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MASS TRANSFER PHENOMENA OF PARTIALLY MISCIBLE LIQUIDS UNDER LIQUID-LIQUID SLUG FLOW IN A CIRCULAR MICROCHANNEL

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Abstract

Microreactors have been demonstrated to become an effective tool for increasing mass and heat transfer in heterogeneous chemical processes that are unachievable in batch or continuous stirred tank reactors. The key to superior performance is the availability of a large specific surface area (A/V) in the microsystem for mass and heat transfer between phases. Among the flow patterns generated in the two-phase liquid-liquid flow, slug is an ideal flow with a high stability characteristic, regular velocity, and uniform dimension throughout a microchannel system. The flow behavior gives slugs great potential to increase mass and heat transfer between phases, so optimizing the utilization of microreactors in many chemical process applications needs an in-depth understanding of the liquid-liquid hydrodynamic. Therefore, the current work aims to determine *the mass transfer coefficient under liquid-liquid slug flow and the influence of slug dimensions, channel material, volumetric flow rate, and the flow rate ratio of the organic-aqueous phase on the mass transfer coefficient.* The experiments were performed in 1 mm (ID) circular PTFE and silicone tubes, with a 30-40 ml/hour flow rate, and sodium hydroxide concentrations of 0.1, 0.13, and 0.15 M were used. The results exhibit that the most prominent overall mass transfer coefficient was 0.121/s, attained at 0.15 M sodium hydroxide concentration, with 40 ml/hour total discharge within the silicone channel. *Microchannels generated smaller slug sizes at a higher aqueous phase flow rate than the organic phase. The smaller size provided a large specific surface area (A/V) for enhancing the mass transfer coefficient.*

Introduction

Microdevice technology plays an increasingly important role nowadays and has many positive impacts on the development of science and industrial applications. One of the rapidly developing fields in microtechnology is microstructure reactors (microreactors). Microreactors consist of several channels with an inner diameter of less than 1 mm (Prakash and Ghosh, Feb. 2021, Zhang et al., Feb. 2019). Because of the small dimension, the surface area *(A)* to volume *(V)* ratio that can be provided on a micro-scale under a hydraulic diameter range of ten to hundreds of micrometers is very high, attaining 10,000 – 50,000 m^2/m^3 (Dessimoz et al., 2008), a much greater value than conventional reactors which are only able to provide 100 – 400 m^2/m^3 (Jähnisch et al., 2004). The high surface area to volume ratio is a crucial aspect required for enhancing mass and heat transfer performance between phases in a unit process. The huge specific surface found in a microfluidic system is able to encourage the provision of a high mass and heat transfer coefficients up to 1.1/s (Ratchananusorn et al., 2011) and 25,000 $W/m^2.K$, respectively, which allows intensification of highly exothermic and or endothermic takes place effectively (Dessimoz et al., Aug. 2008, Wang et al., Nov. 2020, Ghaini et al., Apr. 2010).

The hydrodynamic of two-phase microfluidic systems has three different aspects to be considered: the first, the surface area to volume ratio; the second, the flow is characterized by a small Capillary number (C_a), which expresses that the influence of surface tension force is more predominant than viscous force in the system; and the third, micro-roughness and wettability channel wall exert significant influence to the formation of flow pattern. In a laminar multiphase flow system, the domination of interfacial tension and inertial force leads to various interfaces in regular shapes (Plutschack et al., 2017). There are many types of liquid-liquid flow patterns on a micro-scale. The main flow patterns observed in silicone and Teflon microchannels are annular, slug, and droplet flow (Tsaoulidis et al., 2013), but among the flow patterns, slug flow is ideal for enhancing mass and heat transfer due to the high stability characteristics shown by this flow pattern, and its ability to generate a substantial specific surface. Annular flow appears while inertial force works more dominant than interfacial tension force. Interfacial tension force tends to reduce interfacial area, whereas inertial force breaks the shape and drags the interface in the flow direction. As the competitive nature between inertial and interfacial tension forces, annular flow can be destabilized by changing the flow rate of both phases and the flow rate ratio. Slug and droplet flows are extensively studied because of the ease of hydrodynamic control and potential applications in fine chemical synthesis. Operations in the slug flow regime have been demonstrated to become a valuable tool for increasing reaction performance limited by mass and heat transfer. Furthermore, the ease of slug dimensions control allows a more effective mass transfer process and reaction control, which is unachievable in batch and continuous stirred tank reactors (Ratchananusorn et al., 2011). In this case, two mechanisms are found to be responsible for mass transfer between liquid-liquid two-phase flow: first, internal circulations within the slug; second, gradient concentration between successive slugs leads to diffusion between phases. With its advantages, microfluidic systems are employed for increasing mass and heat transfer in many industrial purposes, such as the dehydration of bioethanol (Suerz et al., 2021), synthesis of ethyl methyl oxalate from diethyl oxalate (Ji et al., 2020), Biodiesel synthesis through transesterification (Wu et al., 2016, Tiwari et al., 2018, Mazubert, 2014, Jamil et al., 2016), fabrication of wrinkled protein microcapsules (Feng and Lee, 2019), and generation of gelatin emulsions and microcapsules (Yeh et al., 2013).

Compared to conventional systems, microdevices offer other advantages, such as effective mixing processes and process safety, mainly for processes employing hazardous and toxic chemicals such as nitration and polymerization. However, the microreactor performance still needs improvements, considering the potential occurrence of fouling in some chemical process applications due to chemical degradation, which is difficult to detect and clean because of the small diameter of the channel. The emergence of this problem will undoubtedly be troublesome when the scale-up process is carried out. Therefore, more in-depth and comprehensive studies regarding the scale-up process toward industrial capacity are still urgently needed (Jensen, 2017).

Flow behavior greatly determines the sustainability of the mass and heat transfer processes on the micro-scale. Mass transfer characteristics on a micro-scale differ from those on a macro-scale, mainly related to thin liquid film formation in a narrow space between the slug and channel wall and the diffusion process between phases. On a micro-scale, liquid films emerge in a more delicate dimension. Hence, the distance of molecular diffusion on a micro-scale becomes shorter and increases the mass transfer coefficient significantly. Some previous researchers have worked on the mass transfer phenomena in microreactors. What distinguishes the current study from previous studies is the channel cross-section, channel material, and the liquid phase used. Previous studies employed channels with square (Dessimoz et al., 2008) and triangular cross-sections (Basher, 2021), while this study utilized circular cross-sections. The effect of channel materials was observed using glass tube (Dessimoz et al., 2008), PTFE (polytetrafluoroethylene) (Ghaini et al., Apr. 2010, Xu et al., Jul. 2013), PMMA (Kovalev and Yagodnitsyna, 2021), and PFA (Wang et al., 2020), while this study employed two different channel materials, i.e., silicone and PTFE. Sodium hydroxide-NBF ester was used as a liquid-liquid mixture (Ghaini et al., 2010), whereas the current study uses the liquid mixture of water-NaOH and CCl_3COOH-toluene as aqueous and organic phases. The tube cross-section, channel material, and different physical properties of the liquid determine the characteristics of the slug flow pattern generated. The various contact angles and liquid wettability on a channel wall affect the overall mass transfer coefficient (Wang et al., Nov. 2020, Liu et al., 2021, Antony et al., 2014).

The studies on mass transfer phenomena in microfluidic systems have been widely published (Zhang et al., Feb. 2019, Ratchananusorn et al., Nov. 2011, Wang et al., Nov. 2020, Liu et al., 2021, Zhang et al., 2012), but the previous results did not explain the correlation between slug dimensions and the mass transfer coefficient. In addition, previous studies have yet to study the impact of various channel materials on the mass transfer process between phases. Therefore, this work focused on exploring the liquid-liquid mass transfer phenomena regarding slug flow pattern and the influence of slug dimensions, channel material, volumetric flow rate, and the organic-to-aqueous flow rate ratio on the mass transfer coefficient.

Section snippets

Material

The hydrodynamic of two-phase microfluidic systems has three different aspects to be considered: the first, the surface area to volume ratio; the second, the flow was characterized by a small Capillary number (C_a), which expresses that the influence of surface tension force is more predominant than viscous force in the system; and the third, micro-roughness and wettability channel wall exert significant influence to the formation of flow pattern. The two-phase flow pattern observed in the…

Results and Discussion

The hydrodynamics were studied in 1 mm inside diameter using PTFE and silicone tubes at various sodium hydroxide concentrations and the organic-aqueous flow rate ratios at various total flow rates $(Q_d + Q_c)$ in the range of 30 – 40 ml/h. Among the liquid-liquid flow patterns that may be formed, slug flow exhibits the most stable characteristics. It has good potential for industrial applications involving mass and or heat transfer. Therefore, this study observed slug flow patterns focusing on mass…

Conclusions

Slug flow patterns were observed in the PTFE and silicone channels. In the microsystem, the gravitational force did not influence the flow formation. In the observed system, surface tension, inertial, and viscous force were the predominant forces influencing the flow system. Sodium hydroxide solution as an aqueous (dispersed) phase and CCl_3COOH-toluene as an organic (continuous) phase have better wettability in the silicone channel than the PTFE channels. The wettability of the sodium hydroxide …

Declaration of Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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