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Experimental study of the contribution of liquid film for liquid-liquid Taylor flow mass transfer in a microchannel

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HIGHLIGHTS

- Taylor flow mass transfer in channels with 0.6–2.0 mm diameters was studied.
- Mass transfer arises on both the end and the side surface of the droplet.
- Little dependency of droplet length on mass transfer was revealed experimentally.
- A mass transfer estimation method using a correlation between *Sh* and *Re* was proposed.

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ABSTRACT

Liquid-liquid two-phase Taylor flow mass transfer in microchannels, which has many advantages for heat and mass transfer operations, should be studied to develop practical applications in various chemical processes. To understand the effects of various parameters on mass transfer and develop an estimation method of mass transfer performance, mass transfer in microchannels was examined by an evaluation of phenol extraction from dodecane to water. Under stable Taylor flow conditions in a 1 mm inner diameter glass channel, the organic phase (dodecane) formed liquid droplets and the aqueous phase (water) formed liquid slugs with a liquid film enclosing the organic phase droplets. The effect of the droplet length on mass transfer was investigated with a microchannel device that consists of a circular channel with a 1 mm inner diameter and union tees with inner diameters of 2.0 mm and 2.4 mm. Although the droplets lengths were varied in the range of 7.4 mm to 30.5 mm, approximately the same volumetric mass transfer coefficients (*Ka*) were obtained. This result suggested that the side interface of the droplets that contact the thin liquid film becomes a mass transfer interface as well as an end interface. This occurs because the side interface area increases with increasing droplet length and droplet volume while the specific interfacial area remains approximately constant. The volumetric mass transfer coefficients were obtained for various cross-sectional shapes and hydraulic equivalent channel diameters at various flow velocities. The mass transfer coefficients were derived by dividing the volumetric mass transfer coefficients by the specific interfacial area. It was found that the Sherwood number (*Sh*), which is a dimensionless expression of the mass transfer coefficient, has excellent correlation with the Reynolds number (*Re*); thus, the estimation of the mass transfer performance for a given channel and given operational conditions can be determined using the correlation between *Sh* and *Re*.

1. Introduction

Liquid-liquid two-phase systems are widely operated in industrial chemical processes such as solvent extractions and catalytic reactions. Several types of apparatuses are used in two-phase operations. Because mass transfer is one of the most important functions for these apparatuses, many studies have been conducted to improve the mass transfer performance for conventional devices such as agitation vessels and columns. Much attention has been directed to a microchannel device, also called a micro device or a micro-structured device, which is an apparatus consisting of small spaces and channels. In a broader sense, a device with spaces and channels on the order of millimeters is also considered

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Nomenclature

a	specific interfacial area (m^{-1})
A_{end}	end interfacial area of droplet (m^2)
A_{side}	side interfacial area of droplet (m^2)
C	concentration of solute (mol m^{-3})
C^*	equilibrium concentration of solute (mol m^{-3})
C_0	initial concentration of solute (mol m^{-3})
Ca	capillary number
d	channel diameter (m)
d_d	droplet equivalent diameter (m) (diameter of a spherical droplet with the same volume)
d_H	hydraulic equivalent diameter of channel (m)
d_J	inner diameter of two-phase junction of channel device (m)
D_d	droplet side diffusion coefficient (m^2 s^{-1})
E	extraction efficiency (%)
E^*	equilibrium extraction efficiency (%)
h	film thickness (m)
Ka	volumetric mass transfer coefficient (s^{-1})
K	mass transfer coefficient (s^{-1})
L	channel length (m)
L_d	droplet length (m)
r	channel radius (m)
Re	Reynolds number
Sh	Sherwood number
t_e	exposure time (s)
u_{cir}	velocity of internal circulation (m s^{-1})
U	two-phase mixture velocity (m s^{-1})
U_d	droplet velocity (m s^{-1})
V_d	droplet volume (m^3)

Greek symbols

σ	interfacial tension (N m^{-1})
ρ_c	continuous phase density (kg m^{-3})
μ_c	continuous phase viscosity (Pa s)
ρ_d	dispersed phase density (kg m^{-3})
μ_d	dispersed phase viscosity (Pa s)
ρ_{mix}	mixture density (kg m^{-3})
μ_{mix}	mixture viscosity (Pa s)
δ	thickness of boundary layer (m)
τ_{nor}	dimensionless circulation time
τ_{cir}	circulation time (s)
τ_{travr}	time that the droplet travels a distance its own length (s)

a microchannel device. Microchannel devices offer more efficient mass transfer and significant intensifications for two-phase operations. Much research on the hydrodynamics and mass transfer characteristics for two-phase systems in microchannel devices has been conducted.

Various two-phase flow patterns in microchannel devices have been reported for both gas-liquid and liquid-liquid systems (Dreyfus et al., 2003; Triplett et al., 1999). Taylor flow is one of the common flow patterns observed in microchannel devices. In Taylor flow, the dispersed phase is enclosed by the liquid slug and the liquid film of the continuous phase to form a liquid droplet or a gas bubble. The liquid slugs are arranged on both adjacent sides of the droplet or the bubble, and the liquid film is formed on the channel inner wall, connecting with the liquid slugs. Thus, the dispersed phase droplets are separated from each other by the continuous phase. The internal circulation flow caused by the shear between the wall surface and the axis of the droplets and slugs is also known as a characteristic of Taylor flow in microchannels.

The existence of the liquid film in Taylor flow has been demonstrated for both gas-liquid and liquid-liquid systems. The thickness of the film (h) in gas-liquid Taylor flow has been correlated with the capillary number (Ca) by Bretherton (1961) as follows:

$$h = 1.34 r Ca^{2/3} \tag{1}$$

$$Ca = \frac{\mu U}{\sigma} \tag{2}$$

where r is the radius of the channel, μ is the viscosity, σ is the interfacial tension and U is the average velocity of two-phase flow.

Ghaini et al. (2011) revealed a fully developed liquid film for liquid-liquid Taylor flows with water and various organic compounds in a glass channel and a PTFE channel of 1 mm diameter. They used the laser induced fluorescence (LIF) method and quantified the film thickness by analyzing the fluorescence intensity profile on the channel cross section. The results demonstrated that the dependency of the liquid properties and mixture velocities on the film thickness qualitatively agreed with the tendencies demonstrated in the gas-liquid system by Bretherton (1961). The research showed that the film thickness increases with increasing mixture velocity and viscosity; the thickness decreases with interfacial tension.

Studies on the internal circulation flow for Taylor flow in microchannels have also been conducted using experimental techniques and simulations.

Thulasidas et al. (1997) applied particle imaging velocimetry (PIV) to determine velocity distributions inside liquid slugs for gas-liquid Taylor flow in circular and square channels. The recording images indicated that the counter rotating vortexes inside the liquid slug which enhance mixing inside the slugs. To characterize mixing inside the slug, the average circulating time of a fluid particle trapped in the counter rotating vortex was defined as the time for the liquid to move from one end of the liquid slug to the other. The nondimensional circulation time, which is defined as the ratio of the circulation time to the time for the slug to travel a distance twice its length, were demonstrated to be relatively constant for Ca values less than 0.01 and rapidly increase with Ca values greater than 0.01 in both the circular and square channels. The nondimensional circulation time values for $Ca < 0.01$ were approximately 2.0 and 3.0 for circular channels and square channels, respectively.

Tsaoulidis and Angeli (2016) conducted μ-PIV experiments to investigate the hydrodynamics of liquid-liquid Taylor flow in circular channels. The working fluids used were a 30 vol% tributylphosphate (TBP)/ionic liquid ([C_4mim][NTf_2]) mixture and a 3 mol L^{-1} HNO_3 solution as the continuous phase and the dispersed phase, respectively. The circulation flow within the droplets was quantified with the nondimensional circulation time, and the results showed that the nondimensional circulation time increases slightly (3.1–3.7) with increasing mixture velocity (0.01 m s^{-1} and 0.03 m s^{-1}), indicating mixing enhancement for the 1 mm diameter channel. In these results, however, the nondimensional circulation times were nearly constant in different droplets length (0.8–2.3 mm) for each mixture velocity (0.01 m s^{-1} and 0.03 m s^{-1}). The tendency of nondimensional circulation time with