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The effects of viscosity, surface tension, and flow rate on gasoil-water flow pattern in microchannels

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Abstract−A microchannel was fabricated with glass tubes to investigate the effect of viscosity, surface tension, and flow rate on the liquid-liquid two-phase flow regime. Water and gasoil were selected as aqueous and organic working fluids, respectively. The two fluids were injected into the microchannel and created either slug or parallel profile depending on the applied conditions. The range of Reynolds and capillary numbers was chosen in such a way that neither inertia nor interfacial tension forces were negligible. Xanthan gum was used to increase viscosity and Triton X-100 (TX-100) and Sodium Dodecyl Sulfate (SDS) were used to reduce the interfacial tension. The results demonstrated that higher value of viscosity and flow rate increased interfacial area, but slug flow regime remained unchanged. The two surfactants showed different effects on the flow regime and interfacial area. Addition of TX-100 did not change the slug flow but decreased the interfacial area. In contrast, addition of SDS increased interfacial area by decreasing the slug's length in the low concentrations and by switching from slug to parallel regime at high concentrations.

Key words: Gasoil-water System, Microchannel, Slug Flow, Parallel Flow, Interfacial Area, Flow Regime

INTRODUCTION

Microchannels have recently drawn worldwide interest due to their significant advantages. In comparison with conventional reactors, these devices create higher surface to volume ratio, provide more efficient mass and heat transfer [1], and produce less chemical wastes [2]. Moreover, they are cheap, small and safe. Easier control of process parameters and reduced environmental hazards are other advantages of microchannels [3,4]. These devices have been fabricated using various materials with different methods [5].

Microchannels improve mass and heat transfer between two immiscible phases; that is why they have been used widely in the research programs [2]. Microfluidic behavior of gas-liquid systems has been studied in detail by many researchers [6-10]. Despite its broad application in microreactors, microextractors, and emulsification devices, fewer works have been carried out in liquid-liquid systems [11].

Water and gasoil as two immiscible liquids are brought into contact in some chemical processes such as gasoil refinery processes, in which mass transfer takes place between the two phases. The main barrier in mass transfer between water and gasoil is their immiscibility. Interfacial area and flow regime impinge directly on mass transfer between the two immiscible fluids [12-14]. Higher interfacial area and internal circulation of the flows in microchannels decrease the diffusion path, which results in higher rate of mass transfer between water and gasoil.

When two immiscible liquids are pumped into the microchannel, two types of flow regimes, namely parallel and slug, are observed depending on the operational conditions [15]. The flow pattern properties in the microchannels are mainly influenced by the channel geometry, interfacial tension, as well as the fluxes and viscosities of the two fluids [16]. Parallel flow consists of annular and parallel flows with wavy or smooth interface, in which two phases are flowing parallel, continuous, and concurrent. In slug (zebra, or segment) flow, continuous and dispersed phases coexist. Mass transfer in the slug flow is controlled by two mechanisms of internal circulation and concentration gradient between two adjacent slugs [12].

The flow pattern in microchannels is explained by both dimensionless numbers and physical properties of channel and fluids. The physical properties of microfluidic system are used to investigate the variation of interfacial area and drawing the flow map. It was observed that addition of Tween20 and Span80 surfactants had negligible effect on the slug length of deionized water-anhydrous octane system [17]. The slug volume or average slug length decreased as the flow velocities increased [18]. Flow regimes for three Y-junction microreactors (0.5, 0.75 and 1 mm internal diameters) as a function of volumetric flow rates of water and cyclohexane have been studied [19]. Three different regions were observed: region of drop flow at high volumetric flow rate of cyclohexane and low volumetric flow rate of water, region of deformed interface flow at high volumetric flow rate of water and low volumetric flow rate of cyclohexane, and a region of slug flow between these two regions [19]. The flow pattern maps for square, trapezoidal, Y-rectangular and concentric microchannels were investigated as a function of superficial velocities of water and toluene; no difference was observed in their performances [20]. The flow pattern map for silicone oil-water flow in a 250 μm microchannel initially saturated with oil versus Reynolds numbers of two phases was studied [21]. Slug flow was observed at low Reynolds numbers of water, while annular flow was observed at high Reynolds numbers [21]. Flow maps based on mean Capillary and mean Reynolds numbers were examined [12].

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The slug flow was dominant at high Reynolds and low capillary numbers, whereas parallel flow was dominant at low Reynolds and high capillary numbers.

Several organic compounds such as silicone oil and kerosene have been used as the organic phases in the liquid-liquid microfluidic systems [15,18,21-24]. The water-gasoil microfluidic system has not previously been examined, although it could be a potential candidate in the field of gasoil bio-refinery processes such as biodesulfurization. The main goal of this study, therefore, was to examine the flow pattern of simultaneous flows of water and gasoil in microchannels as a function of flow properties. The effects of interfacial tension, viscosity, and flow rate were studied. The results are not limited to gasoil-water system and may be extended to other two-phase flow processes in microchannels.

MATERIALS AND METHODS

1. Microchannel Apparatus

The experimental setup was composed of a microchannel, Y-junction, syringe pump (Fig. 1). Microchannel was fabricated with circular glass tubes (74 mm length×0.95 mm diameter) connected using thermal varnish (WOER RSFR Tube, Φ=1.0 mm) to form a channel with the total length of 170 cm. The Y-junction was made of a 10-mm-thickness Plexiglas sheet. Two syringes loaded with the fluids were connected to the inlet channels. Flow rates of the fluids were controlled by a self-made syringe pump. This pump gave a pulse-free and stable liquid flow.

2. Chemicals

Gasoil was obtained from Mashhad Oil Company in Iran. TX-100 was purchased from Merck. SDS was purchased from Guangdong Chemical Reagent Engineering-Technological Research and Development Center. Xanthan gum was obtained from a domestic market. Distillated water was used to prepare aqueous solutions.

3. Data Analysis

The ratio of interfacial area to volume was determined by number of slugs times the fluids interfacial area per volume of microchannel occupied by the fluids. The number of slugs was counted all over the microchannel length (170 cm) after the steady state condition was reached. The two liquids were introduced into the Y-junction by syringe pump at a ratio of 1 : 1, each with the flow rate of 90 μl·min^{-1}, unless otherwise noted.

Higher number of slugs is formed when slug lengths are shorter, which in turn results in higher interfacial area. Moreover, the internal circulation takes place more efficiently in a shorter slug length, because a shorter distance between the stagnant region and slug interfaces exists. Thus, mass and heat transfer are improved by higher number of slugs per unit length of a microchannel. In other studies slug lengths or volumes have been used to predict the available interfacial area to volume ratio [17-19]. The number of slugs per unit length of the channel is used in this study, because it has a direct relation to the interfacial area to volume ratio.

Reynolds and capillary numbers are the two most common dimensionless numbers used to characterize two-phase flow in microchannel reactors. Reynolds number is the ratio of inertial to viscous forces and the capillary number is the ratio of viscous to surface tension forces. The flow in the microchannels is dominated by inertial forces in high Reynolds numbers and by surface tension forces in low capillary numbers [22].

RESULTS AND DISCUSSION

1. Distillated Water-gasoil System

Upon injection of water and gasoil into the microchannel, a slug flow profile was observed in the range of Reynolds number between 3 and 60 and a capillary number between 10^{-5}-10^{-3}. The microscopic image of this zebra laminar flow is depicted in Fig. 2(a). The slug's curvature on the wall showed that the glass tubes were hydrophilic. The slugs resembled capsules, the most stable form, and did not change for several days even after stopping of the flow of the fluids. Fig. 2(b) illustrates water-gasoil slug flow and the internal circulation in the slugs. The fluids near the wall are at a slower speed compared to the central sections. Thus, in order to have stable slugs, the fluid next to the wall must migrate to the channel center.

2. Viscosity Effect

Xanthan gum has been used as a rheology control agent in aqueous systems and as a stabilizer for emulsions and suspensions. Viscosities of aqueous solutions are significantly increased even by addition of small amount of xanthan gum. The viscosity of aqueous solution versus xanthan gum concentration has been measured and the effect of xanthan gum concentration on the viscosity followed

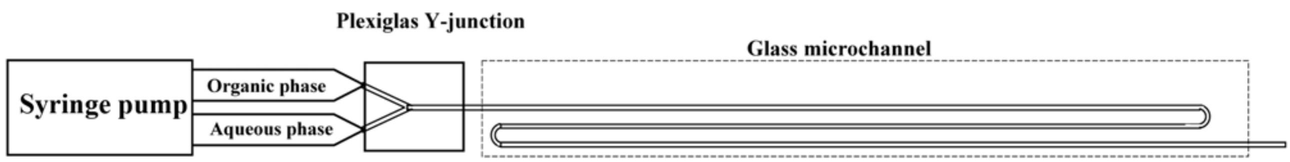

Fig. 1. Y-channel design of the microchannel.

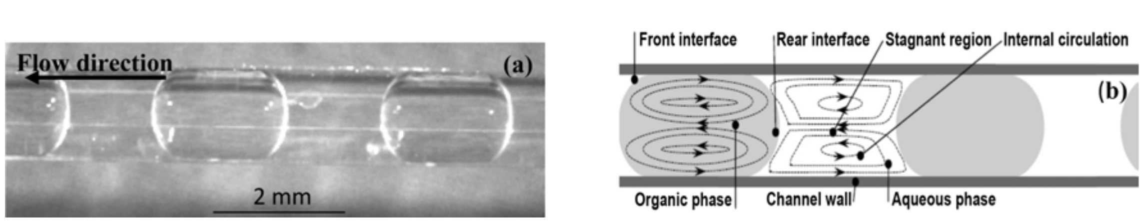

Fig. 2. Water/gasoil slug flow. (a) Microscopic image, (b) flow patterns. Flow direction is from right to left.

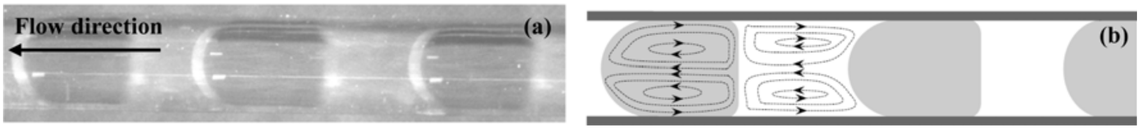

Fig. 3. Xanthan gum solution/gasoil slug flow. (a) Microscopic image, (b) flow patterns. Flow direction is from right to left.

an exponential relationship [25]. The gum increases water viscosity without significant changes in other properties of water.

The behavior of gasoil-water system containing 0-8 gL^{-1} xanthan gum was investigated in the microchannel. A slug flow profile was observed in the range of Reynolds numbers between 0.003 and 10 and capillary numbers between 10^{-5}-10^{-2}. As shown in Fig. 3(a), each gasoil slug looks like a bullet. The viscosity of aqueous phase containing 8 gL^{-1} xanthan gum was about 500 times that of the organic phase. The higher the xanthan gum concentration was, the sharper the front interface of the bullet slugs became.

The front face of the gasoil bullets was curved due to the effect of both viscous and interfacial tension forces. But the rear face of each slug was flat, because the surface tension and the viscous force acted in opposite direction and neutralized each other. Upon stoppage of the streams, the viscous force became zero and the surface tension force changed the gasoil bullets to capsules.

The polymeric bonds between xanthan molecules increased the solution viscosity, which in turn resulted in the formation of more slugs. The internal circulation of the slugs of xanthan gum solution is illustrated in Fig. 3(b). The distance between a stagnant region and slug interface was decreased in this case, which could give rise to more efficient diffusion process between the phases. As depicted in Fig. 4, the slug's number per unit length of microchannel was increased 14 percents at xanthan gum concentration of 6 gL^{-1} compared to pure water as the aqueous phase. However, the number of slugs was not increased by xanthan gum concentrations higher than 6 gL^{-1}.

3. Interfacial Tension Effect

Scientifically, in the absence of surface tension, every interface status is possible. But in the presence of surface tension, the interface must be circle or circular arcs limited at the channel wall [26]. SDS and TX-100 have been used to decrease the interfacial tension between water and gasoil. The surface tension of solution versus the concentration of TX-100 or SDS was measured by Soboleva and Erdogan respectively [27,28]. The surfactant concentrations were less than the critical micelle concentrations (CMCs) of the surfactants.

3-1. Effect of TX-100 Addition

TX-100 is a nonionic surfactant. A slug flow profile was observed in the range of Reynolds numbers between 3 and 57 and a capillary number between 10^{-5}-10^{-3}. Addition of this surfactant to aqueous phase decreased the number of slugs per a defined length of microchannel (Fig. 5). At constant Reynolds numbers, addition of TX-100 reduced the surface tension, which in turn increased the capillary number. Therefore, the effect of surface tension forces was decreased, which resulted in larger length and lower number of slugs. The slug shapes were similar to distillated water slugs shape. No parallel regime was observed during the TX-100 runs.

3-2. Effect of SDS Addition

Depending on the SDS concentration in the aqueous phase, two different regimes were observed. At low concentrations of SDS (less than 1.0 $g\cdot lit^{-1}$) slugs tend to form, but the flow regime was annular

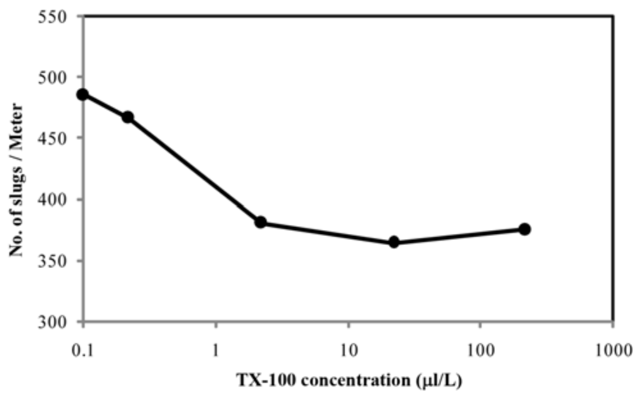

Fig. 5. Effect of TX-100 concentration on the number of slugs per 100 cm of microchannel length.

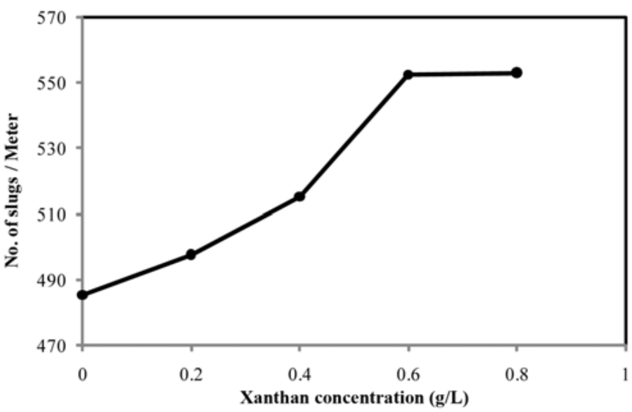

Fig. 4. Effect of Xanthan gum concentration on the number of slugs per 100 cm of microchannel length.

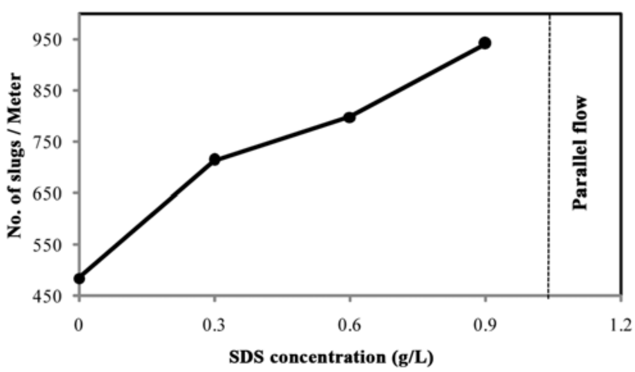

Fig. 6. Effect of SDS concentration on the number of slugs per 100 cm of microchannel length.

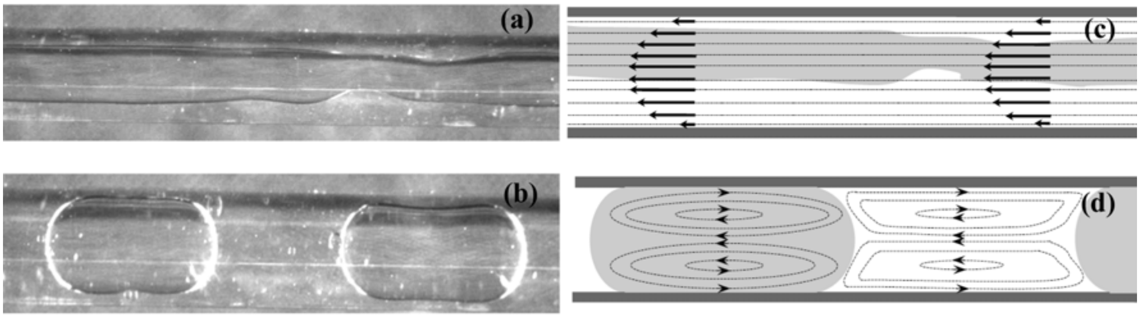

Fig. 7. SDS solution-gasoil system. (a) Microscopic image of annular parallel regime, (b) Microscopic image of slug regime, (c) Parallel flow velocity profile, (d) Slug flow velocity profile.

in the SDS concentrations in the range of 1.0-7.5 g·lit^{-1}. The numbers of slugs in the unit length of the microchannel as a function of SDS concentrations are depicted in Fig. 6. The number of slugs was doubled upon addition of 0.9 gL^{-1} of the surfactant.

The microscopic images of the SDS solution-gasoil system in the annular parallel and slug flow regimes are illustrated on Figs. 7(a) and 7(b), respectively. The interfacial area of the annular parallel flow regime (5052.6 m^2/m^3) was more than the highest value obtained for the slug flow (941.3 m^2/m^3) in this study. Although the parallel flow provides larger interfacial area compared to the slug flow regime, slower diffusion is expected in this flow pattern due to the lack of internal circulation phenomenon. The SDS solution-gasoil system in the annular parallel and slug flow regimes is illustrated in Figs. 7(c) and 7(d), respectively. Both fluids had the same flow rate and the parabolic laminar velocity profile was dominant in the channel. Thus, in the annular regime, the internal fluid had a higher velocity and a smaller cross sectional area than the fluid near the wall.

The behavior of SDS solution-gasoil two-phase flow can be explained based on the concentration of the surfactant. SDS is an anionic surfactant and the negative charge of its polar head may be the cause of its different behavior in comparison with the nonionic surfactant (TX-100).

Since the SDS molecules have negative charged head, they tend to be in close contact with the glass wall. Only at high concentrations of SDS were there enough molecules of the surfactant to cause the aqueous phase to cover the glass channel surface and flow all over the internal channel wall. Consequently, the organic phase was forced to flow in the center section of the channel and an annular parallel regime was formed. But below a low concentration of SDS, there were not enough SDS molecules to bond with the channel wall, so slug flow regime was formed.

In contrast, when a nonionic surfactant was added to aqueous solution, the organic head of surfactant molecules migrated to the two-phase interface. This was the reason for longer slug's length. As it was shown in Fig. 6, addition of low amounts of the anionic surfactant increased the slug's number. The charged head of SDS molecules as an anionic surfactants tended to be near the channel wall. Hence, there were not enough molecules to participate in the interface zone to increase the slug's length. Addition of SDS not only did not decrease the slug's number, even increased it. Unlike the surfactant charged head, the organic head of SDS molecules likes to be on the two phase interface; the tendencies of these two heads cause the shorter length and higher number of slugs.

4. Flow Rate Effect

The flow rates were changed using syringes with different volumes. Three flow rates were 90, 405, and 540 μl/min and the corresponding numbers of slugs per unit length of the microchannel (m) were measured to be 485, 522, and 553, respectively. The slug's curvature, however, remained unchanged and was independent of velocity. Interfacial tension force and hydrodynamic force between two fluids were the two important key factors involved in this phenomenon. An increase in velocity caused an increase in the hydrodynamic force, whereas it did not influence the surface tension. Thus, the slug's formation time was reduced and the slug's number increased subsequently.

Contact time is an important parameter that should be considered in the microchannels. Although the higher flow rate enhances the interfacial area and internal circulation, it decreases the contact time of the two phases. Reduction of contact time decreases the efficiency of the transport phenomena. That's why higher velocities are not recommended.

CONCLUSION

The effects of viscosity, nonionic and anionic surfactants, as well as the flow rates of two immiscible liquids (i.e., water-gasoil system) in a self-made microchannel were examined. It was revealed that different types of surfactants had different effects on the flow regimes. The results indicated that the ratio of interfacial area to the volume of fluids increased by increasing the xanthan gum concentration (viscosity factor), fluids flow rates, and SDS concentration (both interfacial tension and surface properties) and decreased by increasing the TX-100 concentration (interfacial tension factor). Higher values of interfacial area impinged on shorter both slugs' length and the distance between stagnant region and slug's interfaces, which could improve heat and mass transfer phenomena between the phases. The maximum enhancement of interfacial area was about 94% at 0.9 g·L^{-1} of SDS solution.

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