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Flow pattern diagrams of oil-water two-phase microflows and stable parallel flows obtained at low Reynolds numbers

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ABSTRACT

Immiscible liquid-liquid parallel microflows are useful in many applications. However, the immiscible two-phase flow behaves nonlinearly and it is a challenging task to control and stabilize the liquid-liquid interface. Oleic acid-water immiscible two-phase flow in microchannels was studied in the present work. The flow pattern diagrams were measured. Four different flow patterns, namely the single-phase flow of the aqueous phase, the droplet flow with the organic droplet dispersed in water, the parallel flow, and the single-phase flow of the oil phase were identified. Special attention was paid on transitions between the parallel flow and the other patterns of flow. Parallel flow formed under a proper balance between the driving force, the friction resistance, and the interfacial tension. The liquid-solid interaction as well as the liquid-liquid interaction played an important role in manipulating the liquid-liquid interface. Adding surfactants in the aqueous phase and raising the temperature altered both the liquid-liquid interfacial tension and the liquid-solid interaction. This led to a rather complicated transition behavior between the parallel flow and the droplet flow. The surface states of the solid walls were found to be the dominant condition in controlling the flow patterns in many cases. By applying a certain level of vacuum at the outlet of the channels, the first layer of water attached to the solid walls became lower in density and thicker in thickness. The flow resistance for both phases were remarkably lowered. Stable parallel flows at the Reynolds numbers of the oil and the aqueous phases as low as 1×10^{-5} and 8×10^{-3}, respectively, have been obtained without any chemical modifications of the solid surfaces, additional channel structures, or surfactants. A straight interface as long as several to tens of millimeters between the two immiscible liquid flows was successfully established and maintained stably by hours. Driving the flow with a negative pressure was verified to be a simple, effective, and controllable method for producing liquid-liquid immiscible parallel flows. It can be applied to various solvents and solutes and has potential in various applications, especially at low Reynolds numbers.

1. Introduction

Immiscible liquid-liquid two-phase microflows are widely used in fields like petroleum industries, chemical engineering, and biomedical science for mixing (Cheng et al., 2015), separation/extraction (Azimi et al., 2015; Bonet et al., 2015), reaction (Hisamoto et al., 2003; Zhao et al., 2002; Liu et al., 2014a, 2014b; Sugiura et al., 2008), emulsification (Liu et al., 2014a, 2014b; Xu et al., 2006), and partitioning (van der Linden et al., 2006). All these applications are related to the mass and heat exchange on the interface developed between the two phases. Under different flow conditions, the liquid-liquid interfaces formed in microchannels can take various morphologies. Flow patterns, such as droplet flow, segmented/slug flow, annular flow, and parallel flow, are classified accordingly (Guillot and Colin, 2005; Yagodnitsyna et al., 2015; Zhao et al., 2006; Azarmanesh and Farhadi, 2016; de Menech et al., 2008) and show great influences on the mass and heat transfer processes in microchannels (Kashid et al., 2011; Dessimoz et al., 2008; Chinaud et al., 2017). Multiple reagents can be transported in small droplets without dispersion (Song et al., 2006) while intensified mixing is realized by the internal circulation within the liquid segments or slugs (Günther et al., 2004). For applications like continuous flow chemical processing (CFCP) (Aota et al., 2009), it is then necessary to produce stable parallel microflows. The

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continuous interface established between the two immiscible phases flowing side-by-side enables the mass exchange or chemical reactions between the two streams to occur continuously. An important application of CFCP is the so-called multiphase laminar flow patterning (Zhao et al., 2002; Kenis, 1999). According to this technique, when reactive solutions are flowing in the microchannels in well-controlled parallel flow manner, various materials can be deposited on or removed from the inner walls of the microchannels to generate desired structures. This is a flexible method with good process compatibility for fabricating electrical electrodes and other integrated parts in situ in the microchannels.

However, due to the existence of the interface, immiscible two-phase parallel flows behave nonlinearly and the hydrodynamic instability may cause difficulties in controlling the interface morphology. These make it in many cases a challenging task to control and stabilize the liquid-liquid interface, especially at small size. For example, a stable parallel flow is obtained and maintained relatively easily when the Reynolds numbers for both phases are large and the inertia dominates. When the characteristic dimensions of the microchannels are very small and the Reynolds numbers become much smaller than 1, it is then easier to obtain droplet flows. If a long and stable parallel flow is expected at low Reynolds numbers, it has to be realized usually with the help of complicated fabrication processes of the flow devices, such as selective chemical surface modifications of the microchannel walls (Zhao et al., 2002; van der Linden et al., 2006; Aota et al., 2012) or guide structures on the solid surfaces (Aota et al., 2007). Microchannels with characteristic dimensions around or even smaller than 10 μm are usually found in microfluidic devices integrated in complicated multifunctional micro-electro-mechanical systems (MEMS) with miniaturization requirements or experimental simulation systems to study the geometrical confinement effects (Cui et al., 2004) and flow in quasi-2D porous media (Scholz et al., 2012). In such channels, additional wall modification and patterning may greatly increase the difficulty of the fabrication processes. To find out the determining factors for manipulating the two-phase microflow patterns without wall modification and patterning, the flow pattern diagrams are usually plotted against the superficial flow velocities (Salim et al., 2008), the flow rates (Guillot and Colin, 2005; Kashid and Kiwi-Minsker, 2011), the Reynolds number (*Re*) (Azarmanesh and Farhadi, 2016), the Weber number (We) (Yagodnitsyna et al., 2015; Zhao et al., 2006), or the Capillary number (Ca) (Azarmanesh and Farhadi, 2016).

In this paper, the flow pattern diagrams for oleic acid-water microflows in channels of 240 μm in width and 8 μm in depth were mapped. Special attention was paid on transitions between the parallel flow and the other flow patterns. Sodium dodecylsulphate (SDS) was applied as a surfactant and the temperature in the microchannels was varied to control the liquid-liquid interfacial tension. The flow pattern diagrams were plotted according to the driving pressures instead of the dimensionless numbers. This made it possible for us to understand better the pressure balance in such flow systems. A rather complicated transition behavior indicated that the liquid-solid interaction played an important role in modulating the flow patterns at small Reynolds numbers. By applying a certain level of vacuum at the outlet of the channels, the flow resistance for both liquids were remarkably lowered. Stable parallel flow at $Re < 10^{-2}$ has been obtained without any surface modifications, additional channel structures, or surfactants. A straight and stable interface as long as several to tens of millimeters between the two immiscible liquid flows was successfully established and maintained stably by hours. This is a simple and controllable method for producing liquid-liquid immiscible parallel flows. It can be applied to various solvents and solutes and has potential in various applications.

2. Materials and methods

2.1. Chemicals

The immiscible two phases were water and oleic acid, respectively. Water was purified by reverse osmosis with a Molecular Molelement 1805A system (Molewater) and possessed a resistivity of 18.25 MΩ·cm at 25 °C. In some experiments, SDS (chemically pure) was dissolved in the aqueous phase. SDS and oleic acid (analytical reagent) were used as received. A silicone elastomer kit (Sylgard 184) including a base material and a curing agent was purchased for making poly (dimethylsiloxane) (PDMS, Dow corning, USA) microchannels. A SU-8 2005 photoresist was obtained from Micro Chem.

2.2. Fabrication of microfluidic devices

The layout of a 'Y' shape microchannel is shown in Fig. 1a. It consists of two inlet channels and a main channel. The channel depth is 8 μm. A relief structure of the channel was first made by lithography with the SU-8 2005 photoresist on the surface of a silicon wafer as a replication master. Channels were then made in PDMS by replication molding. Each PDMS replica was cured for 45 min at 60 °C first and then 135 min at 100 °C. Inlet and outlet openings were punched in the PDMS piece. A top and a bottom plate made of glass or quartz cleaned with acetone and ethanol were bonded irreversibly to the PDMS replica to form a closed channel as shown in Fig. 1b. In some cases, a thin layer of PDMS precursor was spin-coated and solidified on the surface of the bottom plate. And in this case the top, bottom, and side walls of the corresponding microchannels were fully made of PDMS. This was called a homogeneous wall condition. If not, a heterogeneous wall condition was present. The top and side channel walls were made of PDMS but the bottom walls glass or quartz.

Before bonding, the plates and the PDMS replica were treated by O_2 plasma produced by a plasma cleaner (PDC-32G-2, Corning. Inc., USA) for 2 min and 30 s, respectively, to obtain hydrophilic channel surfaces. The top plate had a hole pattern drilled according to the placement of the inlet and outlet tubes. Polytetrafluoroethylene (PTFE) tubes were connected to the holes with silicone bushings used to fix and seal the join. The bottom could either be an ordinary microscope slide or a piece of conductive glass (1 mm in thickness) with an indium tin oxide (ITO) film sputtered on its surface. The square resistance of the ITO film was measured to be 13Ω/□. When an ITO glass plate was applied, electrodes were welded at the two ends of the ITO film. Thus a heater was integrated in the microfluidic device. The fluids in the microchannels could be heated by supplying a direct current through the ITO film. A K-type thermocouple was embedded in the PDMS layer to measure the temperature at the place of the main channel.

2.3. Flow control and visualization

The main channel was first saturated by water injected from inlet 2 through capillary effect. Water was then partially displaced by oleic acid, the oil phase, injected from inlet 2 to form the two-phase flow. High pressure gas (N_2 or Ar) was supplied form high-pressure tanks and connected to the inlets by PTFE tubes. The gas pressure was adjusted by needle valves and measured by pressure gauges installed just before the inlets. The outlet of the main channel could simply be exposed to atmospheric pressure or connected to a U-gauge by a three-way valve to supply a certain level of vacuum.

A microscope (TS100-F, Nikon, Japan) was used to observe the flow inside the microchannels. The microscope was focused on the upper surface of the bottom plate. Photographs and videos