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Hydrodynamics and mass transfer in liquid-liquid non-circular microchannels: Comparison of two aspect ratios and three junction structures

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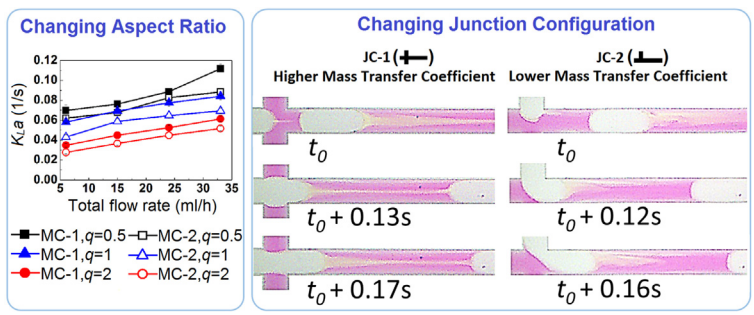
HIGHLIGHTS

- Mass transfer using different aspect ratios and mixing junctions were compared.
- K_La and K_L in a square channel were higher than those in a rectangular channel.
- Using a cross-shaped junction improved K_La up to 35 percent.
- Effect of the junction structure on mass transfer was greater than the aspect ratio.

GRAPHICAL ABSTRACT



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ABSTRACT

Hydrodynamics and mass transfer in liquid-liquid slug flow inside a square microchannel (MC-1, aspect ratio 1) and a rectangular microchannel (MC-2, aspect ratio 2) with identical hydraulic diameter (400 μm) were investigated experimentally. The results showed that the overall volumetric mass transfer coefficient (K_La) in MC-2 was 16.2% lower than that in MC-1 due to enlargement of the dispersed phase slugs in MC-2, leading to development of stagnant zones within plugs and reduction of wall film renewal. Three different junction configurations (JC) were studied. K_La was enhanced by up to 34.8% using JC-1, compared to JC-2 or JC-3 due to greater movement of the biphasic interface, resulting in higher mixing within slugs using JC-1. The junction arrangement influenced not only the mass transfer during the slug-formation stage, but also the slug-flowing stage due to effects on the vortex patterns along the microchannel.

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1. Introduction

Multiphase flow in microchannels has been of scientific interest due to high surface to volume ratio in the range of 5000–30000 m^{-1} [1], leading to intensified heat and mass transfer [2,3]. Besides the high interfacial area, narrow drop size [4] and residence time distributions [2] as well as decrease of diffusion distances [5,6] are among other pros of microstructured devices. Mass transfer coefficients in two-phase microstructured contactors are two to three orders of magnitude higher than those of conventional equipment [7] such as mixer-settler [8], rotating disk [9], packed [10], pulsed [11] and spray [12–15] columns.

Mass transfer in liquid-liquid systems has been studied in the literature. Skelland and Wellek [16] investigated mass transfer through droplets in which internal circulations were formed and the interface had no oscillation. This flow structure was close to

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Nomenclature

a	specific interfacial area (m^2/m^3)
A_{int}	interfacial surface of two phases (m^2)
A_{mc}	cross sectional area of microchannel (m^2)
C_{aqu}	bulk concentration of solute in aqueous phase (mol/m^3)
C_{org}	bulk concentration of solute in organic phase (mol/m^3)
C^*_{org}	equilibrium concentration of solute in organic phase corresponding to bulk concentration of solute in aqueous phase (mol/m^3)
C^0_{org}	initial concentration of solute in organic phase (mol/m^3)
C^τ_{org}	concentration of solute in organic phase at color change length (mol/m^3)
$C^0_{NaOH,aqu}$	initial concentration of NaOH in aqueous phase (mol/m^3)
Ca	*Capillary* number, $Ca = \mu U/\gamma$
D	depth of microchannel (m)
d_d	droplet equivalent diameter (m)
d_H	hydraulic diameter (m)
Ð	diffusion coefficient (m^2/s)
Fo	*Fourier* number, $Fo = ÐL_f/U_d h_f^2$
h_f	wall film thickness (m)
K_L	overall mass transfer coefficient (m/s)
K_La	overall volumetric mass transfer coefficient (1/s)
L_c	continuous phase slug length (m)
L_d	dispersed phase slug length (m)
L_f	film length (m)
L_{mt}	color change length (m)
L_{uc}	unit cell length (m)
m	partition coefficient
q	ratio of aqueous to organic phase flow rates
Q_{aqu}	volumetric flow rate of aqueous phase (m^3/s)
Q_{org}	volumetric flow rate of organic phase (m^3/s)
Q_{tot}	total volumetric flow rate (m^3/s)
Re	*Reynolds* number, $Re = \rho U d_H/\mu$
U	superficial velocity (m/s)
S_{caps}	surface of two caps of a plug (m^2)
V_d	volume of one dispersed phase slug (m^3)
V_{mc}	volume of microchannel (m^3)
V_{org}	volume of organic phase inside microchannel (m^3)
W	width of microchannel (m)

Greek symbols

β	fitting parameter in Eq. (1)
γ	interfacial tension (N/m)
δ	fitting parameter in Eq. (2)
λ	fitting parameter in Eq. (3)
μ	dynamic fluid viscosity (Pa s)
ρ	density (kg/m^3)
τ	color change time (s)
φ	holdup

Subscripts

aqu	aqueous phase
c	continuous phase
caps	caps of a dispersed phase slug
d	dispersed phase
f	film (cylindrical) section of a dispersed phase slug
M	mixture
org	organic phase
tot	total flow

that of liquid-liquid slug flow in microscale contactors [17]. Therefore, Di Miceli Raimondi et al. [17] compared their experimental results obtained in liquid-liquid microchannels with the values predicted by the Skelland and Wellek model (Eq. (1)).

$$K_La = \frac{\bar{D}_d a}{d_d}\beta_1\left(\frac{\rho_c U_d d_d}{\mu_c}\right)^{\beta 2}\left(\frac{\mu_c U_d}{\gamma}\right)^{\beta 3}\left(\frac{4\bar{D}_d U_d}{(L_f+\pi d_H/2)d_d^2}\right)^{\beta 4}\left(\frac{\mu_c}{\rho_d \bar{D}_d}\right)^{-0.125} \quad (1)$$

Among different flow regimes observed inside liquid-liquid microchannels, slug flow has been considered as an important one [18]. Di Miceli Raimondi et al. [19] proposed a model to predict mass transfer coefficients during liquid-liquid slug flow in microchannels:

$$K_La = \delta_1\frac{a}{d_d}\left(\frac{V_d}{V_{uc}}\right)^{\delta 2}(U_d d_H)^{\delta 3}\left(\frac{U_d}{\gamma}\right)^{\delta 4}\left(\frac{d_H}{d_d}\right)^{\delta 5} \quad (2)$$

where d_d is the diameter of a spherical droplet with the volume equal to that of the dispersed phase slug (droplet equivalent diameter). The model suggested by Di Miceli Raimondi et al. includes the size of the microchannel and is developed based on 2D direct numerical simulations [19]. Kashid et al. [20] developed an empirical correlation to predict the overall volumetric mass transfer coefficient in liquid-liquid slug flow in polytetrafluoroethylene (PTFE) capillaries. Physical properties of the phases and the microchannel size are taken into account in their model.

$$K_La\frac{L_{mt}}{U_{tot}} = \lambda_1\left(\frac{\mu_M U_{tot}}{\gamma}\right)^{\lambda 2}\left(\frac{\rho_M U_{tot} d_H}{\mu_M}\right)^{\lambda 3}\left(\frac{d_H}{L_{mt}}\right)^{\lambda 4} \quad (3)$$

Influence of physical properties of liquids on mass transfer performance of liquid-liquid microchannels has been presented in the literature [2,20–27]. Sattari-Najafabadi et al. [26] studied hydrodynamics and mass transfer in slug flow using n-hexane and toluene as organic solvents. The overall volumetric mass transfer coefficient in toluene/water slug flow was significantly greater than that in n-hexane/water slug flow. The higher viscosity of toluene as well as the lower interfacial tension between water and toluene, compared to those of *n*-hexane led to improvement of the internal circulations within slugs, resulting in enhancement of the mass transfer coefficients [26]. Some researchers have investigated the influence of microchannel size on mass transfer in liquid-liquid slug flow [18,21,23,28–30]. Kashid et al. [29] examined mass transfer coefficients in slug flow inside PTFE capillaries with internal diameters of 0.5, 0.75 and 1 mm. Decreasing the capillary size led to enhancement of both extraction efficiency and K_La. Identical sizes of PTFE tubes were employed by Ghaini et al. [23]. They reported insignificant enhancement of the mass transfer coefficients by reducing the capillary size. Recently, the influence of non-circular microchannel size on hydrodynamics and mass transfer in slug flow has been studied by Sattari-Najafabadi et al. [30]. They used three square microchannels with the hydraulic diameters of 200, 400 and 600 μm. Their findings showed that reducing the microchannel size enhanced the interfacial area because of enlargement of the dispersed phase slug length and deteriorated mass transfer resistances due to improved internal circulations, resulting in considerable enhancement of the mass transfer coefficients. Regarding their findings, the effect of size on the specific interfacial area was remarkably greater than that on mass transfer resistances [30].

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