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Ultra-fast microfluidic mixing by soft-wall turbulence

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HIGHLIGHTS

• Flow-soft wall interaction causes transition to turbulence in a microchannel.

- Multi-fold reduction in transition Reynolds number when compared to rigid channel.
- Post-transition, mixing time 5 orders of magnitude lower than that for laminar flow.
- Pressure drop significantly lower than rigid channel due to soft-wall deformation.
- Simple, low-cost, energy efficient strategy for ultra-fast mixing in microfluidics.

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ABSTRACT

Slow cross-stream mixing in micro-fluidic devices poses a significant challenge in realising efficient labon-a-chip technologies. Due to the small dimension and flow velocity, the flow is in the laminar regime, and this results in slow molecular cross-stream diffusion (in contrast to the fast turbulent mixing by cross-stream eddies in industrial applications). Here, we demonstrate a simple and powerful strategy for ultra-fast mixing in a microchannel with one soft wall with height as low as 35 μ m at a Reynolds number as low as 226. There is a spontaneous transition from a laminar flow to a turbulent flow state when the flow rate increases beyond a threshold value, resulting in complete cross-stream mixing. After transition, the mixing time across a channel of width 0.5 mm is smaller, by a factor of 10⁵, than that for a laminar flow, and complete mixing is achieved within a channel length of 2 cm. The increased mixing rate comes at very little energy cost, because the pressure drop is comparable to that required in current microfluidic devices, and it increases continuously and modestly at transition. This is because the channel length required to achieve complete mixing, 2 cm, is much smaller than that used in microfluidic devices that employ diffusive mixing; in addition, the deformation of the soft wall decreases the resistance to flow. © 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Advances in microfabrication have made possible the miniaturisation of complex reaction networks onto 'lab-on-a-chip' devices, in which the reactors and channels have smallest dimension in the range 10–100 μ m or less. Miniaturisation has several potential benefits, such as fast reactions in controlled environments with small volumes of reagents in devices of small size. However, the absence of quick and efficient sample preparation methods presents a technological barrier to realising the true potential of microfluidic technologies. Though applications that do not require sample preparation, such as blood glucose monitoring systems, have already been successfully commercialised, there are no successful commercial microfluidic systems which involve reactions and transformations of multiple fluid streams. This is because

http://dx.doi.org/10.1016/j.ces.2016.04.001 0009-2509/© 2016 Elsevier Ltd. All rights reserved. reactions take place only when the reactant streams mix at the molecular level, and it has long been recognised that slow mixing could be a bottleneck in the use of microreactors in commercial applications (Whitesides, 2006; de Mello, 2006).

The mixing rates depend on the fluid flow characteristics, which in turn depend on the dimension and the velocity of flow. For small channel widths and velocities in microfluidic applications, the flow is in the 'laminar' regime, and consists of smooth parallel streamlines with no cross-stream flow. Mixing across the channel in a laminar flow takes place by molecular diffusion, which is a very slow process with mixing times of the order of seconds or more. The time required for two streams to mix across a channel of width *W* can be estimated as (W^2/D) , where *D* is the molecular diffusion coefficient. The diffusion coefficient for small molecules in liquids such as water is of the order of 10^{-9} m²/s, while that for complex and polymeric molecules could be up to four orders of magnitude lower. Based on this, the time required for diffusion across a channel of width 0.1–1 mm is approximately

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10-1000 s.

Due to the relatively long time required, mixing in microfluidic devices is usually achieved using long channel lengths, of the order of a many tens of centimeters. A specific example of blood cell counting devices is discussed to provide a better context for the technological difficulties in microfluidic mixing. Proposed blood cell counting devices (van Berkel et al., 2011) employ microfluidic path lengths of 60 cm–1 m in order to achieve lysing of the red blood cells, and subsequent quenching in order to count white blood cells. Such long path lengths require high pressures for driving the flow of the order of a few atmospheres or more, and this increases the complexity of the auxiliary equipment required for generating such large pressures, as well as the chip fabrication procedures due to the strength required for withstanding high pressures.

Several strategies have been proposed for increasing mixing in microreactors. These can be broadly classified into passive strategies where the flow is steady but the streamlines are curved, and active strategies where time-dependence is introduced into the flows by external actuation. Passive mixers rely on generating complex flow pathways with curved streamlines, where fluid elements are stretched and rotated along different directions in order to increase mixing. These include channels with repeated bends to curve the streamlines (Kane et al., 2008; Jiang et al., 2004; Liu et al., 2000), wall grooves to introduce secondary flows (Stroock et al., 2002), hydrodynamic focusing where fluid streams with substantially different flow rates come into contact (Knight et al., 1998), split-and-recombine strategies (splitting the inlet into a large number of small streams using channel bifurcations and then recombining them by an inverse bifurcation) either in parallel (Bessoth et al., 1999) or in series (Lee et al., 2006), and chaotic advection inside microdroplets. Active strategies include pressure pulsing (Glasgow and Aubry, 2003), electrokinetic disturbances induced due to fluctuating electric fields (Bazant and Squires, 2004; Posner and Santiago, 2006), actuation by acoustic waves (Ahmed et al., 2009), and micron sized stirring devices (Mensing et al., 2004). In active strategies, there is additional energy input either by micron sized moving parts, or by external fields such as electric and ultrasound. In passive strategies, there is no external energy input. However, there is an energy cost because the curved streamlines, tortuous paths in split-and-recombine strategies, or secondary flows due to wall groves, dissipate additional energy due to fluid friction, and the pressure difference required to drive the flow is higher than that for a straight microchannel. These strategies are not amenable to economical scaling-up (or numbering-up) because they involve complicated micromachining of micron sized moving parts or actuators.

In nature, fast fluid mixing at large scales is facilitated by a transition to a turbulent flow, where there are large velocity fluctuations and significant cross-stream mixing due to turbulent 'eddies'. This results in mixing rates that are many orders of magnitude higher than those for a laminar flow at the same flow rate and conduit dimensions. The flow becomes turbulent at a Reynolds (Re = $(\rho Vh/n)$) number higher than about 1200 in a channel and about 2100 in a tube (Reynolds, 1883; Patel and Head, 1969). Here ρ and η are the fluid density and viscosity, V is the average velocity and h is the height of the channel or the tube diameter. Such high Reynolds numbers have been achieved in Polydimethylsiloxane (PDMS) based microfluidic devices of dimension about 150 µm (You et al., 2015), but these require very large velocities of the order of tens of meters per second, and very large flow rates of the order of 40 ml/min. The large pressure gradient required to drive the flow, of the order of 1-3 atmospheres, also requires very strong bonding between the surfaces. Strong bonding required for withstanding such high pressures is a formidable challenge, which has been overcome using advanced

vapour deposition techniques (You et al., 2015), and this represents a significant advance in fabricating high pressure microfluidic devices.

Recent experiments (Verma and Kumaran, 2013; Srinivas and Kumaran, 2015) have shown that there could be a transition to turbulence in a microchannel of smallest dimension as low as 100 µm if one of the walls is made sufficiently soft. These experimental studies followed the theoretical prediction of different types of instability in a channel/tube with soft walls. When the walls are soft, the transition Reynolds number depends on the parameter $\Sigma = (\rho G R^2 / \eta^2)$, the ratio of thicknesses of the wall material and fluid and the ratio of dissipation (viscosity) of the wall and fluid materials. Here, ρ and η are the fluid density and viscosity, G is the shear elasticity of the wall material¹ and R is the characteristic dimension (tube diameter or channel height). The transition Reynolds number decreases as the ratio of the wall thickness and fluid characteristic dimension increases, but it tends to a constant in the limit where the wall thickness is much larger than the fluid thickness, since the penetration depth for the wall displacement fluctuations is comparable to the fluid thickness. The transition Reynolds number does depend on the ratio of wall and fluid viscosities, as well as the details of the wall model, for the low Reynolds number instability (Chokshi and Kumaran, 2008), but there is little dependence on the wall dissipation at high Reynolds number. The transition Reynolds number shows systematic variations with the parameter Σ , which is the ratio of elastic stresses in the wall material and viscous stresses in the fluid. Different scalings of the transition Reynolds numbers with Σ have been derived in different asymptotic limits. It has been shown that there could be an instability even in the limit of zero Reynolds number (Kumaran et al., 1994; Kumaran, 1995; Chokshi and Kumaran, 2008) when the parameter (V_{η}/GR) exceeds a critical value, and the transition Reynolds number increases proportional to Σ in this limit. At high Reynolds number, there are primarily two modes of destabilisation. The high Reynolds number inviscid modes (Kumaran, 1998; Shankar and Kumaran, 1999, 2000) are a modification of the mode of instability in a rigid channel, and the transition Reynolds number increases proportional to $\Sigma^{1/2}$ for these modes. There is another class of instabilities called the high Reynolds number wall modes (Kumaran, 1996; Shankar and Kumaran, 2001), where the viscous effects are restricted to a thin region near the wall of the channel/tube; the transition Reynolds number increases proportional to $\Sigma^{3/4}$ in this case. The flow is destabilised by the transfer of energy from the mean flow to the fluctuations due to the shear work done at the solid-fluid interface. Weakly non-linear analysis has shown that the low Reynolds number instability is sub-critical (Shankar and Kumaran, 2001; Chokshi and Kumaran, 2008), whereas the high Reynolds number instability (which triggers the transition to turbulence in the present application) is super-critical (Chokshi and Kumaran, 2009). A summary of the linear stability studies is provided in Kumaran (2003) and Shankar (2015).

The low Reynolds number instability has been verified in experiments (Kumaran and Muralikrishnan, 2000; Muralikrishnan and Kumaran, 2002). Recently experiments in flexible tubes of diameter ~1 mm (Verma and Kumaran, 2012, 2015) have demonstrated that there is a flow instability at a Reynolds number as low as 500, which is significantly lower than the transition Reynolds number of 2100 for the flow through rigid tubes. The transition is induced by a dynamical coupling between the fluid and the soft wall material which results in wall motion and fluid

¹ The polymer gels that are used for the soft walls have a compression modulus that is an order of magnitude larger than the shear modulus, so they can be considered incompressible to a good approximation (Verma and Kumaran, 2012).

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