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Experimental study on the dispersed phase macro-mixing in an immiscible liquid–liquid stirred reactor

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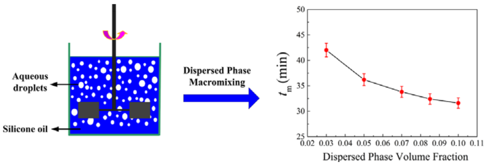
HIGHLIGHTS

- Novel experiment is designed to quantify the macro-mixing of a dispersed phase.
- Dispersed phase macro-mixing behavior differs from that of the continuous phase.
- Effect of impeller type is different to that on the continuous phase macro-mixing.
- The dispersed phase mixing time increases as the impeller clearance decreases.
- The dispersed phase mixing time decreases with the increase of the dispersed phase.

GRAPHICAL ABSTRACT



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ABSTRACT

The spatial nonuniformity of solute concentration in the dispersed phase of immiscible liquid–liquid dispersion is smeared out by coalescence and breakup of drops, which has a very significant effect on the productivity of relevant chemical processes. Novel experiments have been designed to visualize and quantify accurately the macro-mixing within the dispersed phase using the planar laser induced fluorescence (PLIF) method combined with a refractive index matching technique in this work. Batch experiments are carried out in a stirred reactor for an electrolytic solution dispersed in silicone oil. It is revealed that the dispersed phase macro-mixing behavior differs much from that of the continuous phase of multiphase systems and is heavily dependent upon the drop interaction rate and thus power consumption. The dispersed phase mixing time decreases with the increase of the dispersed phase volume fraction. Dispersed phase mixing time varies inversely with the power consumption per unit volume dispersion. The effect of impeller type on the dispersed phase mixing time is greatly different from that on the continuous/single phase mixing time. The effect of radial impeller clearance on the dispersed phase mixing time is contrary to that on the continuous/single phase mixing time.

1. Introduction

Liquid–liquid dispersions in mechanically agitated vessels are frequently encountered in chemical industry. Some typical applications are emulsion/suspension polymerization, solvent extraction, and production via chemical reaction. The intense drop breakup and coalescence as well as possible phase inversion make such processes extremely difficult to be well controlled. Numerous investigations have thus been devoted to the two-phase dispersed systems. Popular subjects of previous investigations included the minimum agitation speed for complete liquid–liquid dispersion (Armenante and Huang, 1992; Cheng et al., 2011; Skelland and Ramsay, 1987), drop breakage

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and coalescence (Tsouris and Tavlarides, 1994), interaction rate (Coulaloglou and Tavlarides, 1977; Madden and Damerell, 1962; Tobin et al., 1990), mean drop size and size distribution (Calabrese et al., 1986a, 1986b; Wang and Calabrese, 1986; Zhou and Kresta, 1998), phase inversion phenomenon (Kato et al., 1991), flow structure and turbulence (Laurenzi et al., 2009; Svensson and Rasmuson, 2006; Svensson and Rasmuson, 2004), and numerical simulation of flow characteristics (Laurenzi et al., 2009; Wang and Mao, 2005).

Mixing in liquid–liquid dispersions is a very complicated process, which occurs either in the continuous phase or in the dispersed phase, even in both phases. Although a considerable body of knowledge has been obtained (Leng and Calabrese, 2004), there is still very little information regarding the mixing in either or both phases in stirred liquid–liquid dispersions. The knowledge of macro-mixing, which is usually globally characterized by mixing time, is essential when the mixing rate is comparable with or slower than the chemical reaction rate (Raghav Rao and Joshi, 1988). Under these circumstances, the liquid–liquid mixing effect is found to have a profound impact on the overall performance, e.g., production cost and product selectivity. Recently, our group (Zhao et al., 2011; Cheng et al., 2012) studied the macro-mixing in the continuous phase of immiscible liquid–liquid systems using the conductivity technique, in which the tap water was the continuous phase and several inert oil phases were used as the dispersed phase, and these studies revealed a non-linear effect of the volume fraction of the dispersed phase on the mixing time of the continuous phase. The macro-mixing process in the continuous phase of multiphase systems or in single phase systems is achieved by convective (bulk) and turbulent (eddy) motion. However, the problem of mixing amongst the dispersed drops/lumps, i.e., the dispersed phase mixing, is of special interest for such practical applications as solvent extraction and suspension/emulsion polymerization with reactions occurring in the dispersed phase, in which the dispersed phase mixing can greatly influence the productivity and selectivity. Experimental measurements and theoretical studies by Curl (1963) and Miller et al. (1963) showed that the dispersed phase mixing had a very significant effect on the average reaction rate and product selectivity in non-first order or mass transfer controlled reactions occurring in the dispersed phase.

The dispersed phase mixing is fulfilled by means of intense drop interactions (dispersion, breakup and coalescence), which control the species concentration distribution among drop population. Usually, a drop dispersed in the continuous phase mixes with another through coalescence and breakup or re-dispersion, which tends to eliminate the concentration differences among drops. The drops are naturally completely segregated in liquid–liquid dispersions, if there is no dispersed phase mixing. On the other hand, there would be no segregation (no concentration difference among drops) if an infinite dispersed phase mixing rate (or interaction rate) among the dispersed drops is available. Rietema (1964) stated that the two extreme cases did not really exist in practical industrial processes and usually there was a finite interaction rate or partial segregation. The higher the rate of the dispersed phase mixing, the more uniform will the drop concentration distribution become. On analyzing the problem, there raises the question to what an extent the drop concentrations can be mixed and how long it usually takes for a pulse-added non-transferring tracer to be mixed to an expected extent.

To the best of our knowledge, the previous works on the dispersed phase mixing were mainly focused on the drop interaction rate by either the chemical reaction method (Hillestead, 1965; Komasawa et al., 1969; Madden and Damerell, 1962), the pure physical means with tracer dye (Curl, 1963; Miller et al., 1963; Ross and Curl, 1973) or impeller speed changes (Howarth, 1967; Mlynek and Resnick, 1972), and other methods (Groothuis and Zuiderweg, 1964). It is particularly noted that quantitative measurement of the macro-mixing process in dispersed phases is still unresolved, despite of its practical importance in design and control of such liquid–liquid operations.

Many measurement techniques have been developed over the past years such as visual observation of decoloration/coloration, transient conductivity/thermal/pH measurement, liquid-crystal thermograph, electrical resistance/impedance tomography and planar laser-induced fluorescence (PLIF) techniques (Nere et al., 2003). The local measurement-based methods such as transient conductivity/thermal/pH measurement techniques are not normally applicable to determine the mixing time in the dispersed phase for the drops break up and coalesce constantly. On the other hand, the complex image reconstruction, requirement of a certain kind of expertise and high instrument cost restrict the application of 3D measurement of electrical resistance/impedance/liquid-crystal tomography techniques.

The planar laser induced fluorescence (PLIF) is a non-intrusive optical diagnostic technique based on laser sheet visualization and image processing, by which the mixing process can be visualized and the corresponding tracer concentration distributions can be measured with high spatial and temporal resolutions. The principles of the PLIF technique were described in many papers (Law and Wang, 2000; van Cruyningen et al., 1990). This technique has matured during the past decades, and has been successfully adopted to quantitatively determine scalar concentrations and mixing processes by many investigators in various flow configurations; e.g., impinging jet (Unger and Muzzio, 1999), tee mixer (Pan and Meng, 2001), T-shaped micro-mixer (Hoffmann et al., 2006), confined wake flow (Hjertager et al., 2003), and stirred reactors (Fall et al., 2001; Guillard et al., 2000; Houcine et al., 1996). These reports demonstrated that the PLIF technique was a powerful and promising tool with capability of providing local data simultaneously at multiple positions (Guillard et al., 2000). However, application of PLIF is limited mainly to the measurement of tracer concentrations in single phase systems. The main obstacle to the application of PLIF to multiphase systems is that the refraction of light passing through liquid–liquid, gas–liquid and solid–liquid interfaces. The method of choice to eliminate the refraction problem in multiphase systems is to match the refractive index of two phases to avoid the refractive interference (Budwig, 1994).

This work contributes a novel experiment to visualize accurately and measure quantitatively the macro-mixing processes of a non-transferring dye in the dispersed phase in an immiscible liquid–liquid stirred reactor by PLIF combined with the refractive index matching for the immiscible liquid–liquid pair. The dispersed phase macro-mixing time measurement in this work involves the addition of a small volume of dye tracer solution into the dispersion in the vessel and recording its concentration variation within the monitored zone concurrently.

2. Experimental

2.1. Stirred reactor configuration

Experiments are carried out in a batch manner in a transparent plexi-glass stirred reactor (cylindrical, flat-bottom). The diameter (T) of the stirred reactor is 120 mm and the total height is 216 mm. Four vertical baffles with width (B) of 12 mm are equipped equally-spaced at the wall. The liquid height for all experiments is set at $H=T$. A tracer (Rhodamine B, A.R., Beijing Yinghai Fine Chemical Co., China) is injected at the liquid surface, 3 mm from the tank wall midway between two baffles. Different types of stirrers are used as shown in Fig. 1, namely, Rushton disk turbine (RDT), half circle blade disk turbine (HCDT), 45° pitched blade turbine downflow (PBTD) and 45° pitched blade turbine upflow