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Layered two-phase flows in microchannels with arbitrary interface-wall contact angles

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HIGHLIGHTS

- A method for computing layered flow with arbitrary contact angles is developed.
- For steady flow, both free and pinned interfacial contact lines are considered.
- Contact angle induced interfacial curvature restricts the range of flow rates.
- The holdup and residence time of a fluid increases with its affinity for the wall.
- The pressure drop is lower when the *more* viscous fluid is more wetting.

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ABSTRACT

In this work, we study unidirectional, fully developed, layered two-phase flows in microchannels, where the interface meets the wall at an arbitrary contact angle. Interfacial tension causes the interface to take the form of a circular arc, with a radius that depends on the contact angle. The momentum equations must therefore be solved on a domain with boundaries that are, in general, not iso-coordinate surfaces. We adopt the technique developed by Shankar (2005a), which extends the use of eigenfunctions to arbitrary shaped domains, and apply it to layered flows for rectangular and circular cross-sections of the channel. This method is computationally efficient and allows us to analyze in detail the effect of the contact angle on flow properties. We focus on the case of a rectangular channel, which is commonly encountered in microfluidic applications, and consider two distinct cases: (a) free interface whose equilibrium contact angle is a function of fluid wetting properties, and (b) pinned interface whose apparent contact angle is determined by fluid flow rates. We calculate the relationship between the volume fractions (holdups) and flow rate fractions of the fluids and show that a non-zero contact angle can significantly restrict the range of permissible flow rates. This range is greater when the less viscous fluid has a greater affinity for the wall. For fixed flow rates, the residence time of a fluid is found to increase as its affinity for the wall increases. The pressure drop, which directly impacts operational costs, is found to be lower when the more viscous fluid is more wetting. This non-intuitive result is explained in terms of the corresponding variation in fluid volume fractions.

1. Introduction

Layered two-phase flow, in which two immiscible fluids flow in parallel alongside one another, has recently emerged as an important flow configuration in microfluidic applications. An experimental realization of fully developed steady layered flow in a microchannel is shown in Fig. 1. In such microchannels, capillary forces dominate over gravitational forces, due to the small length scales involved. This enables the liquids to be stratified either laterally or horizontally regardless of their relative density contrast. Moreover, the dominance of viscous forces over inertial forces damps out fluctuations and enables stable fully-developed layered flow to be realized for a wide range of flow rates. These characteristics of layered micro-flows have allowed them to be used in several microfluidic applications, such as solvent extraction (Assmann et al., 2013; Fries et al., 2008; Okubo et al., 2008; Znidarsic-Plazl and Plazl, 2007), separation of biomolecules (Huang et al., 2014),

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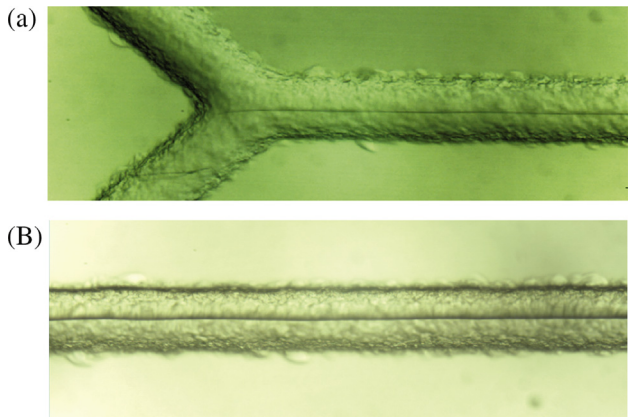

Fig. 1. Steady and uni-directional layered flow of immiscible fluids in a square glass microchannel of width 150 μm and length 5.56 cm. (a) Layered flow of an aqueous two-phase mixture of polyethylene glycol and sodium citrate (viscosity ratio of 5.71). Stable layered flow was obtained over a flow rate range of 69.3–124.8 μL min^{-1} for polyethylene glycol and 73.7–145.6 μL min^{-1} for sodium citrate. (b) Layered flow of water and toluene (viscosity ratio of 0.64), obtained for flow rates over a range of 80.5–220.8 μL min^{-1}, for both fluids. In both (a) and (b) the interface is seen as a thin straight line close to the center of the channel. The interface is observed to be steady and its position is seen to be invariant along the length of the channel, which implies fully developed flow.

treatment of toxic waste streams (Hotokezaka et al., 2005) and phase transfer catalysis (Aljbour et al., 2010; Šinkovec et al., 2013). These applications benefit from the high specific interfacial area, low diffusion path lengths, small inventories, and the ease of separation of outlet streams afforded by layered micro-flows.

In all the studies cited above, stable fully-developed layered two-phase flow was obtained experimentally over a range of moderate fluid flow rates. At low flow rates, capillary forces cause the less wetting phase to form droplets or slugs, whereas at very high flow rates the interface develops waves and ultimately breaks up to result in disordered dispersed flow (Zhao et al., 2006). At intermediate flow rates, however, the fluids are able to flow past the inlet junction of the channel without rupturing the interface, and attain a steady layered-flow configuration. Micrographs demonstrating two examples of fully-developed layered flow in a microchannel are presented in Fig. 1. In these experiments, stable layered flow was obtained over a considerable range of flow rates, of about 50–200 μL min^{-1}, in a square microchannel of width 150 μm (further details are provided in the figure caption).

These experimental observations, along with the many applications of layered micro-flows reported in the literature, have motivated the present study, which aims to develop a theoretical description of steady fully-developed layered flow in three-dimensional microchannels. While there is a vast literature on the dynamics of layered flows in planar geometries, i.e. flow between infinite plates (Pozrikidis, 2004; Valluri et al., 2010; Dietze et al., 2013; Sahu and Govindarajan, 2016), there are very few studies that account for three-dimensional channels and the influence of side-walls. In microchannels, the side-walls have additional significance because the configuration of the inter-fluid interface is dictated by interfacial forces acting at the contact line of the liquids with the side wall. Specifically, the contact angle of the interface with the wall depends on the relative wettability of the fluids with respect to the channel wall. Only if both fluids have the same affinity for the wall, will the contact angle be 90° (degrees). In general the angle will differ from 90°. For unidirectional fully-developed flow, this implies that the interface will not be flat across the width of the channel, but rather curved in the form of a circular arc (as shown later in Section 2). The presence of a curved interface can significantly modify the velocity profile, the flow rates and the pressure drop. The goal of the present work is to quantify and analyze these effects of the contact angle on the flow.

It is straightforward to solve for steady, unidirectional, layered flow in rectangular channels, when the interface between the fluids is flat as is the case when the contact angle is 90°. In this case, the interface and the walls align with Cartesian coordinate lines, allowing the use of the classic method of eigenfunction expansions to solve the associated Poisson problem. Consequently, most previous work has made this assumption, either implicitly or explicitly. Vir et al. (2014b) followed this approach to derive the relationship between the holdup (volume fraction) and the flow rate fraction of the fluids. Stiles and Fletcher (2004) derived a Fourier expansion for estimating laminar hydrodynamic spreading for stratified flows in microchannels. Studies of mass transfer in stratified microflows also make the flat interface assumption (Vir et al., 2014a; Okubo et al., 2008; Fries et al., 2008; Znidarsic-Plazl and Plazl, 2007). In reality, however, most fluids do not wet the wall equally and the contact angle departs significantly from 90°. This is demonstrated by Table 1, which lists some examples of liquid-liquid-wall combinations that are encountered in microfluidics and the corresponding measured contact angles. Here the contact angle is measured within the more wetting fluid, which is the first of the fluid pair.

The difficulty with accounting for contact angles other than 90° is that it implies a curved interface which does not align with the Cartesian coordinate system, thereby preventing the use of the classic eigenfunction expansions method of solution. In this work, we overcome this difficulty by applying the extension of the eigenfunction method developed by Shankar (2005a), which allows the use of eigenfunction expansions on arbitrary shaped domains. The procedure consists of embedding the complex geometry into a simpler larger domain in which a complete set of eigenfunctions exists. A least square approach is used to determine the coefficients of the eigenfunctions numerically, such that the residuals of the boundary conditions are minimized along the complex boundary. This method has been applied successfully to various problems. Shankar (2005b) illustrates the method with an example of two-

Table 1
Contact angles for various systems of two immiscible liquids in contact with a solid wall, as reported in the literature. The contact angle is measured in the more wetting phase, which is the first fluid in the pair. The majority of systems show significant deviations from 90°.

Liquids	Wall	Contact Angle	Reference
Gylcerin/47V2	Precision bore tube	71°	Fermigier and Jenffer (1991)
47V1000/Gylcerin	Standard glass tube	46°	Fermigier and Jenffer (1991)
Water/Toluene	Bare glass	10.2°	Aota et al. (2007)
Toluene/Water	ODS modified glass	12°	Aota et al. (2007)
Butanol/Water	Glass	83°	Kuban et al. (2003)
Water/Choloroform	Glass	74°	Kuban et al. (2003)
$[C_4mim][NTf_2]$/Deionised Water	Borosilicate glass	6°	Tsaoulidis et al. (2013)
Deionised Water/$[C_4mim][NTf_2]$	Teflon	20°	Tsaoulidis et al. (2013)