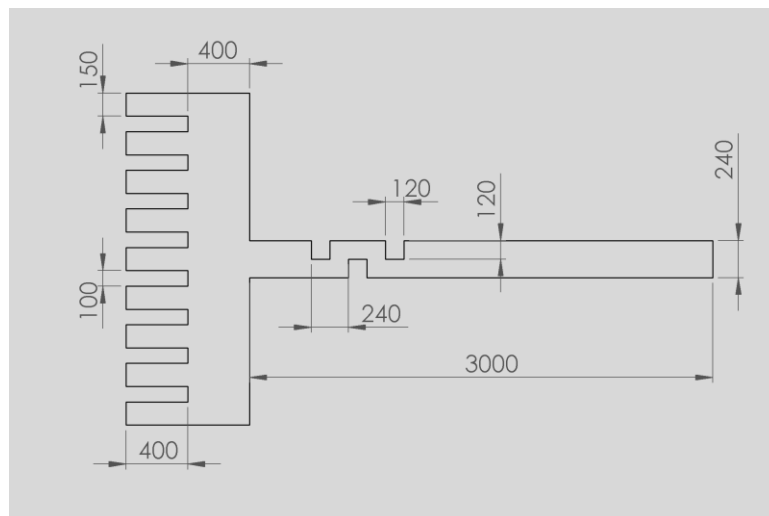


Figure 13. Front view of the tested micromixer. (Dimensions in micron)



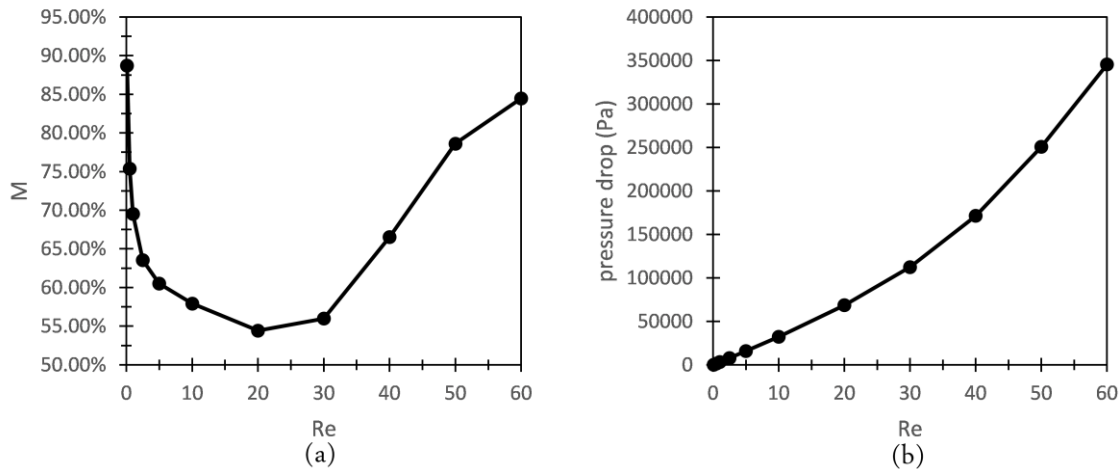


Figure 13 (a) Mixing quality and (b) pressure drop with Re number for the tested micromixer.

TABLE 3  
MIXING QUALITY AND PRESSURE DROP AT DIFFERENT RE NUMBER FOR OIL-ETHANOL MIXTURE

Re	Developed micromixer		Santana et al. [8]	
	Pressure drop (Pa)	M	Pressure drop (Pa)	M
5	60419.6	68.64%	250000	72%
45	755227	63.15%	3600000	80%
90	2443800	85.10%	8650000	97%

TABLE 4  
MIXING QUALITY AND PRESSURE DROP AT DIFFERENT RE NUMBER FOR WATER-ETHANOL MIXTURE

Re	Developed micromixer		Alam and Kim [19]	
	Pressure drop (Pa)	M	Pressure drop (Pa)	M
5	70.9952	77.98%	1190	10%
45	872.27	77.11%	15500	55%
90	2463.22	87.08%	38000	92%

#### IV. CONCLUSION

An efficient uniflow micromixer configuration was introduced, based on multilamination principle. Better mixing quality together with lower pressure drop were observed as compared to other available designs based on multilamination. This is mainly due to the uniflow character of the proposed design.

In addition, abrupt contraction was proposed in order to increase the interface area to volume ratio, which considerably increased mixing quality, at a very small additional pressure drop.

Effect of ribs in enhancing mixing was also numerically investigated. A 3 ribs system with height equal half of the channel width and a pitch between ribs equal the channel width, were more than enough to increase the mixing quality with relatively moderate effect on pressure drop. A developed micromixer configuration was reached and tested with different fluids mixtures.

The enhanced design gave satisfactory mixing quality and

pressure drop even with a low diffusivity mixture such as the mixture of oil and alcohol used in biodiesel production. Enhancement was obtained at very low Reynolds numbers at which a mixture of oil and alcohol should operate.

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