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Liquid–liquid mixing enhancement rules by microbubbles in three typical micro-mixers

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HIGHLIGHTS

- The mixing performance in microchannels under extreme operating conditions is greatly intensified.
- Advantages and disadvantages of three representative types of microchannels in terms of mixing performance with gas enhancement are compared under different conditions.
- The fundamental mechanism of the mixing intensification is figured out.
- Mathematical models are established about liquid–liquid mixing with and without gas phase.

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ABSTRACT

Micro-mixers are good platforms to realize liquid–liquid mixing, but they still need intensification when treating extreme flow conditions, such as relatively higher fluid viscosity than water ($\mu > 3$ mPa s) or high flow rate ratio ($R > 10$). In this work, the quantitative mixing enhancement rules with inert gas phase as enhancing medium in three typical micro-mixers, T-junction microchannel, cross-junction microchannel as well as co-flowing microchannel, are experimentally studied using "Villermaux–Dushman" parallel competitive reaction. The results show that the gas makes *Xs* reduced by 2–5 times in the working systems with various viscosities (0.86 mPa s $\leq \mu \leq$ 11.46 mPa s) and operated in a wide range of flow rate ratio ($1 \leq R \leq 25$). The mixing enhancement mechanism is the two-phase segmented flow and we find only 0.05–0.1 gas–liquid phase flow rate ratio (R_{gas}) is enough for the gas phase to develop its enhancing effect. Based on the characteristics of segmented liquid flow, a mathematical model is established to predict the variation of *Xs* in three micro-mixers.

1. Introduction

Mixing is one of the most common phenomena in the world and the inherent law and mechanism of liquid–liquid homogeneous mixing are very important for many applications, such as in the fields of chemistry, material science, environment, food and pharmaceuticals (Baldyga and Bourne, 1990; Fourcade et al., 2001; Judat et al., 2004; Yoshida et al., 2005). Mixing is one of the core issues in chemical engineering, since fast and uniform reagent mixing is critical to proceed dissolution, reaction and separation process. Owing to the small mixing scale, micro-mixers (Chen et al., 2005; Li et al., 2007; Nguyen and Wu, 2005) have been widely used to carry out liquid–liquid mixing processes in recent years. Different micro-mixers with special mixing structures have been invented (Aoki et al., 2013; Capretto et al., 2011; Hessel et al., 2005; Liu et al., 2009) and it has been demonstrated that these micro-mixers have advantages on shortening molecular diffusion distance (to the range from 10 μm to 1000 μm) while providing large contact areas between fluids.

Although the micro-mixers are effective devices, they still need to be enhanced, due to the low Reynolds number ($Re < 200$) and non-ideal reagent space distribution, which induces long mixing time and low equipment efficiency especially for large mixing flow rate ratios or fluids with relatively high viscosity (Wang et al., 2011). Therefore, some passive and active enhancing methods have been developed in the mixing studies. The passive enhancing method (Long et al., 2009; Schönfeld et al., 2004; Zhang et al., 2010) mainly utilizes the energy from pumps and improves mixing by enhancing relative movement of fluid particles, which results in complex microchannel structure as well as possible dead volume. The active enhancing method, in contrast, is working with the help of extra energy input, such as ultrasound (Yaralioglu et al., 2004), piezoelectrically vibrating (Woias et al., 2000), electric field (Oddy et al., 2001), pressure pulse (Glasgow and Aubry, 2003) *etc*. Other than these direct energy input approaches,

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the introduction of inert fluid is another simple but effective method to intensify mixing process. The purpose of introducing inert fluid, such as gas (Günther et al., 2005; Zhang et al., 2014) or unreactive oil (Matsuyama et al., 2007), is to change the flow pattern of microchannel at low *Re* number, producing inner circular flow in segmented liquid plugs. Matsuyama et al. (2007) showed an example using silicone oil as the enhancing medium in a capillary mixer. "Villermaux–Dushman" parallel competition reaction system was adopted in their study to characterize the mixing performances at constant Peclet numbers ($Pe = ud/D$, where d is the inner diameter of the capillary, u is the average velocity and D is diffusion coefficient). The results showed the axial symmetrical reagent distribution in small water columns attained fast mixing owing to the fast recirculation, while the parallel liquid–liquid flow without inert oil obtained the largest segregation index. Günther et al. (2005) proposed a similar approach to enhance mixing of aqueous solutions by introducing an inert gas phase. This method greatly increased the mixing rate, decreased the mixing distance and obviously avoided complex channel fabrication. Using gas phase has an advantage for its inertia in most working systems and the operating cost is much lower comparing to the application of external fields. Fast gas–liquid phase separation is also easy to realize at the outlet of mixer. The resistance time distribution (RTD) (Chen et al., 2005; Yen et al., 2005a, 2005b) is narrow in this situation for the segmented flow pattern. Yen et al. (2005a, 2005b) made a further research about the channel structure effect on mixing in gas–liquid two-phase flow. However, they focused on the meandering microchannel, while the fluid contact structure of microchannel and the

Table 1
Physical properties of the working systems.

Mass fractions of glycerol (%)	0	20	40	50	65
Viscosity (mPa· s)	0.86	1.47	3.00	4.72	11.46
Density (g/cm^3)	0.997	1.049	1.101	1.127	1.166

Table 2
The concentration of each compound in Solution A.

Compounds	KI	KIO_3	H_3BO_3	NaOH
Concentrations (mol/L)	0.03	0.006	0.09	0.09

Table 3
The concentration of H^+ in Solution B corresponding to feeding ratio.

Feeding ratios (*R*)	5	10	25
Concentration of H^+ (mol/L)	0.025	0.050	0.125

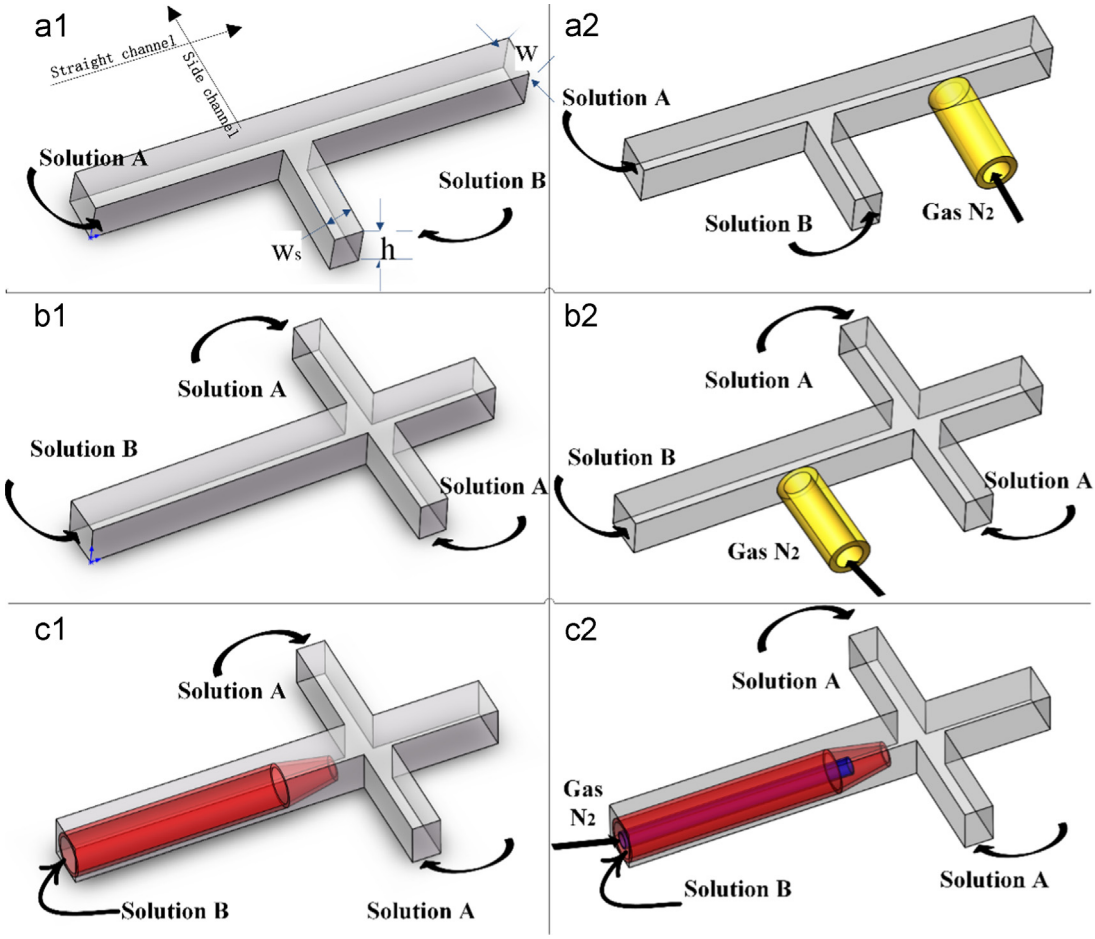

Fig. 1. Schematic diagrams of the three micro-mixers in the experiment. (a1) The T-junction microchannel without gas; (a2) the T-junction microchannel with side gas inlet capillary; (b1) the cross-junction microchannel without gas; (b2) the cross-junction microchannel with side gas inlet capillary; (c1) the co-flowing microchannel without gas; (c2) the co-flowing microchannel with inner gas inlet capillary. Both the inner diameters of the gas capillaries in (a2) and (b2) were 200 μm and the distances from capillary centers to junction centers were equal to 10 mm. For (c1) and (c2), the inner diameter of the tapered capillary was 300 μm in the wide end and 120 μm in the narrow end. In the center of the tapered capillary in (c2), the inner diameter of gas capillary was 100 μm and the outer diameter was 200 μm. The distance from the small capillary end to the big capillary tip was 300 μm, and the distance from the tapered capillary tip to the cross-junction center was 3 mm.