Journal of Colloid and Interface Science 466 (2016) 162–167

Contents lists available at ScienceDirect

Journal of Colloid and Interface Science

journal homepage: www.elsevier.com/locate/jcis

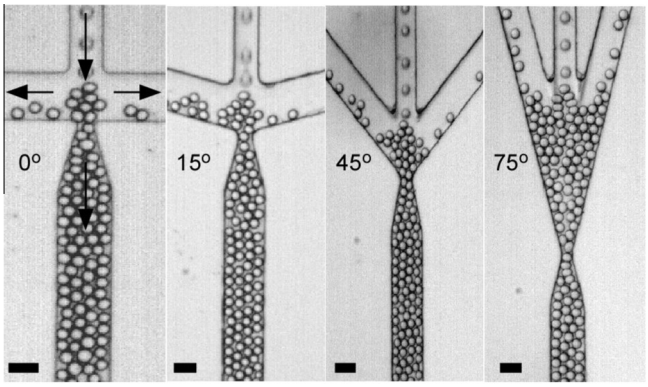
Integrated microfluidic system with simultaneous emulsion generation and concentration

Karuna S. Koppula [a,1], Rong Fan [b,1], Kartik R. Veerapalli [b], Jiandi Wan [b,*]

[a] Department of Chemical Engineering, Rochester Institute of Technology, Rochester, NY 14623, USA
[b] Microsystems Engineering, Rochester Institute of Technology, Rochester, NY 14623, USA

GRAPHICAL ABSTRACT

An efficient microfluidic emulsion generation system integrated with an orifice structure has been presented to separate aqueous droplets from the continuous oil phase. The efficiency of separation is determined by a balance between pressure drop and droplet accumulation near the orifice. Scale bar: 60 μm.



ARTICLE INFO

Article history:
Received 29 October 2015
Revised 11 December 2015
Accepted 17 December 2015
Available online 18 December 2015

Keywords:
Microfluidics
Emulsion drops
Emulsion concentration and separation

ABSTRACT

Because the size, size distribution, and concentration of emulsions play an important role in most of the applications, controlled emulsion generation and effective concentration are of great interest in fundamental and applied studies. While microfluidics has been demonstrated to be able to produce emulsion drops with controlled size, size distribution, and hierarchical structures, progress of controlled generation of concentrated emulsions is limited. Here, we present an effective microfluidic emulsion generation system integrated with an orifice structure to separate aqueous droplets from the continuous oil phase, resulting in concentrated emulsion drops in situ. Both experimental and simulation results show that the efficiency of separation is determined by a balance between pressure drop and droplet accumulation near the orifice. By manipulating this balance via changing flow rates and microfluidic geometry, we can achieve monodisperse droplets on chip that have a concentration as high as 80,000 drops per microliter (volume fraction of 66%). The present approach thus provides insights to the design of microfluidic device that can be used to concentrate emulsions (drops and bubbles), colloidal particles (drug delivery polymer particles), and biological particles (cells and bacteria) when volume fractions as high as 66% are necessary.

* Corresponding author.
E-mail address: jdween@rit.edu (J. Wan).
[1] Karuna S. Koppula and Rong Fan contributed equally to this work.

http://dx.doi.org/10.1016/j.jcis.2015.12.032
0021-9797/

1. Introduction

Microfluidics has been demonstrated as an effective platform for generation of emulsion droplets [1,2] and plays an important role in manufacture of drugs, food products and synthetic materials in the form of microparticles or microbeads, microgels, and microcapsules [3–5]. Recent advances in microfluidics have yielded unprecedented control over the generation of emulsions and we can now manipulate the producing rate, size distribution, and hierarchical structures of emulsion droplets [6–9]. An ultra-high producing rate of emulsion drops over 1 kg/day, for example, has been achieved using microfluidics [10,11]. Emulsions with hierarchical structures including double emulsion [10], triple emulsion [12], and multicomponent multiple emulsion [13] can also be produced via microfluidic approaches.

The majority of microfluidic approaches, however, produce emulsion drops with a low concentration. This is because, in order to generate droplets with a small diameter (~a few to tens of micrometers), the volumetric flow rate of the continuous phase is usually higher than that of the disperse phase [14], resulting in a low volume fraction of drops in the collection reservoir. As a consequence, significant amount of fluids of the continuous phase is required to produce a relatively large amount of emulsion drops. The large consumption of continuous fluids and low concentration of emulsion drops significantly limit its applications in many fields in which large-scale production of drops is preferred [15]. Strategies that can reduce the amount of continuous fluids and concentrate emulsion drops are thus of great interest. Filtering can separate droplets from the continuous fluids but droplets either will break and/or stick to the filter paper during filtering, resulting in a low harvesting yield. In addition, off-line droplet collection and handling may cause contamination and increased emulsion instability, which are undesired for most of the droplet applications. Active sorting of microfluidic-generated droplets, on the other hand, applies external electric [16], dielectrophoretic [17], or magnetic forces [18] to manipulate droplets individually in desired directions and thus have been demonstrated to be able to achieve high volume fractions of droplets. The need of fabrication of complex interfacing components such as electrodes, piezoelectric and mechanical parts in microfluidics, however, limits its application. Although alternative approaches using different geometry of microchannel [19–21] and surface wetting patterns [22] have been developed to manipulate and sort droplets, most of them rely on the complex geometry design and are used for size-dependent droplet separation. Effective strategies to concentrate emulsion drops still remain to be explored.

Here, we demonstrate a simple yet unique microfluidic device with built in facility for simultaneous generation and concentration of uniform sized aqueous droplets in a continuous oil phase. In particular, a flow-focusing design is used to generate droplets whereas a downstream V-shape junction is used to concentrate droplets. The developed microfluidic platform with simple geometry design enables direct size-dependent separation of droplets from the continuous stream, which avoids the using of external forces and the need of extra off-chip separating step. In addition, the integrated microfluidic system is able to simultaneously generate and concentrate monodisperse droplets with a concentration as high as 80,000 drops per microliter (volume fraction of 66%, drop sizes of ~20 μm) at a relatively low shear stress, i.e., ~12.5 Pa. The demonstrated microfluidic system thus provides a potential platform in which emulsion drops, particles or biological cells can be separated and concentrated in an effective manner, which are advantageous for applications in chemical, biomedical, and pharmaceutical engineering.

2. Materials and methods

2.1. Device fabrication and experimental setup

PDMS microfluidics is fabricated using the standard soft lithography technique. The setup for the generation of water-in-oil emulsion drops includes a flow focusing junction continuously generating aqueous droplets in a Novec@ HFE 7500 oil phase (3-ethoxy-1,1,1,2,3,4,4,5,5,6,6,6-dodecafluoro-2-trifluoromethyl-hexane, 99%, 3 M) with 2 wt% Krytox 157FSL (Fluorocarbon surfactant with 21.6% carbon, 9.4% oxygen and 69.0% fluorine, DuPont), as shown in Fig. 1A. Deionized water (18 MΩ cm, 100%) is used as the aqueous phase. The width of the channel for aqueous and oil phase is 100 μm and 150 μm, respectively; the width of the central channel where aqueous droplets are dispersed into the oil phase is 60 μm. The height of the channel is equal to 30 μm everywhere. The width of the constriction of the orifice in outlet 3 is 30 μm. The orifice at the entrance of outlet3 has thus a square section (30 × 30 μm). In the experiment, the aqueous and oil phases are loaded into two different syringes connected to syringe pumps (NE 300) and transported from the syringes to the microfluidic devices using polyethylene tubing (PE 20) connected from the syringe needles to the inlet holes of the device. The interfacial tension between water and HFE 7500 oil phase in the presence of Krytox surfactant is approximately 1–5 mM/m [6]. Because the same aqueous and oil phases are used to generate drops, the interfacial tension of drops is the same. In addition, the fluorinated oil used in the experiment wets the PDMS wall [23] and the interfacial tension between the water and oil phase in the presence of Krytox surfactant is low, surface energy has thus a minor impact on the drop concentration. The generated drops are stable throughout the experiment and during droplet collection, which takes approximately 3–4 h. The aqueous and oil phases have the density of 1 kg/m^3 and 1614 kg/m^3, respectively. Water-in-oil emulsion drops are used for all the experiments. The typical Reynolds number in our experiments is from 15 to 78.

2.2. Image analysis

Generation of droplets is observed using a high-speed video camera (Phantom, Micro M110) mounted on a microscope (Leica). The exit velocity of the droplets and their respective diameters are analyzed using an image analysis program written in Matlab. An algorithm is used to track and count the number of droplets entering the central channel and track the exit path they take in each given device. It uses distance and Hough transforms with image morphological operations to track each droplet's path over a set of frames in the given video of the process. As the accuracy of this algorithm is highly dependent on the video quality of the process, wavelet based de-noising is performed on each of the video frames.

3. Results and discussion

An illustration of the integrated V-junction microfluidic device is shown in Fig. 1A. Aqueous droplets are generated in a continuous oil phase using a flow-focusing design and exit through three outlets, namely, outlet 1, 2 and 3. In a typical experimental setup, it has been observed that the majority of the dispersed droplets exit through outlet 3 while the continuous oil phase exits through outlets 1 and 2 (Fig. 1B), resulting in an effective separation of the emulsion droplets from the continuous phase. This observation of passive hydrodynamic separation of emulsion drops triggers the mechanistic investigation of the effect of flow parameters and design factors on the separation. In particular, the effects of (i) oil-to-water flow rate ratio (Q_o/Q_w), and (ii) angles of orientation