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Development of Microchannel Emulsification Technology for Monodispersed Soybean and Olive Oil-in-Water Emulsions

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Abstract: Emulsions are widely used in food industry, among others. Monosized droplets in emulsions have advantages for control of their properties, and application fields. We have developed microchannel emulsification (MC) technology for monosized droplets formation. The grooved type MC and module were used to characterize MC emulsification, and straight-through MC and module were developed for higher productivity. Both monodisperse oil-in-water and water-in-oil emulsions with average droplets size ranging 2-200 μm and coefficients of variation (CV) lower than 10% were successfully obtained. Emulsification was significantly affected by MC size and structure. A straight–through MC was highly applicable to emulsification. MC emulsification can also be applied to production of solid-lipid and polymeric microparticles, as well as microcapsules for food and bioprocesses.

Keywords: Microchannel emulsification, Monodisperse droplets, Olive oil, Soybean oil

1. Introduction

Recent advances in micro and nanotechnology have enabled their application to food industry, while processing raw materials into valuable products. This may contribute to better nutrition and human health, meanwhile more sustainable processes can be forecasted. Amongst these technologies, emulsification by precisely manufactured microchannels has been developed most recently.

Emulsions are commonly produced using conventional emulsification devices, which apply mechanical shear force to disrupt the dispersed phase into smaller droplets, resulting in emulsions of considerable polydispersity, with a typical C.V. over 20% (McClements, 2004). Nakashima *et al.* (1991) proposed direct production of quasi-monodisperse emulsions with a minimum C.V. of 10% using a microporous membrane. Meanwhile, it is difficult to produce highly monodisperse emulsions owing to the pore size distribution of the membranes used.

Employing microfabrication technology, we have recently developed microchannel (MC) emulsification technology for monosized oil droplets formulation (Kobayashi et al., 2005a, 2005b, 2008).

Microchannels consist of grooves and/or through-holes ranging between 1-1000 μm in size, microfabricated on a chip, as depicted in **Figure 1**.

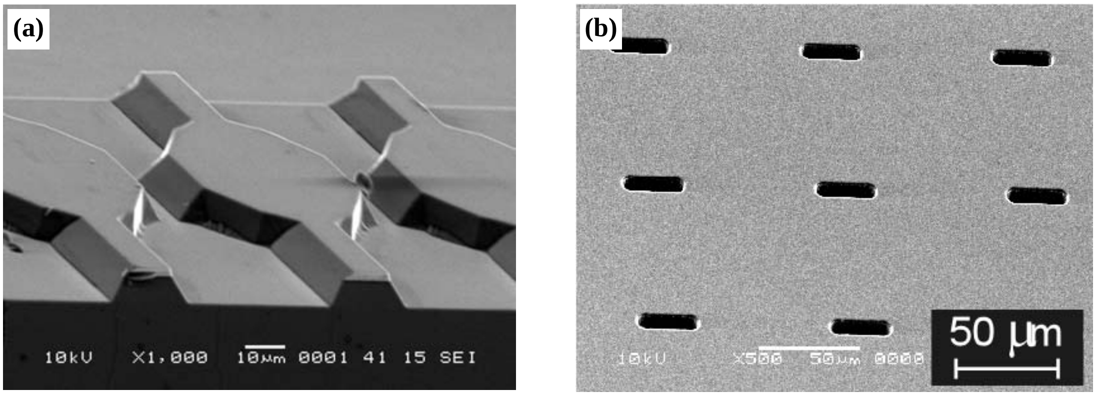

Fig. 1. Scanning electron microscopy (SEM) images of silicon arrays of microchannels: (a) grooved MC; (b) oblong straight-through MC.

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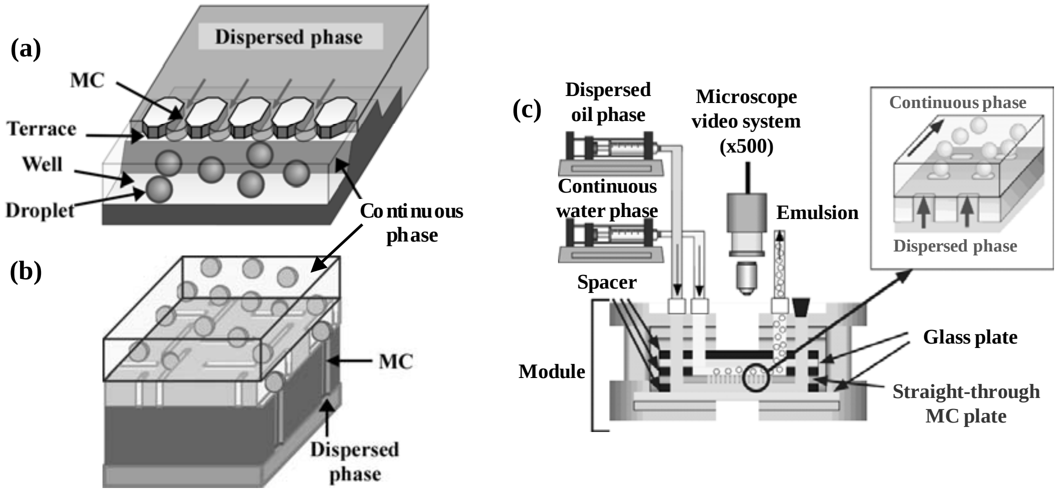

Fig. 2. Schematic illustration of droplet generation: (a) grooved MC; (b) asymmetric straight-through MC; (c) typical experimental setup image for straight-through MC emulsification.

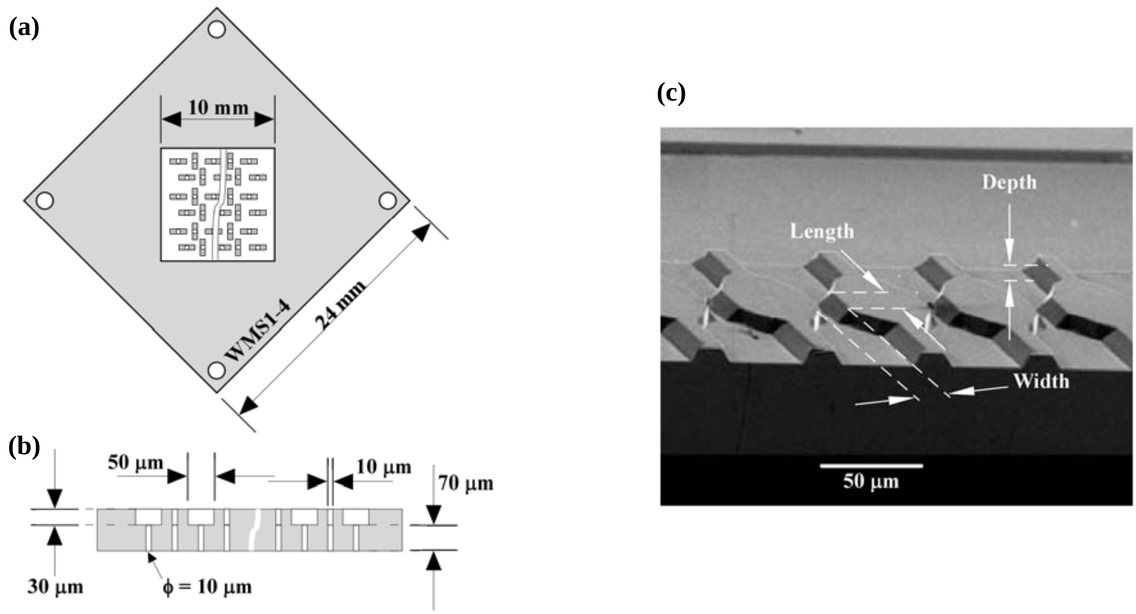

Fig. 3. Schematic illustration of MC: (a) Top view of alternate horizontal-vertical asymmetric straight-through MC plate WMS 1-4; (b) Cross-sectional view of MC WMS 1-4; (c) SEM micrograph of grooved MC plate MS 308, depicting its dimensions.

For most of emulsification technologies using microdevices, the dispersed phase that passed through the channel is directly breakup into droplets. Microchannels with precisely controlled size and size distribution are useful for generation of monodisperse emulsion droplets with a desired size.

In this paper, we review emulsification technologies using microchannels, and show the results of production of emulsions using soybean and olive oils as oil phase.

2. Materials and Methods

The silicon-based grooved type MC was used to characterize MC emulsification, and straight-through MC was also used for high throughput. Soybean and olive oils were tested as dispersed oil phase, in separate. A typical experimental setup for MC emulsification is depicted in **Figure 2(c)**. This emulsification setup consists of an MC plate, a module, apparatuses for supplying the two phases, and a microscope video system. The MC plate is fixed into the module, which has been initially filled with the continuous phase. The pressurized dispersed phase reaches the channel entrance, and then is pushed out into the continuous phase via the channels, to generate emulsion droplets, represented schematically in **Figure 2(a)** for grooved MC, and **Figure 2(b)** for asymmetric straight-through (AST) MC.

The AST MC plate WMS 1-4 has the dimensions 24 mm × 24 mm × 0.5 mm, and consists of about 23,400 MC placed within an area of 10 × 10 mm^2 located at the centre of the plate, as depicted in **Figure 3(a)**. The circular channels have the dimensions detailed in (**Fig. 3(b)**). Due to the oxidation treatment, performed using an oxygen plasma reactor, MC plate surface is hydrophilic, enabling the formulation of O/W emulsions. The grooved MC plate MS 308 has the dimensions 8 mm × 22.5 mm × 0.5 mm, and consists of a microfluidic channel array of uniform size, comprising about 600 channels, which have 10 μm in width, 70 μm in length and 2 μm in depth. Details on the grooved MC dimensions are depicted in **Fig. 3(c)**.

3. Results and Discussion

Using the grooved type MC, successful emulsification was achieved when soybean oil was used as dispersed phase, as shown in **Figure 4**. Emulsification behavior was significantly affected by MC size and structure. Oblong straight–through MC and modified straight–through MC, which were fabricated by deep-reactive-ion etching, were highly applicable to emulsification. Both monosized soybean and olive oil (**Fig. 6**) droplets were obtained. The aspect ratio of the oblong channels considerably affects the droplet generation process in straight-through MC emulsification.

Continuous outflow of the dispersed phase from the channel exit was observed using the channels with a small aspect ratio of 1.9. On the other hand, monodisperse emulsion droplets were stably generated from the channels with aspect ratio of 3.8. The studies in straight-through MC emulsification revealed that monodisperse emulsions were successfully produced using the oblong straight-through MCs with a threshold aspect ratio of approximately three (Kobayashi *et al.*, 2004a).

The droplet diameter was independent of the applied flow rate of the continuous phase. The interfacial-tension-based droplet generation, proposed in grooved MC emulsification, is applicable to successful droplet generation from the oblong channels. Droplets size could be controlled ranging 2-200 μm, and coefficients of variation lower than 10% were successfully obtained.

The oil droplets generation process using the straight-through MCs was also analyzed using the computational fluid dynamics (CFD) method (Kobayashi *et al.*, 2004b). Using the channels over a threshold aspect ratio of 3-3.5, droplets formation was obtained, which was confirmed by both experimental and simulation results (**Fig. 5**).

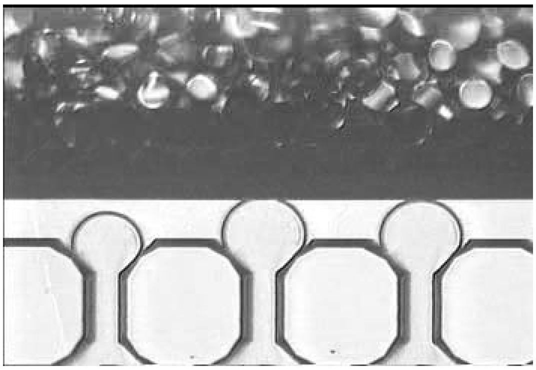

Fig. 4. Microscopic image of emulsification behavior from a grooved MC; regular droplet generation at low dispersed phase flux.

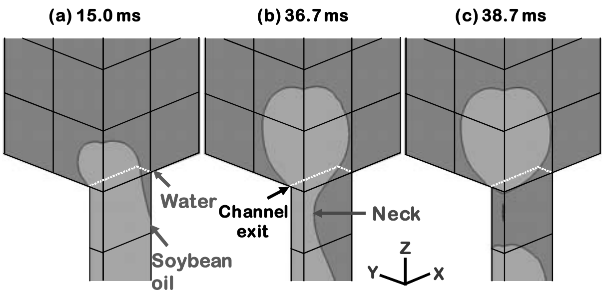

Fig. 5. Emulsification behavior using straight-through MC, calculated by computational fluid dynamics. Successful formulation of a droplet with a diameter of about 35 μm from an elliptic straight-through MC (longer line, 40 μm; shorter line, 10 μm).

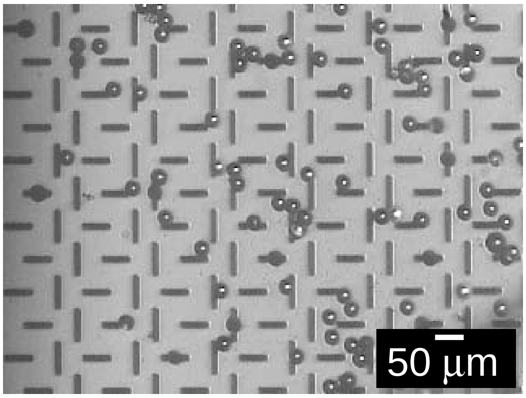

Fig. 6. Microscopic image of emulsification behavior: Olive oil was used as dispersed phase.

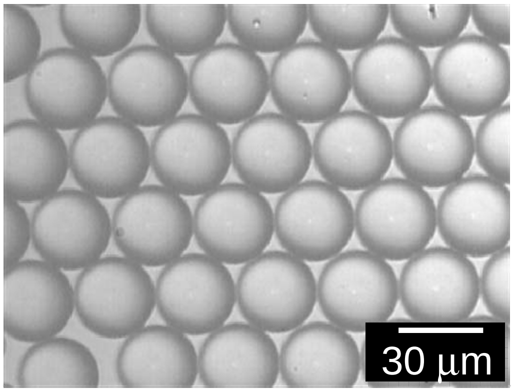

Fig. 7. Microscopic image of olive oil droplets obtained by MC emulsification.

The diameter of the droplets generated by MC emulsification is primarily controlled by the Figure 6 depicts the emulsification behavior using olive oil as dispersed phase, by AST MC, and **Figure 7** shows a magnification of the monodispersed olive oil droplets generated. The resultant droplet diameter was independent of the applied flow rates of the continuous phase. The interfacial-tension-based droplet generation has been proposed in grooved MC emulsification (Sugiura *et al.*, 2001).

The diameter of the droplets generated by MC emulsification is primarily controlled by the channel geometry. The resultant droplet diameter was greatly influenced by the channel depth, and was significantly influenced by the terrace length (Sugiura *et al.*, 2002a), but being independent of the channel width and channel length (Sugiura *et al.*, 2002b). The droplet diameter of the resultant monodisperse emulsions was almost constant below the critical flow rate of the dispersed phase in the channel, which is proportional to the capillary number (Ca [-]) (Sugiura *et al.*, 2002c). Ca is defined as the ratio of the viscous force to the interfacial tension force. The critical Ca was also independent of the channel size and the interfacial tension. Above the critical Ca, a drastic increase in the droplet diameter was observed.

Besides the silicon MCs, stainless steel MCs and acrylic MCs were also used for production of monodisperse O/W and W/O emulsions, respectively. Monodisperse emulsion droplets generated by MC emulsification have been applied to monodisperse lipid microparticles, polymeric microparticles, natural polymeric microbeads, coacervate microcapsules, and W/O/W emulsions. The formulation of microbubble by MC emulsification has been investigated as well.

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