Available online at www.sciencedirect.com
ScienceDirect
Sensors and Actuators B 117 (2006) 431–436
SENSORS AND ACTUATORS B CHEMICAL
www.elsevier.com/locate/snb

Optical detection for droplet size control in microfluidic droplet-based analysis systems

Nam-Trung Nguyen*, Sumantri Lassemono, Franck Alexis Chollet

School of Mechanical and Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

Received 11 May 2005; accepted 2 December 2005
Available online 18 January 2006

Abstract

This paper reports on a hybrid polymeric microfluidic device with optical detection for droplet-based systems. The optical part of the device is integrated by a hybrid concept. The microfluidic structures were fabricated using CO_2 laser on poly methylmethacrylate (PMMA) substrate. The microfluidic network consists of two microchannels for forming droplets of an aqueous liquid in an immiscible carrier liquid. The optical component consists of two optical fibers for guiding laser light from the source, through the detection point, to a photo diode. The formed droplets pass the detection point and diffract the incoming laser light. The detected signal at the photo diode can be used for evaluating droplet size, droplet shape, and droplet formation frequency. The device can detect very high formation frequencies, which are not detectable using a conventional CCD camera/microscope setup.

Keywords: Droplet microfluidics; Polymeric micromachining; Lab on chip; Optical detection

1. Introduction

Droplet-based microfluidics has been emerging in the recent years because of its potential and apparent advantages. One of the key advantages of this concept is the small sample volume on the order of picoliters and nanoliters. A number of concepts such as thermocapillary [1], electrowetting [2,3], or multi-phase flow [4] can be used for generating and controlling of droplets. Droplet formation based on multi-phase flow is easy to implement in a continuous-flow system. In this case, the fluidic system consists of two immiscible phases such as an aqueous liquid and an oil. The balance between the shear stress of the carrier flow and the interfacial tension between the two liquid phases leads to the formation of droplets [5].

Systems generating micro-droplets have been successfully used as microreactors for chemical analysis and protein crystallization [4]. Furthermore, dispersed droplets of one liquid in a second liquid can form an emulsion, which have many applications in food industries and cosmetic industries. Emulsion is important for packaging small amounts of fluid and other active ingredients such as drugs. Encapsulation of nanoliter droplets can also be achieved with a double emulsion. An intermediate fluid layer works as an additional barrier between the inner fluid and the carrier fluid. Recent interest of the research community on double emulsion in micro-scale shows its potential significance [6,7]. Since the droplet's size and its other properties are important for the actual application, a detection system for the micro-droplets is vital for providing a feedback signal to the droplet formation process.

Most of the recent works on micro-droplets only report devices made of polydimethylsiloxane (PDMS) and glass using external syringe pumps. A microscope and a CCD-camera are usually used for characterization of the droplets. The whole setup is rather bulky and the collected data are difficult to analyze automatically. Furthermore, expensive high-speed camera and synchronized strobe illumination are needed for capturing processes with droplet frequencies on the order of tens Hertz and above.

In this paper, we present a poly methylmethacrylate (PMMA) device for droplet formation. The device has a hybrid-integrated optical system for evaluating parameters of the droplet formation process such as the formation frequency, droplet size, and contact angles of the receding and advancing edges. Very high formation frequency can be detected by this device. The paper

* Corresponding author. Tel.: +65 67904457; fax: +65 67911859.
E-mail address: mntnguyen@ntu.edu.sg (N.-T. Nguyen).

0925-4005/$ – see front matter
doi:10.1016/j.snb.2005.12.010

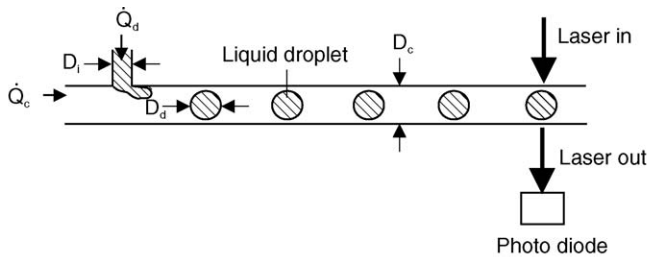

Fig. 1. Concept of formation and detection of liquid droplets in a microchannel.

first discusses a simple theory on droplet formation to identify the key parameters of this process. Next, the fabrication of the device and experimental results are presented and compared with the theory.

2. Formation and optical detection of micro-droplets

Fig. 1 depicts a simple model of the formation process of a liquid droplet in another immiscible carrier fluid. The following model only serves the purpose of understanding the relations between key parameters such as droplet size, formation frequency, flow rates, and most importantly the interfacial tension between the two liquid phases. The model assumes a fixed flow rate ratio between the aqueous liquid and carrier liquid ($\alpha = \dot{Q}_{\rm d}/\dot{Q}_{\rm c}$). We further assume that the droplet size is small ($\alpha \ll 1$). Since the droplets are formed in micro-scale and the flows are in steady state, mass related forces such as inertial force, momentum force and buoyancy force are neglected in this model. If the aqueous liquid contents a surfactant, the surfactant concentration at the droplet surface is not uniformly distributed during the process of droplet growth. The distributed surfactant concentration leads to a gradient of interfacial tension on the droplet surface. This interfacial tension gradient in turn induces a Marangoni force on the droplet. If the surfactant solution is diluted, the Marangoni force is assumed to be small and negligible. The injection channel and the carrier channel are both assumed to be cylindrical.

Considering all the above assumptions, the force balance includes only the drag force of the carrier flow and the interfacial tension at the injection port:

$$\begin{aligned} F_{\rm drag} &= F_{\rm interfacial\ tension} \\ \tfrac{1}{2} C_{\rm D} \rho U_{\rm c}^2 A_{\rm D} &= C_{\rm S} \pi D_{\rm i} \sigma \end{aligned} \quad (1)$$

where $\rho_{\rm c}, U_{\rm c}, A_{\rm D}, D_{\rm i}$, and σ are the density of the carrier fluid, the average velocity of the carrier flow, the effective drag surface, the diameter of the injection port, and the interfacial tension, respectively. In addition, $C_{\rm D}$ and $C_{\rm S}$ are the drag coefficient and the coefficient for the interfacial tension. The coefficient $C_{\rm S}$ depends on the contact angle and the shape of the injection port. In this model $C_{\rm S}$ is assumed to be constant. We assume for $C_{\rm D}$ the drag coefficient of a hard sphere at a low Reynolds number *Re*:

$$C_{\rm D} = \frac{24}{Re}. \quad (2)$$

The effective drag interfacial $A_{\rm D}$ grows with the droplet. Assuming that the droplet is a sphere, the effective drag surface at the detachment moment is:

$$A_{\rm D} = \frac{\pi D_{\rm d}^2}{2} \quad (3)$$

where $D_{\rm d}$ is the diameter of the generated droplet. Initially, the interfacial tension is large enough to keep the small droplet at the injection port. At the detachment moment, the continuous droplet growth makes the drag force large enough to release the droplet. Substituting (3) into (1) results in the droplet diameter:

$$D_{\rm d} = 2\sqrt{\frac{C_{\rm S}}{C_{\rm D}} D_{\rm i} \frac{\sigma}{\rho_{\rm c} U_{\rm c}^2}} \quad (4)$$

The formation frequency can be estimated from the flow rate of the aqueous liquid $\dot{Q}_{\rm d}$ and the droplet volume $V_{\rm d}$ as:

$$f = \frac{\dot{Q}_{\rm d}}{V_{\rm d}} \quad (5)$$

Using the droplet diameter $D_{\rm d}$ and the relation $\dot{Q}_{\rm d} = \alpha \dot{Q}_{\rm c}$, the formation frequency in (5) can be expressed as:

$$f = \frac{3\alpha D_c^2}{16((C_S/C_D)D_{\rm i})^{\frac{3}{2}}} \frac{\rho_c^{\frac{3}{2}} U_c^4}{\sigma^{\frac{3}{4}}} \quad (6)$$

where $D_{\rm c}$ is the diameter of the carrier channel. Eq. (6) shows a nonlinear relation between the formation frequency and the average carrier's velocity ($f \propto U_{\rm c}^4$) or flow rate ($f \propto \dot{Q}_{\rm c}^4$). Furthermore, if the Marangoni force is considered in Eq. (1), and if the surfactant concentration is high, there will be an additional term for such force in the numerator of (6), resulting in a higher formation frequency.

Our droplet-based microfluidic device consists of two parts: a microchannel system for droplet formation and an optical detection system, Fig. 1. The microfluidic network consists of a large carrier channel and a small injection channel. The aqueous liquid enters through the injection channel, while an immiscible carrier liquid is introduced into the carrier channel. The two channels form a T-junction, at which droplet formation occurs. After droplets are formed and stabilized, they can be detected at a downstream position. The detection system is based on the optical concept. Laser light is guided into the microchannel by an optical fiber. After passing through the microchannel, the light is received at the other side by a second optical fiber which leads the laser to an optical sensor. The passing-by droplets change the intensity of the light due to diffraction and absorption. Thus the size and shape of the droplet can be well recorded as the time signal of the optical sensor.

On the receiving side of the microchannel, the laser light diffracted by the propagation and by the different interfaces must enter the fiber within a fixed angle. This angle depends on the optical fiber's numerical aperture (NA) and the refractive indices of the fiber's core and cladding. In our later experiments, the optical fiber had a numerical aperture of 0.22. Theoretically, the maximum angle of the incident light in our device is 12.7°.