



## Comparison of mixing performances of T, Y and arrow-shaped micromixers using Villermaux–Dushman protocol at low Reynolds number

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**Abstract:** Mixing performance is commonly assessed using the Villermaux–Dushman protocol. Mixing in passive mixers may need to be at very low Reynolds number (Re). This study is aimed at comparing the mixing performances of T, Y and arrow-shaped micromixers using the Villermaux–Dushman protocol at Re < 100. The mixing performance test was run at flow rates of 500 to 1 mL h<sup>-1</sup> with the pressure drop measurement. Based on UV absorbance values and experimental mixing times of mixed fluids from the three micromixers, the order of mixing performance at Re > 26.5 was Y < T < arrow-shaped micromixers. At lower Re values, the order of performance based on the absorbance and the experimental mixing times, became inconsistent. However, the comparison of mixing performance based on predicted mixing time gave consistent order of performance at all Re values. The instability in UV absorbance of mixed reagents in Villermaux–Dushman protocol was noted as the major cause of the reported inconsistency at very low Re. The inadequacy of Villermaux–Dushman protocol at low Re was verified via facile solvent extraction test. A more reliable technique should be used to assess the mixing performance of micromixers at very low Re and on-line UV measurement should be adopted for the Villermaux–Dushman experiment.

**Keywords:** efficiency of mixers; iodide/iodate test experiment; passive mixers; micro-channel; laminar flow regime; confluence shape.

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## INTRODUCTION

Most chemical processes require contacting of different substances to achieve homogeneity, emulsification, dispersion or even chemical reaction as in the case of combination reactions where different molecular species have to contact before conversion. One of the most important methods of enhancing mixing at micro level is using micro scale devices.<sup>1</sup> Dating back to the early nineties, the micro scale devices such as micromixers (or microreactors; when chemical reaction is involved) have attracted great attention in laboratory as well as on industrial or commercial scale.<sup>2,3</sup> The values of Reynolds number ( $Re = \rho u d / \mu$ ; where:  $\rho$  is the density,  $u$  is the flow velocity,  $d$  is the characteristic diameter of the micro channel cross section,  $\mu$  is the dynamic viscosity of fluid; assuming that flow regime laws apply in micro-channels) are usually very low in micro-structured devices due to their small dimensions; the internal cross sectional diameter in the order of  $10^2 \mu\text{m}$ .<sup>4</sup> Therefore, the flows in micro channels fall within the laminar regime.

But, if the flow in micro-channels is in the laminar regime, how is the fluid mixing possible then? Based on the way mixing is achieved, there are two classes of micromixers: the active and the passive micromixers.<sup>5,6</sup> Only the passive micromixer, for which fluid–fluid interfacial contacting (mixing) is effected by the energy of flowing fluids (no external energy source like mechanical stirring is employed, as in the case of active ones), is implied throughout this study. The reduced cross section of micro-channels leads to the increased rate of fluid shearing and based on the micromixer geometry or the configuration (point of confluence, obstruction, bend or twist), the fluid in laminar flow gets deformed *via* convection as it flows past such obstructions, bends, twists or as streams meet at point of confluence and that is the mechanism of mixing in passive micromixers.<sup>6,7</sup> Primarily, there is diffusive mixing at molecular level and this diffusion is enhanced by the relatively reduced diffusion path or cross section of flow.<sup>7</sup> In fact, “Molecular diffusion is the ultimate and finally the only process really able to mix components of a fluid on the molecular scale”.<sup>6</sup>

As a result of their small sized channels, the micromixers have often increased fluid surface area to volume ratio and this makes them very useful in high mass transfer and heat transfer applications.<sup>5</sup> Commenge and Falk noted some other interesting applications of micromixers arising from their compact nature, to include: safety of operation in case of highly exothermic and fast reactions involving hazardous chemicals; new and untapped reaction routes for chemical synthesis (such as direct fluorination), which are very dangerous can be accessed and there is improved efficiency in energy utilization by using passive micromixers.<sup>3</sup> In addition, the small sized devices save materials and time; since just little volume of fluid is required to operate them and reasonably short residence

time is needed. Unlike most chemical process equipment that are bulky, micro-mixers are very portable.

Consequent to the relevance of mixing and the beneficial nature of micro-mixers, the assessment of the mixing performance of micromixers is a very important step for their design (making good choices). While it may be so easy to assess the performance of some micro-devices by the comparison of some standard parameters like heat transfer coefficient for heat exchangers, the case of micromixers is quite difficult.<sup>2</sup> A number of techniques have been utilized to assess the mixing performance and they are included in order of increasing preference: the visualization of dyes as their colour fades away, due to the dilution by mixing or as their colour changes, in case of pH sensitive dyes, the visualization of reactions that progress with change in colour and most recently, the monitoring of competitive test reaction schemes.<sup>1–3,8</sup>

Amongst the competitive test reaction schemes used for mixing performance assessment, the Villermaux–Dushman reaction scheme which consists of two parallel reactions: neutralization and redox reactions in competition for proton from acid, is well known and most used.<sup>3,7</sup> The neutralization reaction given by Eq. (1) is instantaneous (extremely fast) while the redox reaction (Eq. (2)) is just fast.<sup>5,6</sup> All species in Eqs. (1)–(3) are aqueous:



A strong acid is used to generate the protons in one reactant (acidic) solution while the second (buffer) solution for the mixing contains:  $\text{IO}_3^-$ ,  $\text{I}^-$  and  $\text{H}_2\text{BO}_3^-$ . Falk and Commenge<sup>6</sup> require that only a critical stoichiometric amount of acid is needed to neutralize the borate ions according to Eq. (1) must be used for the test, while Fournier *et al.*<sup>9</sup> state that the acid should just be the limiting reagent. The concept of the Villermaux–Dushman protocol is briefed as follows.<sup>3,6</sup> When the mixing is perfect, the stoichiometrically fitted instantaneous reaction (Eq. (1)) completely consumes the protons, leaving no room for the slower Eq. (2) to proceed. However, when mixing is poor, there is segregation (heterogeneous fluid behaviour) of the two reactant solutions into acidic and buffer segregates (local sites), making the amount of protons in every acidic segregate to be excessively high to just complete the neutralization of the borate ions in the immediate surrounding (boundary/interphase). The excess protons enable the occurrence of the redox reaction to form iodine. The extent of segregation/size of the segregates depends on the mixing efficiency/performance and that determines the amount of iodine to be produced. Eq. (3), which utilizes the iodine, is very useful for the quantitative analysis because the amount of  $\text{I}_3^-$  produced can be easily measured using UV spectrophotometer at a wavelength of 353 nm. Hence, the relatively

low absorbance values of mixed products indicate relatively high mixing performance and *vice versa*. Ehrfeld *et al.*<sup>2</sup> share similar idea but they utilized acetate in place of borate species in Eq. (1). Some works<sup>2,10</sup> are reported with product absorbance measured at 352 nm, noting that the UV spectrum has a peak at that wavelength, rather than at 353 nm; however, the values are quite close. Commenge and Falk<sup>3</sup> further noted that the test gives optimum (most sensitive) result when the rates are such that the neutralization occurs in zero seconds (quasi-instantaneous) and the redox reaction occurs at roughly the same time as the mixing time. Villermieux–Dushman protocol is also known as the iodide/iodate test method.

Following the Villermieux–Dushman protocol, the performance of a mixer at specific conditions should be assessed based on the observed absorbance value of the mixed products (the higher the absorbance, the poorer the performance and *vice versa*). Unfortunately, several authors<sup>3,6,11</sup> recommend that the mixing performance criteria should not be concentration dependent. Mixing time ( $t_m$ ), according to Commenge and Falk<sup>3</sup> which ought to be adopted by Asano *et al.*<sup>11</sup> is given in Eq. (4). Obviously, the model is concentration dependent, since it includes absorbance and initial concentrations as variables:

$$t_m = 0.33 OD c(H^+)_0^{-4.55} c(KI)_0^{-1.5} c(KIO_3)_0^{5.8} c(NaOH)_0^{-2} c(H_3BO_3)_0^{-2} \quad (4)$$

where  $OD$  is optical density which is equivalent to the absorbance of mixed reagents/sample for cuvette of optical density of 1 cm;  $c(i)_0$  is the initial molar concentration of chemical species  $i$  (in the stock reagent solution).

However, satisfactory attempts to express  $t_m$  as a concentration free parameter (in terms of kinematic viscosity and specific power dissipation rate) are reported somewhere.<sup>9</sup> By the analysis of literature results, Commenge and Falk obtained a relationship between the theoretical mixing time ( $t_{mp}$ ) in s and the specific power dissipation ( $\varepsilon$ ) in  $\text{W kg}^{-1}$  as given in Eq. (5). They recommend that the equation should be used when pressure drop data due, to the flow through micromixer, are available. Eq. (5) was used to predict the mixing time in this work. The specific power dissipation or specific energy dissipation rate is expressed in terms of volumetric flow rate ( $Q$ ), pressure drop ( $\Delta p$ ), density ( $\rho$ ) and control volume of the micromixer ( $V$ ) as shown in Eq. (6):<sup>3</sup>

$$t_{mp} = 0.15 \varepsilon^{-0.45} \quad (5)$$

$$\varepsilon = Q \Delta p / (\rho V) \quad (6)$$

Due to the variations in channel dimensions and fluid flow conditions for different experimental set-ups, the works on micromixer performance are better reported in terms of  $Re$  values rather than volumetric flow rates or velocity. Also, it may not be appropriate to compare the values of performance data from different works because several authors report different reagent concentrations and reagents' flow rate ratios.<sup>11</sup> In the evaluation of mixing performance of T, Y and oriented Y micromixers with the spatially arranged outlet channel using Viller-

maux–Dushman protocol, at minimum flow rate of  $10 \text{ mL min}^{-1}$ , at average fluid velocity of  $0.331 \text{ ms}^{-1}$  (minimum  $\text{Re} = 265$ ), the reduction in segregation index (the improvement in mixing performance) with the increasing flow rate is reported.<sup>5</sup> From this result, by comparison of segregation index values at common flow rates, the oriented Y micromixers (which has arrow-shaped confluence) gave the best mixing performance followed by T and then Y-mixer.<sup>5</sup> Also, the similar result on comparison of mixer performances and the effect of flow rate on mixing performance was earlier reported using an online spectroscopic analysis system (UVmini1240, Shimadzu), still at  $\text{Re} > 265$ .<sup>7</sup> The information on mixing performance at very low  $\text{Re}$  values is scarce and such information may be very valuable in certain applications like drug delivery and in achieving mixing down to cellular level in biological systems.

In consolidation with the works of other researchers, this study aims to compare the mixing performances of three different configurations of micromixers (T, Y and arrow-mixers) using the Villermaux–Dushman test reaction at low  $\text{Re}$  values. To achieve this aim, the Villermaux–Dushman protocol was carried out for the micromixers with pressure measurements and then the absorbance values of mixed products from each of the micromixers, the experimental and predicted mixing times, were compared at various  $\text{Re}$  values.

## MATERIALS AND METHODS

### Materials

T, Y and arrow-shaped micromixers made by 3D printing, each with the uniform internal diameter of 0.5 mm and length of 30 mm after confluence, were used in this study. Two syringes each of capacity 60 mL and 26.72 mm diameter were used to feed the storage tank. Other accessories such as G28 screws, Teflon® and small, micro-sized, pipes were used for connection.

The consumable chemicals, which include: sodium hydroxide pellets (product of Appli-Chem GmbH, Germany), 99 % pure potassium iodide (product of Chem-Lab NV, Belgium), boric acid (product of MP Biomedicals, LLC, France), potassium iodate (Rankem™, RFCL Limited, India), methanol (>99.9 %, product of Fisher Scientific, UK) and hydrochloric acid of 37 % purity and specific gravity of 1.18 (product of Fisher Scientific, UK) were all of analytical grade and accessed from Chemical and Petrochemical Engineering Laboratory, E-JUST, Egypt. Hедеya cooking oil was purchased from Fathalla-Gomla Market, Borg El-Arab, Alexandria, Egypt.

Dual-syringe infusion pump (model: 115 VAC; Cole-Parmer™, USA), KOBOLD hand-held pressure measuring device for differential pressure for 2 external sensors (HND-P215; with accuracy of 0.0005 psi), 2 KOBOLD pressure sensors (HND-PS19) and Hitachi UV spectrophotometer (U-3900; for absorbance measurement, with 0.0005 accuracy), equipped with sample cell/cuvette of optical path length = 1cm, were accessed from different laboratories in E-JUST.

### Methods

*Villermaux–Dushman experiment with pressure measurement.* One liter each of the acid (0.03 M aqueous HCl) and buffer (0.09 M  $\text{H}_3\text{BO}_3$ <sub>(aq)</sub>, 0.09 M  $\text{NaOH}$ <sub>(aq)</sub>, 0.006 M  $\text{KIO}_3$ <sub>(aq)</sub>) and

0.032 M  $\text{KI}_{(\text{aq})}$  solutions for Villermaux–Dushman reaction were prepared and labelled solution 1 and solution 2, respectively.<sup>11,12</sup> Two 60 mL syringes were filled with solutions 1 and 2, labelled and clamped onto the dual syringe pump. The Y-mixer was connected with two pressure probes (sensors) at the mixer inlet and outlet pipes. The experimental set-up is as shown in Fig. 1a and b; it shows details of the micro-channels of the Y, T and the arrow-shaped micromixers displaying the confluence shapes and the angular deviation of fluid streams at point of confluence (with flow directions).

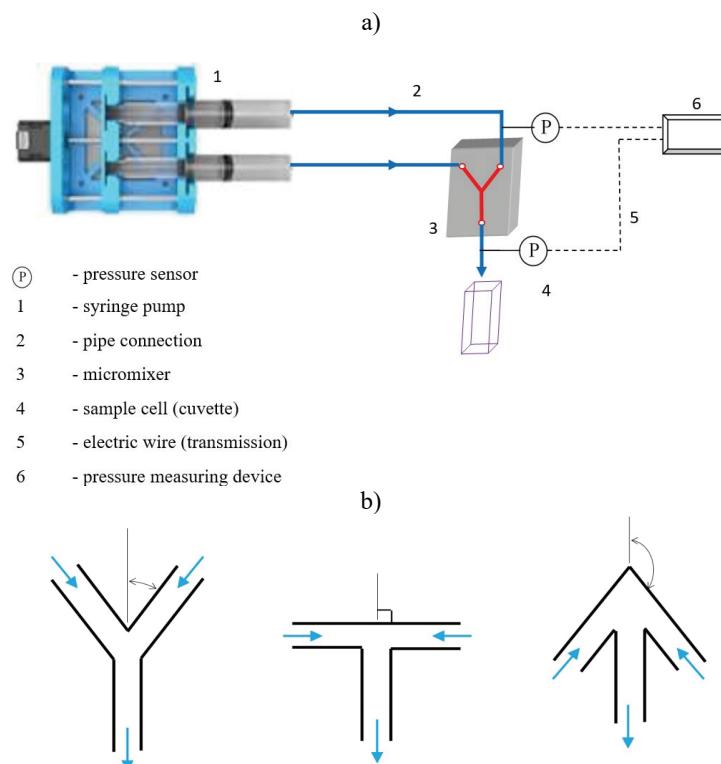


Fig. 1. a) Schematic diagram of the experimental set-up; b) illustrative details of the micro-channels of the mixers showing the confluence shapes and angular deviations (60, 90 and 120°) of fluid streams at point of confluence.

The syringe pump was switched on and set to: volume, diameter and flow rate of 60 mL, 26.72 mm and 500 mL hr<sup>-1</sup>, respectively. Afterwards, the pump was run. The pressure readings were taken after the flow had attained the steady state (as indicated by the constant pressure readings), while 2 mL of mixed sample was collected after discarding the first 20 mL that flowed out in order to ensure steady concentration. UV absorbance of the collected sample was measured as fast as possible within the set wavelength range of 320 to 370 nm, and recorded. Next, the flow rate was successively changed to 400, 300, 250, 200, 150, 100, 70, 50, 30, 15, 7 and 1 mL hr<sup>-1</sup>, while repeating the same mixing performance characterization procedure each time. Afterwards, the Y-mixer was replaced with the T-mixer and then with the

arrow-shaped mixer while repeating the same procedure. Syringes 1 and 2 were refilled with solutions 1 and 2, respectively, each time their content reduced below 25 mL.

All through the experiment, the connected micromixer was fixed at a horizontal position with the pressure probes maintained at the same position to avoid changes in pressure head (variation in hydraulic pressure with height). The upper limit of flow rate used in this study ( $500 \text{ mL h}^{-1}$ ) was fixed by the maximum pressure limit (5.8 psi) of the KOBOLD (HND-PS19) pressure sensors, while the lower limit ( $1 \text{ mL h}^{-1}$ ) was fixed by time consideration.

Finally, the mixing performance of the micromixers was assessed based on the UV absorbance, the predicted mixing time ( $t_{mp}$ ) and the experimental mixing time ( $t_m$ ). The density and viscosity of the dilute solutions used in the experiment were assumed to be the same as that of water (approximately  $1 \text{ g cm}^{-3}$  and  $1 \text{ cP}$ , respectively). With these solution fluid parameters, together with the micro-channel dimension, it was possible to express flows in dimensionless form as  $\text{Re}$ .

## RESULTS AND DISCUSSION

### *Mixing performance characterization based on UV absorbance values*

The acid and buffer concentrations used resulted in high absorbance values at flow rates below  $150 \text{ mL h}^{-1}$  ( $\text{Re} = 26.5$ ). According to Falk and Commenge,<sup>3,6</sup> the absorbance-concentration relationship, otherwise known as the Beer-Lambert law, is valid for the tri-iodide ion at the absorbance range below 2.5. The dilution of the mixed samples of high absorbance is discouraged;<sup>3</sup> a failed attempt to characterize performance by dilution of mixed samples with high UV absorbance is reported in literature.<sup>12</sup> Consequently, the concentrations of solutions 1 and 2 were each halved by doubling their volumes with distilled water before proceeding with the experimental runs for flow rates below  $150 \text{ mL h}^{-1}$ . As a result of this modification, the result (Fig. 2) is presented in two parts.

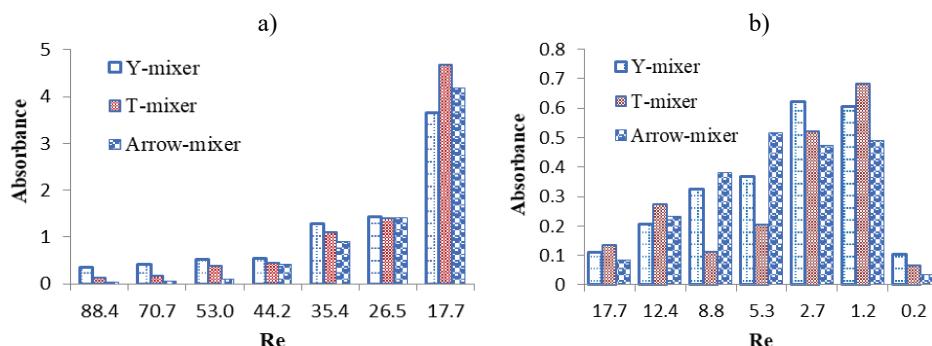


Fig. 2. Effect of Re values on absorbance: at stated reagent concentration for higher flows (a) and with halved reagent concentration for lower flow rates (b).

Fig. 2a and b show the effect of  $\text{Re}$  on absorbance at the stated reagent concentration for higher flows and halved concentration for lower flows, respectively. Fig. 2a clearly shows that the UV absorbance increased progressively for

all the micromixers with the decreasing Re. By comparison, the arrow-mixer gave the least absorbance (best mixing performance), while the Y-mixer gave the highest absorbance (least mixing performance) for flows above the Re value of 26.5. At Re value of 26.5, the three mixers measured up in absorbance, while it is clear from Fig. 2b that there is no consistent comparative trend in absorbance for the micromixers at  $Re < 26.5$ .

In quest for possible reasons for the inconsistency in the absorbance trend at  $Re < 26.5$ , the UV absorbance of mixed samples were measured with respect to time (age after sampling). It was found that the absorbance of mixed samples for all Re values, were not stable; the values reduced with samples' age. Ehrfeld *et al.*<sup>2</sup> noted this instability in absorbance, although they report an increase in the absorbance, using sodium acetate in place of NaOH and boric acid. The observed instability could be due to the reversible nature of most of the reactions in the Villermaux–Dushman reaction scheme. As a result of this, the time lag between samples collection and measurement was kept to the minimum, for greater accuracy. Also, the spectrophotometer was set to cover short range of wavelength (320 to 370 nm) to reduce response time in measurement. The observed sharp drop in absorbance at  $Re = 0.2$ , for all mixers as shown in Fig. 2b, could be due to the long time spent in collecting samples at such low flow rate – allowing such a long time lag for mixed samples absorbance to depreciate. Substantial reasons for the absorbance instability with samples' age cannot be given because the kinetics of Villermaux–Dushman reaction is not well understood.<sup>13</sup>

Again, the mixed samples were quickly collected and analyzed for the UV absorbance at very close time intervals from the on-set of mixing till exhaustion of syringe contents, at different Re values. The result showed that the steady absorbance value (with noise of  $\pm 0.01$  at  $Re < 17.7$  and  $\pm 0.003$  at  $Re \geq 17.7$ ) was attained after collecting at most 5 ml of mixed solution. The observed noise at steady state (at  $Re < 17.7$ ) is quite high, but too small to account for the observed inconsistency, because the observed disorderliness in absorbance (for Fig. 2b) is greater than 0.01. Therefore, the noise could have contributed slightly to the inconsistency. This test justified the assumption of the steady state concentration during sampling (discarding the first 20 mL of mixed sample).

#### *Mixing performance characterization based on mixing times*

The high mixing performance is characterized by the low mixing time. Figs. 3 and 4 show the effect of changing Re on the experimental mixing time ( $t_m$ ) and on the predicted mixing time ( $t_{mp}$ ), respectively. By comparison, as it can be seen from the figures, the values of the experimental mixing time at a given Re are more distinct than the predicted ones which are clustered together. Again, the range of values of  $t_{mp}$  is lower than the experimental mixing time ( $t_m$ ). Fig. 3a and b were derived from the absorbance data presented in Fig. 2a and b, and the

same discussion applies. Although Fig. 4 is not quite clear due to the clustered points; for all  $Re$ ,  $t_{mp}$  was the lowest for the arrow mixer and the highest for the Y-mixer.  $t_{mp}$  increased for all mixers as  $Re$  value was decreased. The increment in  $t_{mp}$  was gradual, but steeped up at  $Re < 12.4$ . The similar step-up with decreasing  $Re$  is also noticeable in Fig. 3a; meaning that the sensitivity of mixing performance to the changes in  $Re$  is higher at low  $Re$ , than at high  $Re$ . The maximum value of  $t_{mp}$  was achieved at the least flow velocity ( $Re = 0.2$ ), as it can be seen from Fig. 4. While from Fig. 3b a very low  $t_m$  (high mixing performance) was obtained, which appears unreasonable because at very low  $Re$ , mixing is wholly dominated by the molecular diffusion without convective (vortex) enhancement. However, numerical simulations have shown that the reduction of  $Re$  at certain range could lead to the increase in mixing quality.<sup>14–17</sup> But that does not justify the inconsistency in order of performance encountered here, which is not different from an earlier report.<sup>12</sup> The predicted mixing time of Fig. 4 does not agree with the theoretical results,<sup>14–17</sup> because the predicted model (Eq. 5)<sup>3</sup> is independent of chemical species in the mixed stream.

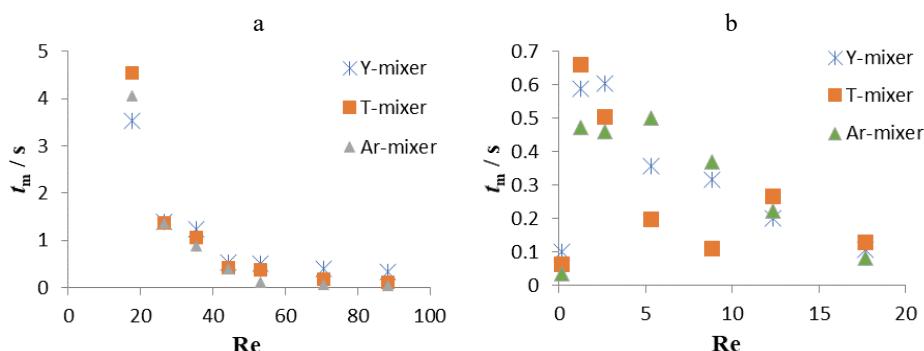


Fig. 3. Effect of  $Re$  values on experimental mixing time: with normal reagent concentration (a) and with adjusted reagent concentration (b).

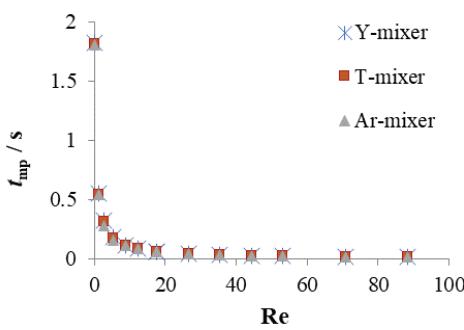
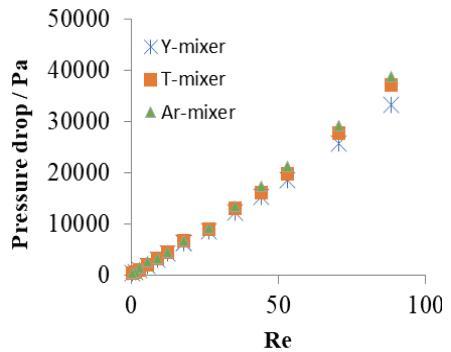
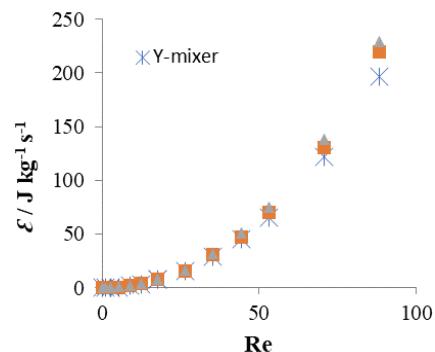


Fig. 4. Effect of  $Re$  values on predicted mixing time.

Effect of the confluence shape on mixing performance can therefore be explained as follows. At a given  $Re > 30$ , there is improvement in performance, since the confluence shape encourages more fluid-fluid and/or fluid–wall impact when two fluid streams meet, and *vice versa*. Referring back to Fig. 1b, two factors are of importance in explaining the order of performance: the impact on collision and the simultaneous angular deviation in fluid flow direction. When the inlet fluid streams approach each other from opposite directions, as in the T-shaped confluence, the fluid-fluid collision impact is “head on” and maximum, compared to the lower impact obtainable when the streams approach each other at an angle (as in the Y and arrow-shaped confluentes). The effect of bend on fluid mixing along channels<sup>18</sup> also plays great role in mixing at confluence points, because with greater deviation at confluence, the enhanced mixing is achievable. The angular deviation in flow direction for T-shaped confluence is just 90° while Y-shape offers the least deviation. The arrow-shaped confluence caused the greatest agitation during the collision of streams due to its maximum angular deviation (>90°), coupled with its slight collision impact. T was better than Y, because it offers the greatest collision impact and better deviation than Y-mixer. In all cases, the mixing is enhanced with increasing  $Re$  value in the range  $> 30$ .

Fig. 5. Effect of  $Re$  on pressure drop.Fig. 6. Effect of  $Re$  on energy dissipation rate.

*Variation of pressure drop and specific energy dissipation rate with Re*

The modification of concentration stated under “Mixing performance characterization based on UV absorbance values” section did not affect the pressure drop and therefore Figs. 4 to 6 are not split into two. The effect of inlet Re on pressure drop ( $\Delta p$ ) as a result of flow through each micromixer is shown in Fig. 5. From the figure,  $\Delta p$  increased somewhat linearly as Re increased. The variation in  $\Delta p$  among the three micromixers becomes wider (diverges) as one moves from low Re to higher Re. The similar trend is reported by Hossain *et al.*,<sup>8</sup> Jafari *et al.*<sup>19</sup> and Kockmann *et al.*<sup>15</sup> To understand this divergence, we consider the possible expressions for  $\Delta p$ , or pressure head, as a function of Re or velocity, hence the Hagen–Poiseuille equation and the Darcy’s equation. But, the flows in micromixers are not ideally laminar to be described as Hagen–Poiseuille flow. Based on Darcy’s relationship,  $\Delta p$  is proportional to  $f u^2$ ; where  $f$  is a dimensionless parameter (a function of flow velocity and other factors), commonly known as the friction factor and  $u$  is the average velocity of flow.<sup>20</sup>

When the fluid streams (from two identical branches of micromixer) collide/meet, mixing takes place at the expense of energy which is felt as a drop of pressure. The effect of Re on energy dissipation rate ( $\varepsilon$ ) is shown in Fig. 6. From the figure, the variation of  $\varepsilon$  with respect to Re is very similar to the observed trend for  $\Delta p$ , but is more of a curve than straight line, as a result of the extra velocity factor, as related in Eq. (6). It is clear that higher energy is spent at higher Re and the consequence of it is the improvement in mixing performance. This is in agreement with the literature.<sup>6,19</sup> The order of energy dissipation rate is Ar > T > Y-mixers.

*Verification of the inadequacy of Villermaux–Dushman protocol at low Re values*

The simple interphase mass transfer between two immiscible liquids (water and crude biodiesel) was used to verify the observed performance inconsistency in this study.<sup>21</sup> Briefly, the hydrophobic (crude biodiesel) phase resulting from ultrasonic assisted-methanolysis of fresh cooking Hedeya oil in presence of 1 % KOH as homogeneous catalyst for 40 min, with methanol to oil mole ratio of 6:1, at 60 °C; was contacted with the alkaline water (0.016 M KOH) in the same experimental set-up of Fig. 1, at flow rates of 70, 50, 20, 7 and 1 mL h<sup>-1</sup>, each for T, Y and arrow-shaped micromixers, respectively. After micromixing, 5 mL of the resulting aqueous phase was titrated over 0.03 M HCl, using phenolphthalein as an indicator. We are currently working to fully develop this performance characterization technique and the details would be published soon.

Titrimetry revealed a progressive increase in the alkalinity of biodiesel wash water, for all the micromixers, as the flow rate was increased. Table I shows the titrimetric result of aqueous extract phase. Also, at every flow rate, the order of mixing performance was arrow > T > Y mixer.

TABLE I. Titrimetric result of aqueous extract phase

Flow rate, mL h <sup>-1</sup>	Volume of HCl used, mL		
	Arrow-mixer	T-mixer	Y-mixer
70	3.65	3.50	3.15
50	3.35	3.20	3.00
20	3.10	2.95	2.85
7	2.95	2.90	2.85
1	2.85	2.85	2.80

## CONCLUSION

The mixing performances of T, Y and arrow-shaped micromixers have been characterized by the Villermaux–Dushman protocol at low Re. Based on UV absorbance, experimental and predicted mixing times; comparison of mixing performance of the micromixers was made at various Re. Facile solvent extraction test was performed for the result verification. From the results of this study, it can be concluded that the arrow-shaped confluence offers the best mixing performance, while the Y-shape offers the least of the three confluence shapes at  $Re > 30$ . The Villermaux–Dushman protocol is not adequate at lower Re. Increase in Re within the range  $30 < Re < 100$ , leads to the improvement in mixing performance. Mixing performance is more sensitive to changes in Re at low Re values. Improvement in mixing performance, due to the shape of confluence or to the increase in Re value occurs at the cost of fluid energy per unit time (power).

A more reliable mixing performance characterization method has been used to verify the inadequacy of the Villermaux–Dushman protocol at very low Re. Finally, in order to reduce the effect of time lag, on-line (*in-situ*) absorbance measurement should be adopted.

## NOMENCLATURE

Re	Reynolds number
Ar	Arrow (micromixer)
$\rho$	density of fluid, kg m <sup>-3</sup>
$\mu$	dynamic viscosity of fluid, kg m <sup>-1</sup> s <sup>-1</sup>
$u$	flow velocity of fluid stream, m s <sup>-1</sup>
$d$	characteristic diameter of flow channel, m
UV	ultraviolet
OD	optical density or mixed sample absorbance
$c_{i,0}$	initial molar concentration of chemical species $i$ , mol L <sup>-1</sup>
$\varepsilon$	specific power dissipation or specific rate of energy dissipation, W kg <sup>-1</sup> or J kg <sup>-1</sup> s <sup>-1</sup>
$Q$	volumetric flow rate, mL h <sup>-1</sup>
$\Delta p$	pressure drop, Pa
$V$	control volume for mixing, m <sup>3</sup>
$t_m$	experimental mixing time, s
$t_{mp}$	predicted mixing time, s
$f$	dimensionless friction factor in Darcy's equation

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## ИЗВОД

ПОРЕЂЕЊЕ ПЕРФОРМАНСИ МЕШАЊА ЗА Т, У И МИКРОМИКСЕРЕ ОБЛИКА  
СТРЕЛИЦЕ КОРИШЋЕЊЕМ VILLERMAUX–DUSHMAN ПРОТОКОЛА ПРИ НИСКИМ  
ВРЕДНОСТИМА Re БРОЈЕВА

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Перформасе мешања се уобичајено процењују коришћењем Villermaux–Dushman протокола. Мешање у пасивним миксерима може бити пожељно при веоме ниским вредностима Ренолдсовог (Re) броја. Циљ овог истраживања је поређење перформанси мешања за Т, У и микромиксер облика стрелице, применом Villermaux–Dushman протокола, за  $Re < 100$ . Тестови за испитивање перформанси мешања су вршени приprotoцима од 200 до 1 ml/h, при чему су вршена мерења пада притиска. На основу вредности UV абсорбација и експерименталних времена мешања за 3 микромиксера, добијен је следећи редослед перформанси мешања за  $Re > 26,5$ :  $Y < T <$  микромиксер облика стрелице. При нижим вредностима Re броја, редослед перформанси мешања заснован на абсорбанси и експерименталним вредностима времена мешања, је постајао неконзистентан. Међутим, поређење перформанси мешања заснованих на предвиђеном времену мешања је дало конзистентан редослед за све вредности Re броја. Примећена је нестабилност у мерењима UV абсорбације помешаних реагенаса у Villermaux–Dushman протоколу, која представља главни разлог неконзистензог понашања при веома ниским Re бројевима. Неадекватност Виль Villermaux–Dushman протокола при ниским Re бројевима је потврђена помоћу једноставног теста екстракцијом растворачем. Да би се испитале перформансе мешања при веома ниским Re бројевима требало би користити поузданiju методу, као и онлајн UV мерење које би требало усвојити за Villermaux–Dushman експеримент.

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## REFERENCES

1. J. Aubin, M. Ferrando, V. Jiricny, *Chem. Eng. Sci.* **65** (2010) 2065 (<https://doi.org/10.1016/j.ces.2009.12.001>)
2. W. Ehrfeld, K. Golbig, V. Hessel, H. Löwe, T. Richter, *Ind. Eng. Chem. Res.* **38** (1999) 1075 (<https://doi.org/10.1021/ie980128d>)
3. J. M. Commenge, L. Falk, *Chem. Eng. Process. Process Intensif.* **50** (2011) 979 (<https://doi.org/10.1016/j.cep.2011.06.006>)
4. P. Garstecki, M. J. Fuerstman, H. A. Stone, G. M. Whitesides, *Lab Chip.* **6** (2006) 437 (<https://doi.org/10.1039/b510841a>)
5. Ma. Rahimi, N. Azimi, M. A. Parsamogadam, A. Rahimi, M. M. Masahy, *Microsyst. Technol.* **23** (2017) 3117 (<https://doi.org/10.1007/s00542-016-3118-6>)

6. L. Falk, J. M. Commenge, *Chem. Eng. Sci.* **65** (2010) 405 (<https://doi.org/10.1016/j.ces.2009.05.045>)
7. M. Rahimi, P. Valeh-e-Sheyda, M. A. Parsamoghadam, N. Azimi, H. Adibi, *Chem. Eng. Process. Process Intensif.* **85** (2014) 178 (<https://doi.org/10.1016/j.cep.2014.09.001>)
8. S. Hossain, I. Lee, S. M. Kim, K.-Y. Y. Kim, *Chem. Eng. J.* **327** (2017) 268 (<https://doi.org/10.1016/j.cej.2017.06.106>)
9. M. C. Fournier, L. Falk, J. Villermaux, *Chem. Eng. Sci.* **51** (1996) 5187 ([https://doi.org/10.1016/S0009-2509\(96\)00340-5](https://doi.org/10.1016/S0009-2509(96)00340-5))
10. K. Kunowa, S. Schmidt-Lehr, W. Pauer, H. Moritz, C. Schwede, *Macromol. Symp.* **259** (2007) 32 (<https://doi.org/10.1002/masy.200751305>)
11. S. Asano, S. Yamada, T. Maki, Y. Muranaka, K. Mae, *React. Chem. Eng.* **2** (2017) 830 (<https://doi.org/10.1039/c7re00051k>)
12. O. Okwundu, M. Fuseini, *Mixing performance of micromixers by Villermaux–Dushman reaction protocol at low Reynolds Number*, Technical Repor” (2018) (<https://doi.org/10.13140/RG.2.2.33028.83847/1>)
13. J. R. Bourne, *Chem. Eng. J.* **140** (2008) 638 (<https://doi.org/10.1016/j.cej.2008.01.031>)
14. M. Engler, N. Kockmann, T. Kiefer, P. Woias, *Chem. Eng. J.* **101** (2004) 315 (<https://doi.org/10.1016/j.cej.2003.10.017>)
15. N. Kockmann, T. Kiefer, M. Engler, P. Woias, *Sensors Actuators, B* **117** (2006) 495 (<https://doi.org/10.1016/j.snb.2006.01.004>)
16. N. Kockmann, C. Foll, P. Woias, *Microfluid. BioMEMS, Med. Microsystems* **4982** (<https://doi.org/10.1111/12.478157>)
17. A. Soleymani, E. Kolehmainen, I. Turunen, *Chem. Eng. J.* **135** (2008) S219 (<https://doi.org/10.1016/j.cej.2007.07.048>)
18. N. Aoki, R. Umei, A. Yoshida, K. Mae, *Chem. Eng. J.* **167** (2011) 643 (<https://doi.org/10.1016/j.cej.2010.08.084>)
19. O. Jafari, M. Rahimi, F. H. Kakavandi, *Chem. Eng. Process. Process Intensif.* **101** (2016) 33 (<https://doi.org/10.1016/j.cep.2015.12.013>)
20. J. R. Welty, C. E. Wicks, R. E. Wilson, G. L. Rorrer, *Fundamentals of Momentum, Heat and Mass Transfer*, 5<sup>th</sup> ed., John Wiley & Sons Inc., New York,, 2007
21. Y. Zhao, G. Chen, Q. Yuan, *AICHE J.* **53** (2007) 3042–3053 (<https://doi.org/10.1002/aic.11333>).