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On nature of mass transfer near liquid-liquid interface in the presence of Marangoni instabilities

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HIGHLIGHTS

- Physics of drop rise in quiescent channel with mass transfer is studied.
- Variation of length scales near the drop interface is analyzed.
- Energy transfer rate between length scales is calculated near the drop interface.
- Small scale structures control mass transfer near the drop interface.
- Variation mass transfer coefficient near the drop interface in time is reported.

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ABSTRACT

We report on mass transfer statistics in low Reynolds number system ($Re_p \sim 0.053$) with Marangoni instabilities. Data from our previous paper (Khanwale et al., 2015a) has been used concerning velocity and concentration fields around the drop interface measured using combination of Particle Image Velocimetry (PIV) and Planar Laser Induced Fluorescence (PLIF) techniques. To understand the effect of interfacial instabilities on mass transfer process, fluid dynamical variations resulting due to relative motion of two phases and the influence of deformation of interface are made negligible. This has been achieved by selection of fluid system with particle Reynolds numbers (Re_p) of 0.053 (creeping flow), Eötvös number, $Eo = 1.95$, and Morton number, $M = 78.20$. The multi-scale nature and similarity of physics with intermittent turbulence of velocity fields, arising due to Marangoni instabilities are reported in detail by Khanwale et al. (2015a). Calculation of mass transfer coefficient due to the aforementioned multi-scale nature of velocity field is non-trivial, and requires detailed investigation of flow structures. We employ advanced wavelet based methods to calculate length scales of flow structures, which are then used for detailed calculations of mass transfer coefficients using the small eddy formalism. Being an inherently dynamic system the temporal variations of mass transfer statistics are also reported.

1. Introduction

Understanding of mass transfer near interface of liquid-liquid systems with interfacial instabilities (Marangoni) is very important for efficient design of liquid-liquid contactors. Due to the multi-scale nature of velocity field near the interface in case of Marangoni instabilities; mechanism governing mass transfer is non-linear in nature. It is clearly seen from previous studies (for review see Chen et al., 2015) that interfacial instabilities enhance mass transfer, but precise calculation of local mass transfer statistics is non trivial because of entangled nature of concentration gradients with velocity gradients. Local mass transfer coefficients due to multi-scale nature of velocity fields similar to intermittent high Reynolds number turbulence vary spatially. But, previous studies (for review see Chen et al., 2015) mainly focused on calculation of overall mass transfer enhancement, seldom investigating local mass transfer statistics.

Number of authors (Lewis, 1954; Sherwood and Wei, 1957; Olander and Reddy, 1964; Bakker et al., 1966; Javed and Thornton, 1984; Javed et al., 1989) measured overall mass transfer coefficients for number of ternary systems. They observed higher mass transfer rates for the systems where interfacial turbulence was present. Sherwood and Wei (1957) reported that existing theories of mass transfer with chemical reaction, based on molecular diffusion, are not applicable in presence of interfacial turbulence.

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More recently effect of Marangoni instabilities on mass transfer efficiency has been studied by various authors (D'Aubeterre et al., 2005; Wegener et al., 2007, 2009b; Wang et al., 2011; Zheng et al., 2014). They also observed higher mass transfer rates in presence of Marangoni instabilities. These authors also studied the effect of drop size, initial solute concentration on mass transfer rates. It is well known that classical mass transfer models are unsuccessful in predicting mass transfer coefficients in the presence of interfacial instabilities (Henschke and Pfennig, 1999; Rose and Kintner, 1966; Sawistowski and Goltz, 1963; Wang et al., 2011; Handlos and Baron, 1957; Hubis and Hartland, 1986; Kronig and Brink, 1951; Steiner, 1986; Steiner et al., 1990; Wegener and Paschedag, 2012). Based on classical models of inter-phase mass transfer, various correlations (Kronig and Brink, 1951; Handlos and Baron, 1957) were reported to estimate mass transfer coefficient from single rigid drop to continuous phase (for details see Chen et al. (2015)). These models do not take into account physics involved in presence of Marangoni instabilities but were used to quantify the effect of Marangoni instabilities on mass transfer coefficients (Henschke and Pfennig, 1999; Wegener et al., 2007; Wang et al., 2013). If we look into the mass transfer process, the interfacial tension variations on the interface due to mass transfer are not strictly a fluid dynamical phenomena. However, it has causation, which results into a coupling of fluid dynamics and mass transfer. This coupling causes non-uniform shear distribution along the interface which leads to interfacial instabilities also known as Marangoni instabilities. The Marangoni instabilities manifests themselves in various forms of flow structures at liquid-liquid interface such as surface ripples, interfacial turbulence, and localized eruptions (Orell and Westwater, 1962; Maroudas and Sawistowski, 1964; Sawistowski, 1973; Pertler et al., 1995). Marangoni instabilities help to enhance mixing of fluid elements near the interface resulting in increase of surface renewal rates thereby promoting mixing in the interfacial region (Sherwood and Wei, 1957; Banerjee et al., 1968; Turney and Banerjee, 2013). This enhanced surface renewal rate thereby results in higher mass transfer rates (Olander and Reddy, 1964; Berg and Haselberger, 1971; Lode and Heideger, 1970; Takeuchi and Numata, 1977; Javed et al., 1989; Henschke and Pfennig, 1999; Wegener et al., 2009a). The development of flow structures has a strong influence on the velocity field near the drop interface in both phases (Sternling and Scriven, 1959; Wegener et al., 2009a). Thus, concentration field is coupled with velocity field through a advective term in the species transport equation given by $u_i \partial C/\partial x_i$. This results in local concentration variation at the interface thereby influencing interfacial tension gradient given by $\frac{\partial \sigma}{\partial C}\frac{\partial C}{\partial x_i}$, which further leads to Marangoni stresses given by $\hat{n}((\partial\sigma/\partial x_j)\hat{n}_j)$, where $\hat{n}_j$ represents tangential vector to the interface. This cycle is pictorially depicted in Fig. 1. Thus calculation of mass transfer coefficients would require a detailed analysis of length scales involved in the chaotic flow structures near the interface, which can be done using the wavelet based methodologies used by Khanwale et al. (2015b) and Sona et al. (2014) for heat transfer.

There are number of theories in the literature which predict mass transfer statistics near interfaces in inertial flows, *vis-a-vis* small eddy model, large eddy model, surface divergence model, etc. (for a review see Mathpati and Joshi (2007)). These theories make a basic assumption about the nature of probability density function (pdf) of length scales of flow structures near the interface, which are responsible for scalar transport (concentration, temperature, etc.). The small eddy model by Banerjee et al. (1968) and Lamont and Scott (1970) assumes that the mass transfer near an interface is primarily due to the motion of dissipation scale flow structures which justifies the namesake. In our analysis we try to revisit this assumption by actually calculating the pdf of length scales and the respective energy transfer rates within the flow structures from our experimental measurements.

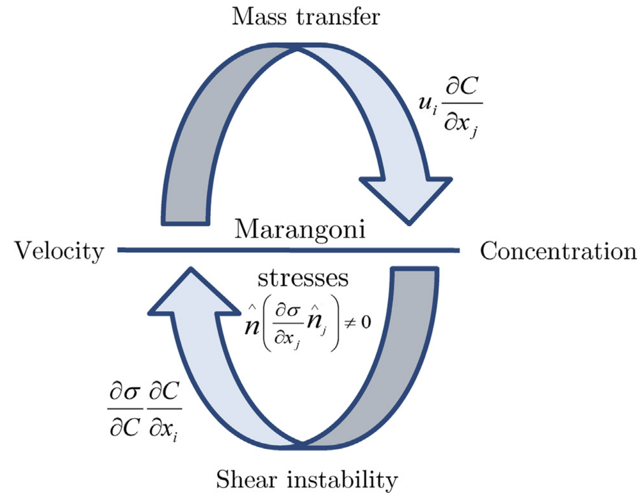

Fig. 1. Coupling of mass transfer and fluid flow in drop rise.

We use simultaneous Particle Image Velocimetry (PIV) and Planar Laser Induced Fluorescence (PLIF) data reported in our previous paper (Khanwale et al., 2015a) which includes velocity and concentration measured on a plane. To understand the effect of interfacial instabilities on mass transfer process, fluid dynamical variations resulting due to relative motion of two phases and the influence of deformation of interface are made negligible. To achieve this, the system we selected was a drop of toluene and acetone (dispersed phase) mixture rising in Glycerol (continuous phase), which corresponds to particle Reynolds numbers ($Re_p = \rho_c v_{drop} d_p/\mu_c$) of 0.053 (creeping flow), Eötvös number, ($Eo = g\Delta\rho d_p^2/\sigma$) of 1.95, and Morton number, ($M = g\mu_c^4\Delta\rho/\rho_c^2\sigma^3$) of 78.20. Where ρ_c is the density of continuous phase (kg/m^3), v_{drop} is drop rise velocity (m/s), d_p is diameter of the drop (m), μ_c is the viscosity of continuous phase (Pa · s), g is acceleration due to gravity (m/s^2), $\Delta\rho$ is density difference between the continuous phase and dispersed phase (kg/m^3), σ is interfacial tension (N/m). The Reynolds number (Re_D) based on the channel width comes out to be 0.517 which still is in the creeping flow regime. Analysis of this data showed that nature of velocity and concentration fields due to presence of Marangoni instabilities is similar to high Reynolds number intermittent turbulence (Khanwale et al., 2015a).

Using the aforementioned data sets we study parameters which govern the evolution of mass transfer statistics like pdfs of velocity and length-scales calculated using wavelet post-processing methodologies previously used in Deshpande et al. (2008), Roux et al. (1999), Sona et al. (2014), and Khanwale et al. (2015b). It is non-trivial to calculate these local mass transfer coefficients from the experimental data, once the system is multi-scale and turbulent like as explained earlier. This is where, we take the aid of the wavelets methodology, to analyse the experimental data in a systematic scale space. We also calculate the distribution of energy transfer within various length-scales of flow structures. We finally report on the variation of mass transfer coefficient near the drop interface as the drop moves upward in time.

2. Experimental method

Panel (b) of Fig. 2 shows a schematic of the experimental set-up used for the current investigation. The set-up includes a rectangular channel (with cross-section of 50 mm × 50 mm and height of