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Unstable parallel flows triggered by a fast chemical reaction

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ABSTRACT

We present an experimental investigation of an instability triggered by a fast chemical reaction in a low inertia parallel flow in a small channel. Two fluids evenly injected in a straight channel react creating a strongly stratified distribution of viscosity near their interface, which destabilizes the flow. Depending on the flow rates and the aspect ratio of the flow channel, several unstable regimes are observed: mixing regime, weakly stratified regime and a stable stratified regime. In channels of 1:1 aspect ratio we find most efficient mixing (and highest flow resistance) at an intermediate pressure drops, as the flow transitions between a fully 3D regime and more stratified regime. For channels of small aspect ratio the non-monotonicity is less evident: higher pressure drops lead to increased mixing.

1. Introduction

This paper continues our study of reactive multi-fluid flows in ducts at low *Re*, initiated in [3]. Duct flows of non-Newtonian fluids abound in industrial settings, from process and polymer engineering applications to oil well hydraulics and slurry pipelining. While many of these flows involve a single liquid a significant number involve multiple liquids. Two generic types of such flow are multi-layer flows and displacement flows. A difficult technological objective in such flows is to control the degree of mixing or interpenetration of the two fluids. Whilst in duct displacement flows some degree of mixing may be beneficial (or not), in multi-layer flows it is usually undesirable.

The underlying technique that we have been interested in is to control the yield stress of one of the fluids and to use this to influence the degree of mixing. In this context, one research direction has been targeted towards *stabilising* multi-layer flows by maintaining an unyielded region adjacent to the interface, e.g. [8,12,13,17]. This method is aimed at applications in co-extrusion, layering flows and lubricated transport, where the underlying process objective is to maintain stability at increasingly high *Re*.

A second research direction has been targeted towards *destabilising* multi-fluid flows by inducing strong non-monotonic viscosity gradients via fast chemical reaction, e.g. [3]. Our initial motivation was for controlling large scale displacement flows, e.g. in oil well construction, where it is sometimes necessary to displace at low *Re*. However, other processes such as those involving microfluidic operate in a low *Re* regime due to their small size, and this provides further potential applications.

In [3], we considered horizontal pipe displacement flows of a aqueous sucrose solution (at high pH, titrated with sodium hydroxide) with an aqueous Carbopol®-sucrose solution (at low pH). Neutralisation takes place where the fluids mix, resulting locally in a yield stress fluid. Thus, for mixtures between the two fluids we have a very high effective viscosity, much higher than the shear viscosity of either pure fluid. This strongly non monotonic viscosity distribution in the flow can destabilize the shear flow even in the absence of any significant inertial contribution. The instability reported in [3] was observed in the flow configuration illustrated schematically in Fig. 1(a). In this flow configuration we have shown that the chemically triggered instability manifests itself by creation of a strong vorticity near the interface between the two fluids which replenishes the reacted fluids as the flow destabilises and mixes. Ultimately this procedure results in effective transverse mixing and hence near complete displacement of the in-situ fluid.

Although the results of [3] were promising from the perspective of improving displacement efficiency, many aspects of the flow instability are poorly understood. A rudimentary linear stability analysis was performed in [3], assuming a non-monotonic viscosity profile depending on a pseudo-steady 1D base concentration field. This showed that linear instabilities are generated in a band of intermediate wavenumbers for sufficiently high viscosity variations. Although this may give insight into the onset of instability, the flows observed in [3] were well into the nonlinear regime, mak-

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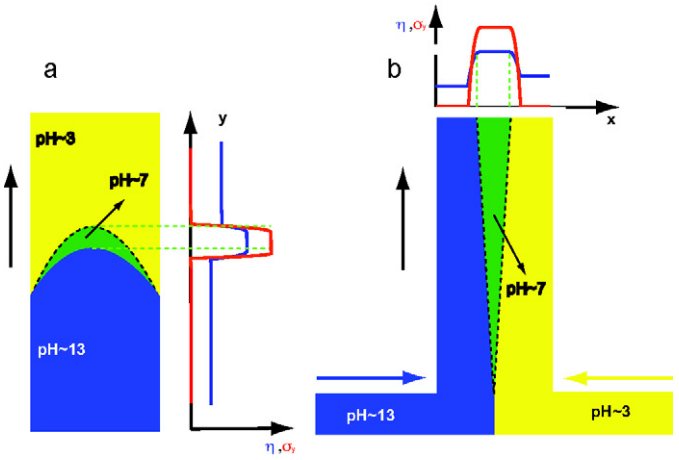

Fig. 1. Schematic view of the flow configurations: (a) displacement flow configuration investigated in [3]; and (b) parallel flow configuration.

ing such inferences hard to substantiate. Equally the mixing flows in [3] were fully 3D consisting of an inhomogeneous mixture of reacted solid-like fluids and regions of pure unreacted low viscosity fluid. This structure (or lack of it) makes traditional continuum modeling approaches difficult. This difficulty is exacerbated by the range of different timescales in the problem. Therefore, the part of the motivation for our study was to better understand the flows in [3] from an experimental perspective. The flow shown schematically in Fig. 1(a) has two main features: a frontal region and a multi-layer region along the sides of a developing displacement finger. In a moving reference frame the secondary flow at the front is extensional, while that along the sides is shear-dominated.

We wished to establish whether or not the shear flow was responsible for the mechanism of vorticity generation and continual replenishment observed in [3]. Therefore, in this paper we investigate flows in channels with 2 parallel injection ports, for which the basic flow configuration is a multi-layer shear flow, see Fig. 1(b). The first part of our study considers such flows in long channels of roughly 1:1 height to width ratio, so that similar 3D effects to [3] could manifest. The second part of our study considers long channels with a small height to width ratio (effectively a Hele-Shaw cell). Here the flow is understandably more 2D and structured.

Some of the objectives here include: (a) Could we chemically create a partition (of unyielded fluid) between two free moving streams of pure fluid? (b) Could one generate a nonlinear flow behavior with vanishingly small inertial contributions?

The latter point may find interesting applications in microfluidics where efficient mixing between two fluid streams is often desired. Methods for generating a nonlinear flow behavior in the absence of inertial effects have already been proposed. They may be based on either a specific patterning of the flow channels which can induce secondary fluid motion [25], or on the use of complex fluids characterized by a strongly nonlinear stress–strain dependence [10].

Other authors have considered reactive viscous displacements and these studies are unquestionably interesting, e.g. [4,14,27]. However, the main focus of these studies is on porous media style frontal displacements and the investigation of frontal instabilities. While the second part of our study is a more Hele-Shaw style geometry, for which a porous media analogy could be established, we shall later see that there is considerable complexity in the flow structures found. Hence establishing a clear relationship to these studies is difficult.

Recently, a number of authors have studied instability in miscible parallel flows, in pipes and channels, using Newtonian fluids, e.g. [5–7,9,11,22,24]. The focus of these recent studies is on convective instabilities promoted by a large viscosity difference between the fluids. In comparison to our work, these studies concern flows at moderate Reynolds number and the instabilities have a strong inertial influence.

An instability triggered by pH differences at the interface between two polyacrylic acid in a radial Hele-Shaw configuration has been reported in [18]. In spite of the differences in the flow configuration, the situation considered in Ref. [18] is somewhat similar to ours in the sense that a fast chemical reaction modifies significantly the rheology of an interfacial layer which ultimately influences the hydrodynamic stability of the flow.

Our paper is organised as follows. In Section 2 we describe the experimental setup, measurement methodologies and fluid rheologies. The results are presented in Section 3. We first discuss the observed flow regimes in each of our channels in Sections 3.1 and 3.2, respectively, using the *LIF* technique. We supplement these observations with results from digital PIV (Section 3.3) and from imaging of the distribution of the pH in the flow from which the rheological properties can be locally assessed (Section 3.4). The paper ends with a brief summary of the principal results.

2. Experimental details

2.1. Setup

All the experiments were conducted in the apparatus illustrated schematically in Fig. 2. Two fluids are injected through two inlets **I_1**, **I_2** at one end of an acrylic flow channel with a rectangular cross section **FC**. Their mixture is collected at the outlet **O**. The flow is smoothly driven by the hydrostatic pressure difference between the inlet fluid containers and the outlet fluid container. The two inlet fluid containers are 100 cc plastic syringes mounted on accurate (within 100 μm) vertical translational stages, fixed at the same height. The characteristic flow rates during the experiments were small and thus the static pressure drop did not change significantly during the experimental runs.

The top and side walls of the flow channel have been carefully polished to enhance their transparency and allow an optimal visualization and illumination of the flow patterns. The flow channel

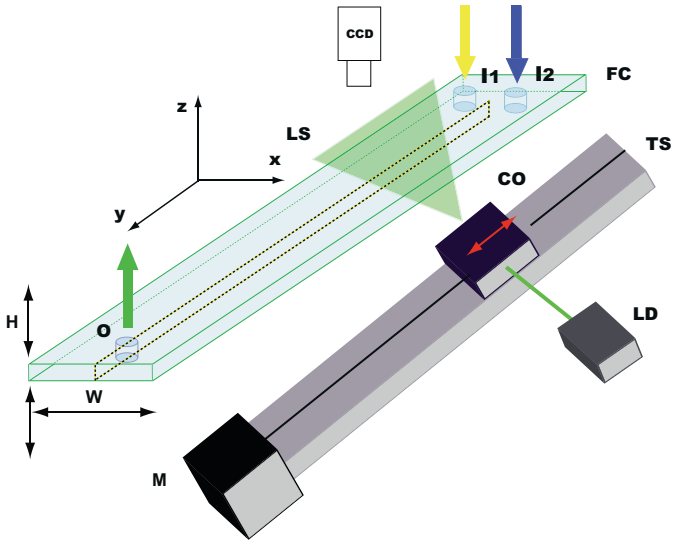

Fig. 2. Schematic view of the experimental apparatus: **FC** – flow channel, **I_{1, 2}** – fluid inlets, **O** – fluid outlet, **CCD** – video camera, **LS** – laser sheet, **TS** – translational stage, **M** – stepping motor, **LD** – laser drive, **CO** – cylindrical optics block.