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Multiphase flows in microfluidics

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Abstract

Various aspects of microfluidic flows with different immiscible constituents are addressed. The fundamental physical characteristics are proposed, as well as the flow regimes which are determined by the wetting and surface tension properties. These fundamental aspects are followed by engineering applications that emerge in microfluidics, namely the creation of microbubbles or drops. Further applications are also discussed, such as the transfer of heat in bubbly flows, or the transport of colloids and emulsions. ***To cite this article: C.N. Baroud, H. Willaime, C. R. Physique 5 (2004).***

Résumé

Écoulements de fluides non miscibles dans des systèmes microfluidiques. Nous présentons ici quelques aspects d’écoulements de fluides non miscibles dans des systèmes microfluidiques. On rappelle les caractéristiques physiques fondamentales, on montre que les propriétés de mouillage des fluides sur les surfaces ainsi que les propriétés de tension superficielle sont déterminantes pour les écoulements. On présentera quelques applications de ces écoulements telle la formation de microbulles, de gouttes. On évoquera quelques applications tels les transferts thermiques, ou le transport de colloïdes ou d’émulsions. ***Pour citer cet article : C.N. Baroud, H. Willaime, C. R. Physique 5 (2004).***

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1. Introduction

We address various aspects of the flows of immiscible fluids in microchannels. The importance of these studies lies in the dominance of interfacial forces over bulk forces at microscales. Furthermore, important engineering applications emerge out of this scaling feature, such as the ability to form micron-sized bubbles or drops which are important in many pharmaceutical and food processing applications.

While the subject of multiphase flows in its full generality would require a very long review, we focus our interest on a subset of the possible systems. For instance, we limit our interest to the flow of fluids in closed microchannels, thus circumventing the rich area of ‘droplet microfluidics’ where one acts on drops that are transported on a solid substrate. Neither do we address the complete spectrum of possible parameter regimes, concentrating our attention for example on the low Reynolds number flows which are more typical for microfluidics.

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The range of parameters, as well as the most important physical mechanisms are discussed in Section 2. This is followed, in Section 3, by a review of the different flow regimes observed for multiphase flows in microchannels. A discussion of the engineering of drops and bubbles in microsystems is taken up in Section 4, finally followed by other engineering applications in Section 5.

2. Physical mechanisms in multiphase flows

In considering the physics of multiphase flows in microchannels, we note that it is largely dominated by the interfacial effects. This statement is quantified in the subsection below by first introducing the dimensionless quantities that come into play in such flows. Then the physical mechanisms of surface tension and wetting are described, followed by a discussion of the techniques to control these phenomena.

2.1. Dimensionless numbers

We are interested in flows that contain a moving boundary between two fluids. Such flows are described by a large number of parameters that describe the surface phenomena, bulk phenomena, as well as the variations in density and viscosity across the two fluids. Luckily, the small scale reduces the number of parameters, since viscosity or surface tension often dominate other effects that can be present in general.

Several parameters are naturally small in this case. For instance, inertial effects are small compared to viscous effects, yielding small Reynolds numbers. Gravitational effects will also generally be small compared to surface tension, yielding a small Bond number.

In contrast, some effects can be strong or weak depending on the details of the flow. In particular, the capillary number describes the relative importance of viscosity and surface tension. It is defined as $Ca = \mu U/\gamma$ where μ is the dynamic viscosity, U is a typical velocity, and γ is the surface tension coefficient. In flows where one fluid replaces another, the coherence or breakup of the interface will depend strongly on Ca, hence on the velocity of the moving interface. See for example [1] for a good review of the motion of drops in capillaries.

Another important dimensionless parameter is the Weber number, $We = \rho U^2 R/\gamma$ with ρ a typical density and R a length scale. The Weber number compares inertial effects to surface tension. Although the length scales involved are small, the strong dependence on the velocity can yield values of We that are large. This is particularly true when drops are ejected from a nozzle at high speed, as in inkjet printing for example.

Finally, in the presence of temperature gradients, buoyancy effects will be negligible (low Rayleigh number) but temperature dependent capillary effects can still exist. The Marangoni number characterizes the variations in surface tension with temperature. It can be written as $Ma = \sigma_T \Delta T d/\rho\nu\kappa$ [2], where σ_T is the surface tension coefficient, ΔT is the temperature difference over a distance d, ν is the kinematic viscosity and κ is the thermal diffusivity. Variations in surface tension due to temperature or surfactant concentration have been used to actuate motion in microfluidic flows, as discussed in Section 5.

2.2. Capillarity and surface tension

Let us now discuss the notions of capillarity and surface tension in some more detail. To understand capillarity, it is useful to look at atomic models of a fluid. At the atomic level, a fluid is a disordered state of matter where thermal agitation of the molecules is in competition with the attractive forces. When the attractive forces overpower the thermal agitation, the fluid goes from the gaseous phase to the liquid phase. Although it is condensed, a liquid is still a disordered phase.

In a liquid, each molecule has its specific potential energy, depending on its location with respect to the rest of the fluid; this energy reaches its minimum in the bulk because the forces from the surrounding molecules balance out. By the same token, it is maximum at the surface because of the unbalanced attractive forces of the medium. In that way, one finds that work is required in order to move a molecule from the bulk to the surface. As a result, liquids will tend to minimize this surface energy and, in the absence of external forces, they do so by minimizing the interfacial area and taking on a spherical shape. The surface tension coefficient (γ with units J/m^2 or N/m) is defined as the energy necessary to increase the surface of the fluid by one unit: $dE = \gamma\, dA$, where E is the surface energy and A is the surface area. Typical values of surface tension are given in Table 1.

Table 1
Typical values of surface tension of liquids in air. Adapted from [3]

Liquid	Helium (4K)	Ethanol	Benzene	Water–oil	Water	Mercury
γ (mN/m)	0.1	23	28	50	72	485

In the context of competition between surface tension and gravity, one can define a length scale below which gravity becomes negligible: the capillary length is therefore defined as $L_c = \sqrt{\gamma/(\rho g)}$ where g is the acceleration of gravity and ρ the density difference between the two fluids. Note that in the case of a liquid in air, ρ is approximately the density of the liquid. If the size of a drop is lower than the capillary length, one can neglect gravity and the shape of the drop is determined solely by the capillary forces. For most liquids in air, the capillary length is on the order of a millimeter, with this length taking on larger values in the case of two immiscible liquids. Since the dimensions of microchannels are generally well below the capillary length, the shape of droplets in such channels will not be influenced by gravity but by capillary forces and the confinement of the system.

The surface tension between two media also causes a pressure jump at the interface; this pressure jump is given by Laplace's law $\Delta P = \gamma(1/R_1 + 1/R_2)$, where R_1 and R_2 are the two radii of curvature of the surface. This pressure jump can become very large as the radius of curvature decreases. For example the pressure drop at the interface of a water drop which is 10 nm in radius would be $\delta P = 150$ atm. Therefore one may imagine that the generation of small droplets will have a lower limit, below which it is impossible to reduce the size of drops.

2.3. *Wetting*

Another aspect that is important in microfluidics is the wetting property of liquids on the substrate. Wetting implies the existence of three interfaces between two fluids and a solid surface. The whole system will minimize its energy by adopting the optimal configuration. In the case of a liquid–gas–solid system, one can define the spreading parameter S as $S = \gamma_{\text{sol/gas}} - \gamma_{\text{sol/liq}} - \gamma_{\text{gas/liq}}$ were $\gamma_{\text{sol/liq}}$, $\gamma_{\text{sol/gas}}$ and $\gamma_{\text{gas/liq}}$ are the interfacial tensions of the solid–gas, solid–liquid and gas–liquid interfaces, respectively.

If $S > 0$ the liquid completely wets the surface since this would be the energetically favorable configuration. If $S < 0$, one can define an angle (θ) created by the liquid/solid and liquid/gas interfaces. Knowledge of the spreading parameter determines θ as: $\cos\theta = 1 + S/(\gamma_{\text{liq/gas}})$. A contact angle only exists if the liquid partially wets the surface, i.e. if $-2\gamma_{\text{liq/gas}} < S < 0$. If $S < -2\gamma$, there will not exist a contact line between the liquid and the solid, as discussed in Section 3.

There exists an empirical criterion [4] stating that a solid can be characterized by a critical tension γ_c, such that if the liquid–gas interfacial tension is lower than γ_c, the liquid will completely wet the surface. The value of γ_c is around 30–40 mN/m for most of plastics, which causes them to be hydrophobic but which allows most oils to wet these surfaces.

In the case when the two fluids are liquids, one has to consider the wetting properties of each liquid on the surface. Two cases are possible: either one of the liquids is non-wetting in which case it is separated from the wall by the other liquid, or both liquids are partially wetting and an equilibrium angle is found.

2.4. *Surfactants*

Surfactants are very often used in multiphase flows. A typical surfactant molecule schematically has two parts: a hydrophobic (e.g., aliphatic) tail and an hydrophilic head (for example a polar head). The surfactant molecules will therefore reside mainly on the surface of a fluid, with only their heads immersed in the aqueous phase and their tails outside it. This strongly modifies the surface properties.

A typical usage of surfactants is in the case of oil and water: depending on the structure of the surfactant, it can favor the formation of oil drops in water or the reverse, as well as the intermediate states consisting of large zones of one liquid in the other. The role of these molecules is to reduce the surface tension in the system. Furthermore, the surfactant is often used to stabilize drops in the flow, for example to avoid the coalescence of the droplets in an emulsion.

Finally, we note that there exists a critical concentration of surfactant, known as the Critical Micellar Concentration (CMC). It is the concentration above which surfactant molecules organize themselves into micelles (bilayers, spherical or cylindrical micelles). At concentrations lower than the CMC, the molecules are mostly at the interface of the fluid. Once the surface of the fluid is saturated by the surfactant, the concentration of molecules in the bulk increases allowing them to organize into micelles. The CMC depends on the characteristics of the solvent and is also affected by pH, temperature and ionic strength.

2.5. *Surface treatment*

As the dimensions of the channels shrink, the interactions between the fluids and the solid surfaces become very important. It can be useful to change the critical tension γ_c so as to change the wettability of a fluid on the substrate.

Microfluidic channels are increasingly often made using poly(dimethyl siloxane) (PDMS), a cheap and easily molded material [5]. However, PDMS is hydrophobic which causes some difficulties in pumping water through these channels. However, the surface properties of PDMS can be changed by various techniques, for example by exposing it to UV light or by oxygen plasma treatment [6]. These methods cause chain scissions in the polymer and therefore change the wetting properties of the surface.

It can also be useful to make a surface, such as glass or silicon, hydrophobic. Hydrophobic microchannels can be created by coating the walls with a hydrocarbon chain having a tri-chlorosilane head group, such as, e.g., octadecyltrichlorosilane (OTS). This process is known as silanization, which is described in detail in [7].

Surfactants can also be used as surface agents in order to change the wetting properties. For instance, flowing a fluid with a high concentration of SDS (well above the CMC) makes silicon and glass channels completely hydrophobic; adversely, flowing water containing a high concentration of Span 80, makes the same substrate totally hydrophilic.

3. Flow regimes

The patterns formed by two phase flows in microchannels have attracted a lot of attention. The space of parameters that control these patterns is very large, it involves the nature of the two fluids, the details of the injection, the use of surfactants, as well as the surface characteristics, etc. All this makes the problem difficult to describe in a simple way. However, different groups have looked into the regimes that appear in these flows in the cases of liquid–liquid and liquid–gas systems. Below, we describe some of the important contributions to the question.

3.1. Liquid–liquid

Several authors have noted the importance of the fluid interaction with the walls in the pattern that is formed. In order to generate structures of one fluid (fluid A) within another (fluid B), fluid B must usually completely wet the walls of the microchannel while fluid A must be non-wetting. This fact was noted by Nisisako et al. [8] who showed that it was impossible to generate oil droplets in water in PMMA channels, since PMMA is hydrophobic. Tice et al. [9] described the formation of plugs, defined as droplets that fill the width of the channels but do not wet the walls. These plugs are carried by the flow of the continuous phase which also acts to lubricate their motion along the walls by wrapping them with a thin film. Dreyfus et al. [10] undertook a systematic study of the flow regimes obtained in the case when both fluids partially wet the walls. In this case, the flow regimes were found to be less reproducible. The interface separating the two fluids was found to take an irregular shape which was stationary in time. Droplets were also found to adhere to the channel walls instead of being advected by the mean flow. These observations show the importance of controlling the wetting properties of the fluids on the channels. The control can be made by surface treatment or the use of surfactants, as discussed in the previous section.

We concentrate below on the case where only one fluid wets the walls, for the two types of geometries that have been extensively studied: the T-junction and the cross-junction, shown schematically in Fig. 1.

We first discuss the case when the non-wetting fluid enters through the lateral channels. In this case, drops are formed if the non-wetting fluid is pumped at low flow rates for both geometries. The size and the frequency of the droplet formation depends on the ratio of the flow rates and on the channel size [9], with the drops typically remaining larger than the size of the injection channel. The pinching is due to an interfacial instability taking place at the channel intersection, as described in Section 4.

For higher flow rates of the non-wetting fluid, the interfacial instability is quenched and one observes the formation of stable layers: two in the case of the T-junction channels and three for the cross-junction. This regime is called the stratification regime.

In the specific case of the cross-junction, if the fluid entering through the lateral channels is flowing much faster than the main channel fluid, the shear from the outer fluid layers causes the formation of a thin stream of the inner fluid. This technique was named ‘flow focusing’ by Knight et al. [11].

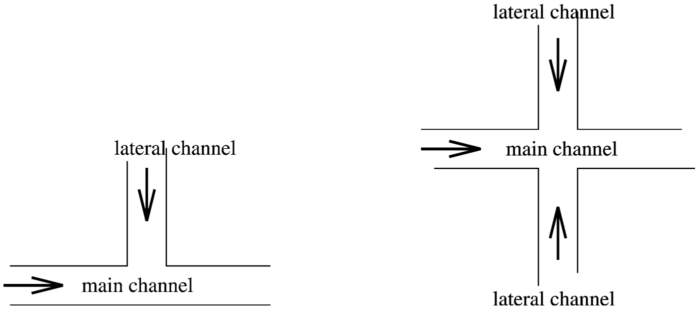

Fig. 1. Junction types: the T-junction on the left, and the cross-junction on the right.

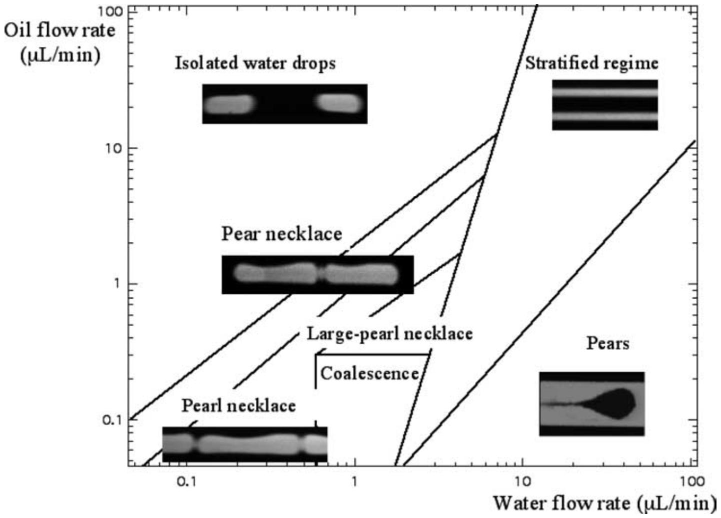

Fig. 2. Phase diagram of flow patterns in a microchannel. Dark regions are the oil phase and light regions are the water phase. From [10].

Finally, for very large flow rate differences between the slow wetting fluid and the fast non-wetting fluid, Dreyfus et al. find the intermittent formation of large drops which pinch off with a low frequency. Note however that now the non-wetting fluid is surrounding the wetting fluid, contrary to other cases described before. A summary of the different flow regimes is shown in Fig. 2.

In the case when the non-wetting fluid is injected in the main channel, the observed regimes differ from the above descriptions and depend more strongly of the geometry of the channels. In the T-junction, we mainly observe the stratified regime. In the cross-junction, at high enough lateral flow rates, one also observes stratification, with the non-wetting fluid stably confined between two layers of the wetting fluid. This regime becomes unstable if the non-wetting stratum becomes thin enough, a Plateau–Rayleigh like instability occurs and droplets of this fluid are advected by the mean flow. Flow focusing is used in generating drops and bubbles, as described below in Section 4.

Finally, we briefly note that similar studies have been conducted on critical liquids: binary mixtures operated near a thermodynamic critical point (see, e.g., [12,13]). Liu et al. [12] looked at the wetting behavior in these fluids, while Tanaka [13] concentrated on the patterns that are formed in capillaries. He found a similar zoology of drops, plugs, stratified, or disordered patterns as in the case of simple liquids.

3.2. Liquid–gas

Liquid–gas systems are gaining in importance as new processes that require a gaseous phase emerge. For instance, reactive liquid–gas systems are of great interest in the fluorination of toluene, an inherently dangerous reaction (see, e.g., de Mas et al. [14]). Running this reaction in a microsystem allows efficient and safe implementation of a process which is difficult on macro-scale. Another application is the cooling of integrated circuits or other MEMS using forced convection, where two-phase flows are promising because of the possibility to use the latent heat absorbed by the working fluid during the phase change from liquid to vapor without increasing its temperature.

As in the liquid–liquid system, it is important to know the different flow regimes that can be obtained in gas–liquid systems. A compilation of various experiments was made by Triplett et al. [15] in the case were the two phases are inert and by Günther et al. [16] in microchemical systems. In both cases, one observes several flow regimes, with observed structures that are similar to the ones in the liquid–liquid systems. For a given liquid flow rate and increasing the gas flow rate: at low gas flow rate, a bubbly regime is obtained, leading to a slug regime as the gas flow rate is increased. For still higher gas flow rates, a slug-annular regime (spatio-temporally varying structures) is observed, followed by a stationary annular state. Intermediate structures such as churns are also observed at high flow rate. In the case of the annular structure, the gas flows inside the liquid. The thresholds of the different transition between the observed regimes depend first on the geometry of the channels and second on the reactivity of the two phases. The main difference between liquid–liquid and gas–liquid two-phase flows is that at high flow rate, the stratified regime is replaced by an annular regime in the liquid–gas system, since the gas cannot wet the boundaries.

For gas–liquid reactions, one typically prefers the slug regime obtained with a low liquid flow rate, while for microheat exchangers the flow rates tend to be much higher, leading to annular or churn regimes.

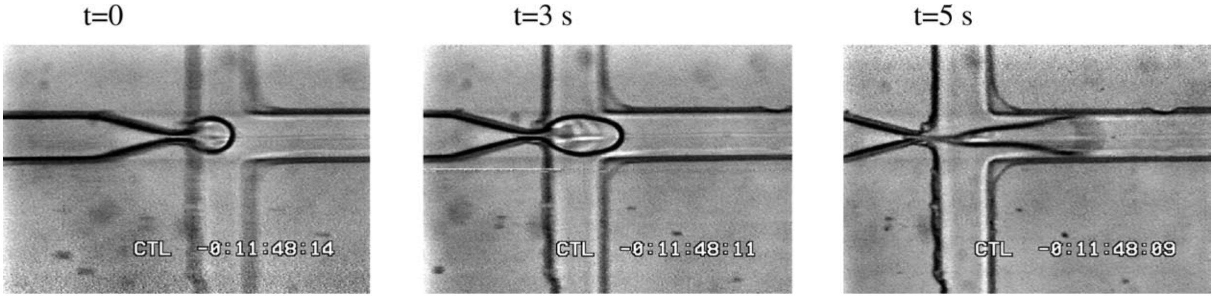

Fig. 3. Formation and detachment of an air bubble in a mean flow. Photo Ch. Baroud.

4. Making drops and bubbles

A major aspect of multiphase flows is the fabrication of microdrops and bubbles. The applications for small, monodisperse, drops range from the pharmaceutical industry to inkjet printing. A microfluidic circuit to fabricate microdrops would also be a useful tool in Micro Total Analysis Systems (µTAS), where these drops can carry reactants or even living cells from one area of the circuit to another. Finally, there is much interest in creating monodisperse colloids and emulsions of microdrops. Before discussing applications of drop making, it is useful to introduce the basic mechanism that breaks a liquid jet into droplets under the action of surface tension: the Rayleigh–Plateau instability.

A detailed review of this instability is given by Eggers [17]. The instability is caused by pressure fluctuations inside the liquid due to small variations in the local curvature; such variations grow to eventually break the liquid film into drops, through a so-called *finite time singularity*. Although the canonical analysis is done for a cylindrical fluid layer, the route to the singularity is quite general since surface tension causes the fluid in most cases to take the cylindrical geometry as it approaches the breaking point. An understanding of the physical mechanisms is therefore useful whenever drop pinching occurs, as shown for example in Fig. 3.

In the case of the simple breakup of a liquid cylinder, one can obtain the wavelength of the *first unstable mode* from simple energetic arguments [17,3]. This wavelength is $\lambda = 2\pi r$, where r is the radius of the unperturbed cylinder. Indeed, the surface energy cost favors drop formation if the drop is large enough to reduce the total area of the interface. Although this is not the *most unstable* mode, experimental measurements show that the two modes are close, so that the perimeter of the jet gives a good approximation for the size of the bubbles. Since the drop size is controlled by the size of the initial jet from which they formed, there has been wide interest in reducing the radius of the jet before allowing it to break.

This technique was used by by Gañán Calvo [18] in a cross-type junction, who showed that a liquid jet surrounded by an air flow will break into monodisperse drops if the Weber number is smaller than a critical value of about 40. In the case of a gas jet inside a liquid flow, Gañán Calvo and Cordillo [19] showed better monodispersity, with bubble sizes ranging between 5 and 120 µm, as shown in Fig. 4.

Also, Anna et al. [20] demonstrated the existence of a wide range of droplet breakup regimes in the ‘focusing’ geometry. They studied the effect of shear by varying the relative flow rates of the two liquids as well as their viscosities. A large array of ordered and disordered patterns were found, as shown in Fig. 5.

In addition to the flow focusing geometry, the T-junction has also been used [21,8,10] to form drops. In this flow, the drop forming fluid is injected slowly from the lateral-channel of the T into a stronger main-channel flow of the surrounding fluid. The shear from the flow forces bubbles to shed from the junction with size depending on the relative flow rates. For very low shear, it is the size of the channels that determines the scale of the ejected drops, as in the case of a drop falling from a pipette.

A variation on the T-junction geometry is shown in Fig. 6. Similar systems to the one above were used for example by Tan et al. [22] and Yi et al. [23]. Both groups demonstrate the ability to produce drops of a few microns, down to a fraction of a micron, as the main-channel flow rate, hence the local shear, is increased. For a discussion of the relation between viscous shear and drop size (in a macroscopic system), see [24].

5. Other applications

We list below two applications that we find important, although the list of applications is very long and very varied.

An interesting route to follow is the use of surface tension modulation to pump fluids through the effect of Marangoni convection. The difficulty of manufacturing micropumps is well known, due to the reversible aspect of low Reynolds number flows. Therefore, any reciprocating motion will lead to zero net flux. By modulating the surface tension on an air bubble, Debar

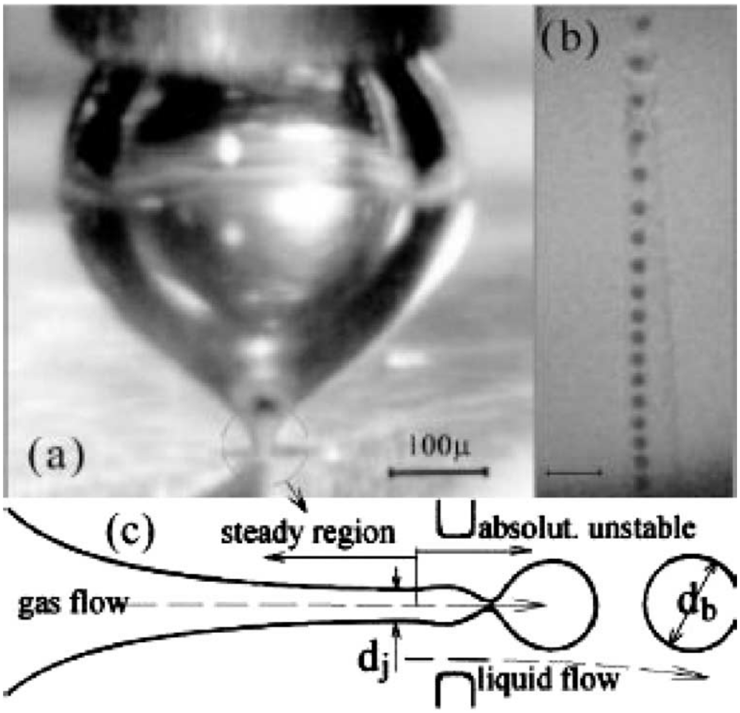

Fig. 4. Formation of gas bubbles in a liquid jet. From [19].

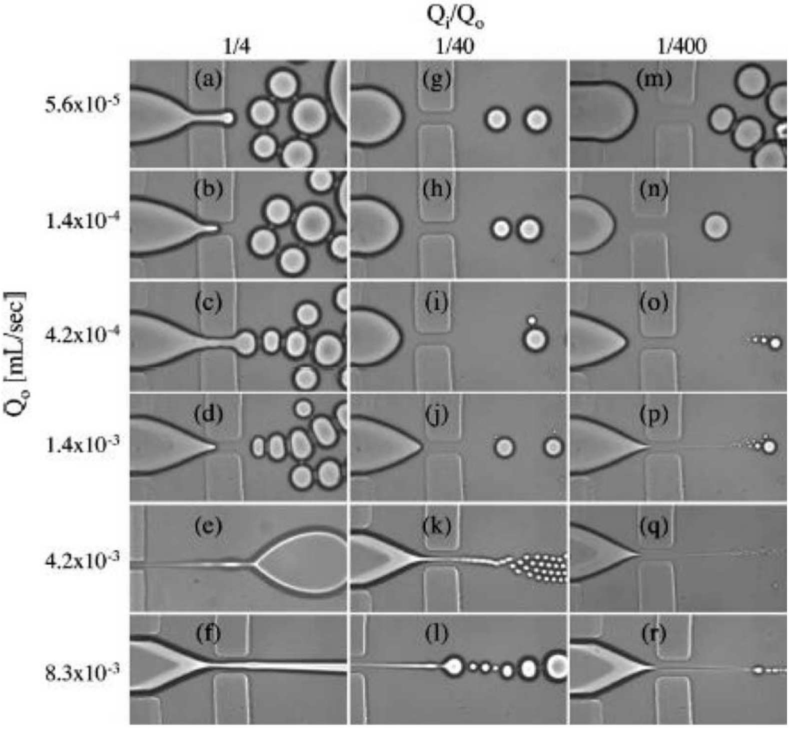

Fig. 5. Regimes of droplet formation by ‘flow focusing’. From [20].

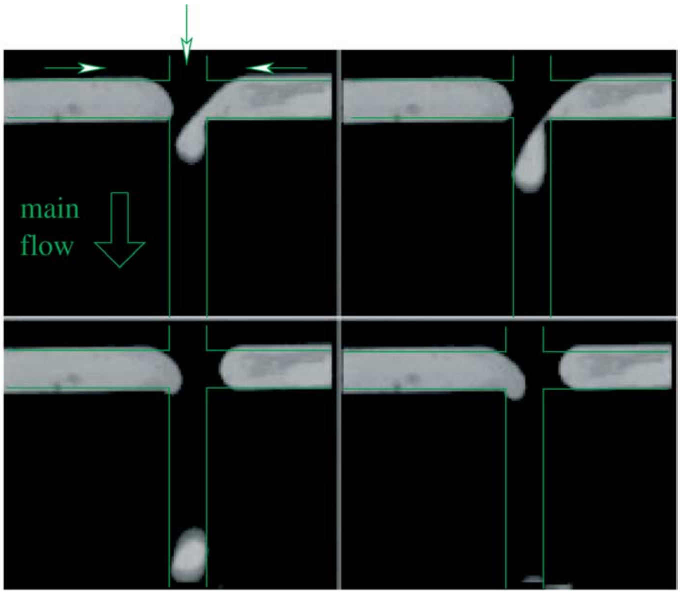

Fig. 6. Forming drops by using the imposed shear from a side-flow. From [10].

and Liepmann have demonstrated that a Marangoni flow is set up at the interface of the bubble, causing a net flux everywhere in the channel [25]. Similar ideas are being explored in open-drop microfluidics (see, e.g., [26]). Finally, an asymmetry in the growth and collapse of a heated gas bubble could be used to pump the surrounding liquid through a microchannel, as suggested through numerical simulations by Ory et al. [27].

Inkjet printing is one of the most important applications of MEMS and a good example of multiphase flow application. Here, the microsystem is used to inject controlled droplet of ink, typically around 50 microns in diameter [28]. This ejection is caused by heating and vaporising a small volume of ink, thus increasing its volume and inducing a liquid jet that is destabilized by capillary forces. Similar injection technology is now being used in different applications such as the deposition of biochemical products on microsystems. It is obvious that in biological application (e.g., DNA chips) the minimization of the volume of each droplet is of great importance and much research is being carried out in this direction.

The study of multiphase flows continues to occupy an important place in microfluidics, both because of the wide range of possible applications and for the understanding of fundamental fluid processes. We have discussed some of the important aspects of these complex fluid flows, their mutual interactions, and interactions with the channel boundaries. The flow regimes vary significantly depending on the many parameters such as wetting properties or flow rates. Furthermore, different techniques of forming droplets and gas bubbles by using the geometry of microchannels were discussed. The list of potential applications is very large and promises to keep growing, thus motivating further research in the field.

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