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Hydrodynamics and mass transfer characteristics of liquid–liquid slug flow in microchannels: The effects of temperature, fluid properties and channel size

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HIGHLIGHTS

- The effect of temperature on liquid–liquid hydrodynamics and mass transfer are studied.
- The flow characteristics are focused on systems with more viscous dispersed phase.
- A general flow map and transition criteria are proposed for wide viscosity range.
- The H_2SO_4 droplet velocity is significantly reduced due to high viscosity.
- Excluding diffusion coefficient, increasing temperature reduces mass transfer rate.

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ABSTRACT

Immiscible liquid–liquid flow patterns and mass transfer were investigated in circular PTFE capillaries, with toluene-sulfuric acid, toluene-water and ethyl acetate-water systems. By comparing the slug flow operation range at different temperatures and capillary diameters, the results revealed the roles of inertia and viscous force in the transition from slug flow to droplet flow, and from slug flow to annular flow. A universal flow map based on composite terms of $Ca_C Re_C^{0.5}$and$Ca_D^{0.7} Re_D^{0.5}$was proposed to represent the competition between interfacial tension and the inertia/viscous force, which can excellently predict experimental data and literature results with fluid viscosity ranging from 0.85 to 1200 mPa·s. Additionally, the effect of temperature on the dispersed phase slug velocity, specific surface area and mass transfer was investigated and discussed, providing incremental understanding on the flow hydrodynamics and better guidance for optimized reactor design.

1. Introduction

Microreactors have been widely applied to intensify processes in the area of emulsion production, synthesis of fine chemicals/pharmaceuticals, nano-material synthesis and other chemical applications [1–3]. Mircoreactors provide unique advantages of excellent mass and heat transfer performance, high specific surface area, easy operation and scalability and enable continuous/fast synthesis [4,5], which has become an important paradigm change in both lab researches and industrial applications [6–8]. Among the numerous successful applications, multiphase liquid–liquid systems are commonly employed [9–12]. The key points of liquid–liquid processes in microreactors are flow regime manipulation and droplet transportation. Accordingly, slug flow is preferred because of short transport path, uniform phase distribution, recirculation and low back mixing.

Besides slug flow, other flow patterns such as droplet flow and annular flow are also observed in liquid–liquid systems. The slug flow formation and stability (*or* transition from slug flow to other flow patterns) are influenced by a number of parameters including fluid viscosity, interfacial tension, phase ratio, channel geometry etc [11,13–15]. Hence, dimensionless numbers such as Reynolds number (*Re*), Weber number (*We*) and Capillary number (*Ca*) were usually used to develop flow maps. Zhao et al. [11] studied kerosene-water flow in a rectangular PMMA microchannel and suggested using *We* of each phase to distinguish flow patterns as interfacial tension and inertial force were dominant at relatively high flow rate. When viscous force is the dominant force to balance the interfacial tension in more viscous systems, *Ca* and *Re* numbers are often applied for each phase due to their

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Nomenclature

a	specific surface area, m^2/m^3
a_{mc}	specific surface area of microchannel, m^2/m^3
Ca_C	Capillary number of continuous phase ($= j_C\mu_C/\sigma$)
Ca_D	Capillary number of dispersed phase ($= j_D\mu_D/\sigma$)
Ca_M	Mean Capillary number of two phase ($= j_{TP}\mu_M/\sigma$)
Ca_{TP}	Capillary number of two phase ($= j_{TP}\mu_C/\sigma$)
d_h	diameter of the microchannel, m
D	diffusion coefficient of ethyl acetate in water, m^2/s
j	superficial velocity ($= 4Q_C/(\pi d_h^2)$ or $= 4Q_D/(\pi d_h^2)$), m/s
k_L	mass transfer coefficient, m/s
k_La	volumetric mass transfer coefficient, s^{-1}
L	length, m
Oh	Ohnesourge number ($= \mu^2/d_h\sigma\rho$)
Q	flow rate, mL/min
Re_C	Renolds number of continuous phase ($= d_hj_C\rho_C/\mu_C$)
Re_D	Renolds number of dispersed phase ($= d_hj_D\rho_D/\mu_D$)
Re_M	Mean Renolds number of two phase ($= d_hj_{TP}\rho_M/\mu_M$)
Sc_D	Schmidt number of the dispersed phase defined by ($=\mu_D/\rho_DD$)
Sh_L	Sherwood number defined by ($=k_Ld_h/D$)
T	temperature, °C
U_D	dispersed phase slug velocity, m/s
We	Weber number ($= d_hj^2\rho/\sigma$)

Greek letters

β	volume fraction of the dispersed phase
μ	kinetic viscosity, Pa·s
ρ	density, kg/m^3
σ	interfacial tension, N/m
μ_M	mean kinetic viscosity, Pa·s ($\mu_M = (\beta/\mu_D + (1-\beta)/\mu_C)^{-1}$)
ρ_M	mean density, kg/m^3 ($\rho_M = (\beta/\rho_D + (1-\beta)/\rho_C)^{-1}$)

Subscripts

C	continuous phase
D	dispersed phase
o	oil phase
w	water phase
TP	two phase
M	mean

different roles in the flow pattern transitions [16–19]. It can be easily concluded that these studies and as proposed flow maps can hardly serve as a universal map for various fluid systems and operation conditions [20]. Recently, composite dimensionless numbers were applied and showed better performance in producing a universal map [15,20,21]. Yagodnitsyna et al. proposed to use the product with Weber number and Ohnesorge number ($We \times Oh$) to map their flow patterns obtained with three different fluid systems. The flow maps could well predict the flow patterns, which included many unconventional patterns obtained in fluid systems where both liquids wetted channel walls. Cao et al. [15] proposed two sets of Re^mWe^{-n} for the slug-annular flow transition and slug-droplet flow transition respectively through multiple regression method. Only very limited extension to other studies was validated from their comparison results. From the review above, it indicates that there are still huge efforts needed to be made in this area.

It was found that in our recent review [20] that most studies only involve low viscous fluids as the dispersed phase, whereas very few consider the condition of high viscous fluid carried by low viscous fluid. In fact, such operation is very beneficial for saving pumping energy (pressure drop) [19,22,23]. This is very important for various applications such as concentrated acids-involved reactions (*i.e.*, nitration, sulfonation, aklylation), ionic liquid-involved processes, polymerization and so on [5,24–27]. While the basic flow structures of slug flow remain similar, there are a lot of differences considering the operation range and mixing behavior [19,23]. A comprehensive knowledge remains lack to a large extent. Another shortage of literature studies on flow hydrodynamics is that they are nearly all conducted in room temperature, which is opposite to the fact that most reactions are operated under different temperatures. Temperature actually has significant influences on hydrodynamics which imposes additional influences on mass transfer and reaction processes. The detailed impacts of temperature on hydrodynamics and mass transfer hence require full investigation for improved reactor design. Based on these facts, this work aims at investigating the hydrodynamic and mass transfer characteristics of immiscible liquid–liquid systems at different temperatures, and particular attention will be paid to the effect of viscosity of the dispersed phase.

In the present study, flow patterns and slug hydrodynamics in circular PTFE capillaries were studied. Water-toluene, concentrated sulfuric acid (92 wt%)-toluene and water-ethyl acetate systems were tested at different temperatures and in various microchannels. The selection of the fluids was to approach real working fluid (sulfuric acid, to investigate concentrated acids-involved processes) or study systems with mass transfer (ethyl acetate) and with different fluid properties, namely viscosity, density and interfacial tension. Based on parametric study and qualitative force analysis, universal flow maps and flow transition criteria are proposed, which are also validated by comparing to other literature data. In this study, the slug flow hydrodynamics and mass

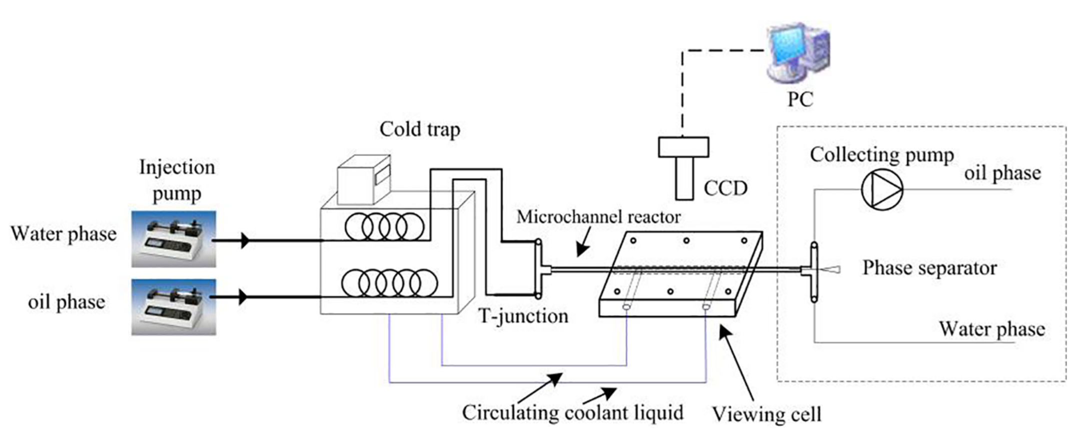

Fig. 1. The structure of hydrodynamics and mass transfer experimental setup.