PRODUCTION OF UNIFORMLY SIZED EMULSION DROPS AT HIGH PRODUCTION RATES USING ASYMMETRIC MICROCHANNEL PLATES

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Keywords: microchannel emulsification; deep reactive ion etching; single crystal silicon; monodisperse drops; microchannel array

Abstract

The purpose of this work was to investigate maximum disperse phase flux required for production of monodisperse drops in a straight-through microchannel (MC) array device. The experiments have been carried out using single-crystal silicon MC plate consisting of about 23,000 asymmetric MCs fabricated by photolithography and deep reactive ion etching (DRIE). Each MC consisted of a rectangular 50×10 μm slot and a circular 10 μm-diameter hole in the middle of each slot. A depth of the hole was 70 μm and the slot depth was 30 μm. The dispersed phase was soybean oil, MCT (middle-chain fatty acid triglyceride) oil

and n-tetradecane with a viscosity at 293 K of 50, 20, and 2.7 mPa·s, respectively. The continuous phase was 2 wt% Tween 20 or SDS.

The maximum oil flux permitted production of monodispersed drops increased with decreasing the oil viscosity and ranged between 120 $Lm^{-2}h^{-1}$ for soybean oil and 2700 $Lm^{-2}h^{-1}$ for n-tetradecane. The maximum drop production rate for n-tetradecane was 250 drops/s per channel and nearly 3 million drops/s per plate at the proportion of active MCs of 50%. A span of particle size distribution of resultant emulsions was below 0.23 for soybean oil and MCT oil and below 0.3 for n-tetradecane. The experimental results were found to be in a very good correlation with CFD predictions.

1. Introduction

Traditional microfluidic drop generators such as flow focusing devices (1) and T-junctions (2) can generate monodispersed drops with a coefficient of variation of less than 3 %. Although the drop generation rate for water-in-oil emulsions can be as high as 12,000 Hz (3), the flow rate of dispersed phase is very limited because the drops are formed from a single microchannel (MC). Much higher dispersed phase flow rates can be achieved using MC array devices where the droplets are simultaneously formed from thousands of MCs. Single-crystal silicon MC arrays can be microfabricated onto the plate surface as open microgrooves (4) or normal to the plate surface as straight-through holes (5). Grooved MC plates for blood rheology measurements have been developed by Kikuchi et al. (6) and optimized for production of emulsions by Kawakatsu et al. (4). Grooved MC plates usually have drop productivity of less than 0.1 mLh^{-1} for food grade oil-in-water emulsions, due to limited number of MCs per plate (100-1500). In contrast, straight-through MC plates with as much as 2×10^5 MCs and a throughput of 20-30 mLh^{-1} have been developed and tested (7). Straight-through MC plates can be fabricated with a symmetric or asymmetric structure. Symmetric MCs mean that MCs are of the same geometry (circular or slotted) on both sides of the plate, while asymmetric channels have different geometry along the cross section (e.g. circular holes at the bottom side of the plate and slotted channels at the top size). Slotted MCs can provide better performance than circular MCs and an aspect ratio of the slots should be greater than 3 to ensure production of highly uniform droplets (8). The aim of this work was to investigate experimentally and using CFD analysis the limiting flux in asymmetric straight-through MC plates.

2. Experimental

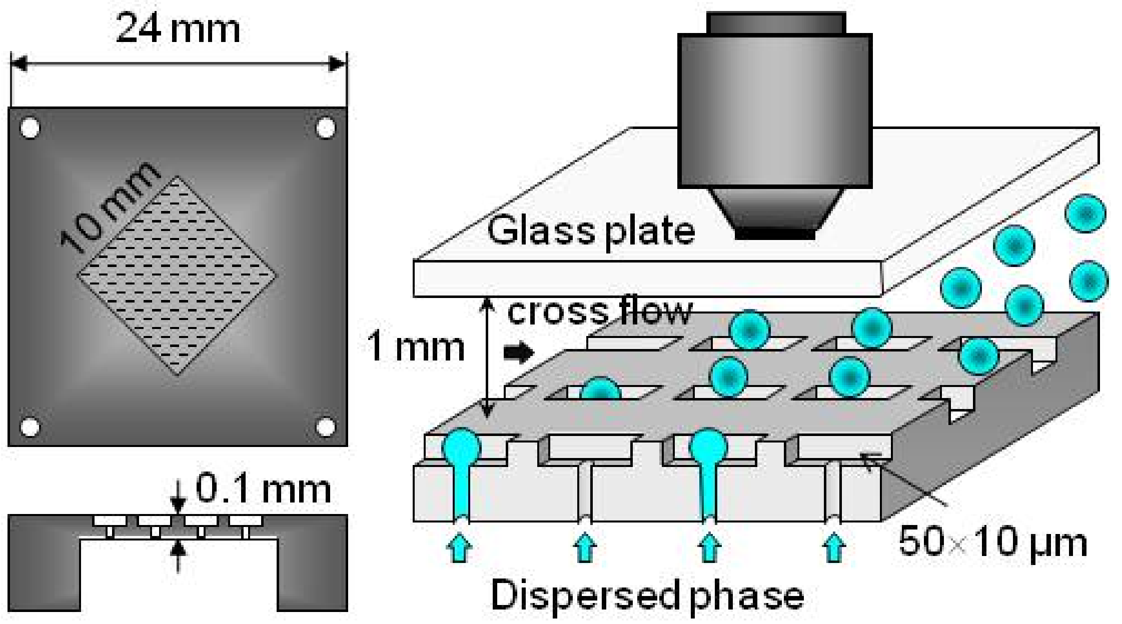

Fig.1. Schematic of the top and cross sectional view of the silicon MC plate used (left) and a schematic view of drop generation from a small portion of the plate (right).

The experiments have been carried out using silicon 24×24 mm MC plate (model WMS1-3, EP. Tech Co., Ltd., Hitachi, Japan) containing 23,489 straight-through MCs arranged within a 10 × 10 mm square area (Fig. 1). The plate was fabricated by photolithography and deep reactive ion etching (DRIE). Each MC consisted of a circular 10-µm diameter hole with a depth of 70 µm and a 50×10 µm slot with a depth of 30 µm. After fabrication the plate was subjected to plasma oxidation to form a hydrophilic layer of SiO_2 on the surface. The dispersed phase was injected through the MC plate by a Harvard Apparatus, model 11 Plus syringe pump. The droplets were carried away from the module by a continuous phase delivered in cross flow through the gap between the plate and transparent cover glass. The drop generation was observed using a fast-speed camera at 600 pps attached to microscope and PC computer. The particle size distribution of the resultant emulsions was measured using a Beckman Coulter LS 13 320 light scattering instrument. The disperse phase was soybean oil, MCT (medium-chain fatty acid triglyceride) oil and n-tetradecane with a viscosity at 293 K of 50, 20, and 2.7 mPa·s, resp. The continuous phase was 2 wt% Tween 20 or 2 wt% SDS dissolved in Milli Q water. No significant difference in drop size between Tween 20 and SDS was observed.

3. Results and discussion

3/1. CFD simulation

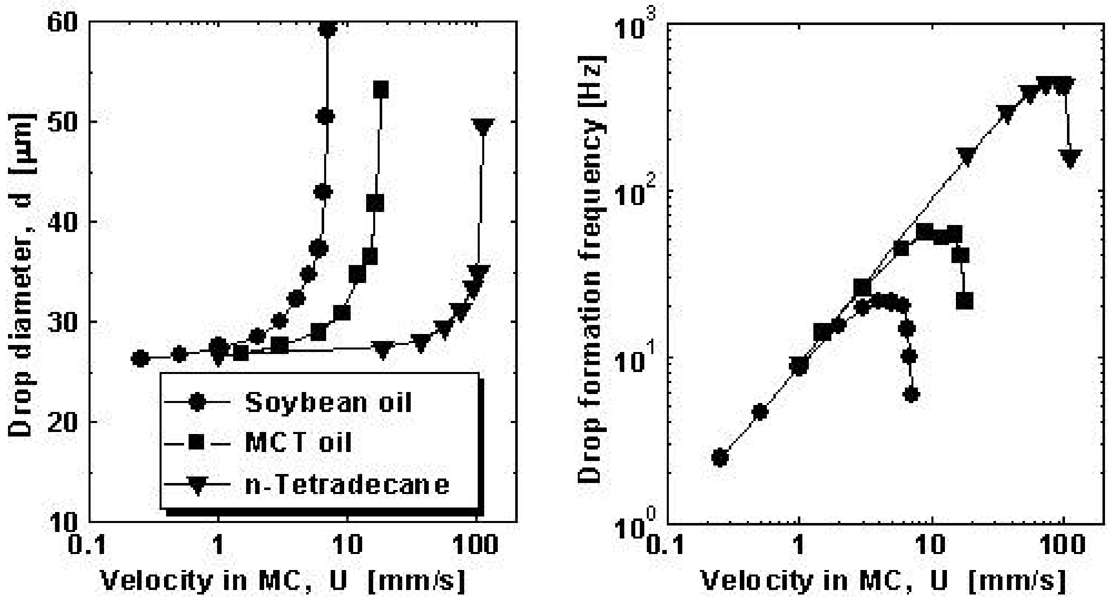

Fig.2. The CFD results showing the effect of oil velocity in a single MC on the drop diameter and the number of drops generated per second.

The CFD results for drop generation through a single MC are presented in Fig. 2. At the oil velocity in MC of less than 1 mm/s, the drop diameter is independent on the oil velocity in MC and the oil viscosity and ranges between 26 and 27.5 µm. The drop diameter sharply increases at a certain critical velocity U_{cr} corresponding to the maximum drop generation frequency. At $U < U_{cr}$ the interfacial tension force dominates the viscous drag force and the drop diameter is only controlled by the MC geometry (9). At $U > U_{cr}$ the viscous drag force dominates the interfacial tension and the drop size is controlled by the balance between the shear force as a result of cross flow and the velocity-dependent viscous drag force in MC. Within this regime bigger drops are formed at the lower formation frequencies and the drop size highly depends on the velocity in MC and the oil viscosity. U_{cr} ranges from 4 mm/s for soybean oil to 92 mm/s for n-tetradecane. Fig 3. shows that on a log-log scale the maximum number of drops that can be formed from a single MC decreases linearly with increasing the oil viscosity and the same type of behaviour was found for the critical velocity. The maximum drop generation frequency from CFD results is 440 Hz for n-tetradecane and 21 Hz for soybean oil.

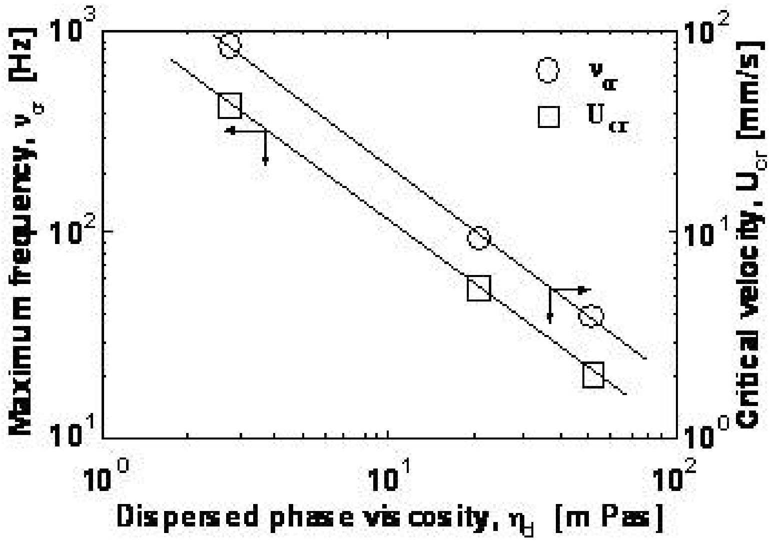

Fig.3. The maximum drop generation frequency from a single asymmetric MC and the critical velocity in MC as a function of the dispersed phase viscosity. The results are obtained using CFD analysis.

3/2. Experimental investigations

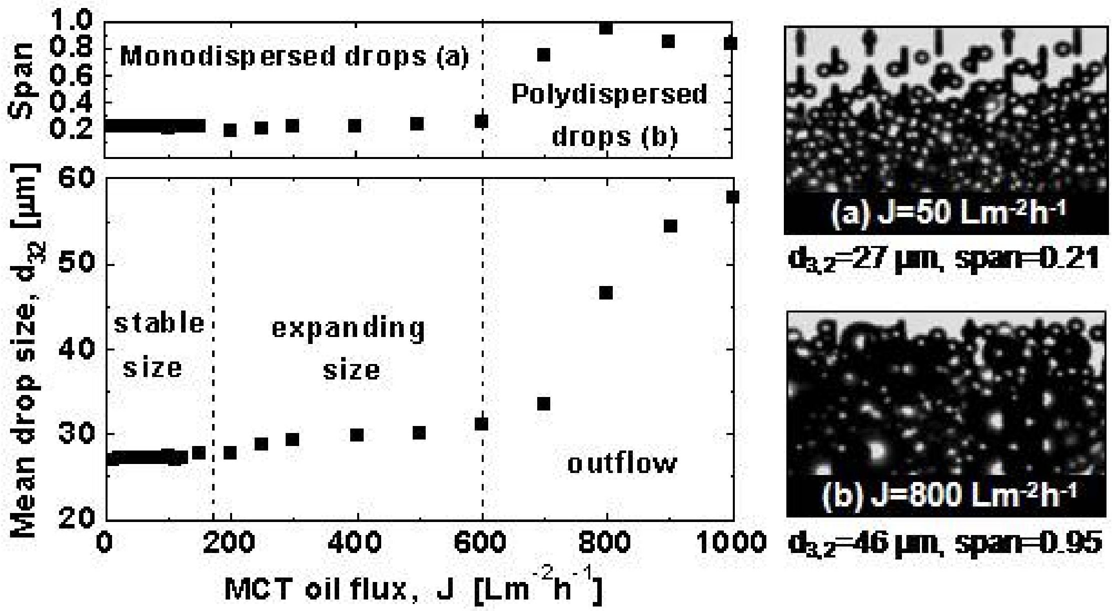

Fig. 4. The effect of oil flux through the MC plate on the mean drop size and the span of drop span distribution for MCT oil. The pictures showing generation of droplets below the critical flux (region a) and above the critical flux (region b) are shown on the right-hand side.

Fig. 4 illustrates the oil flux vs. mean drop size relationship for MCT oil. Monodispersed drops were formed in region (a) which was maintained up to the flux of 600 $Lm^{-2}h^{-1}$. For J < 170 $Lm^{-2}h^{-1}$ the mean drop size was virtually constant ($d_{3,2}$ = 27.1-27.6 µm) with a span of 0.21-0.23 and this range was referred to as the size-stable zone (1). For 170 <J< 600 $Lm^{-2}h^{-1}$ (the 'size-expanding' zone), $d_{3,2}$ expanded from 27.6 to 31.2 µm and the drops retained its monodispersity with a span of 0.20-0.25. Above the critical flux, the majority of MCs produced small uniform drops, but large droplets were formed from some of the MCs. As a result, the drops were polydisperse with a span of 0.75-0.95. The mean oil velocity U in MCs and oil flux J are related by the equation:

$U = 4JA_m/(\pi k d^2 N_0)$ (1)

where A_m is the effective cross sectional area of the MC plate (=10^{-4} m^2), d is the MC diameter (=10^{-5} m), k is the fraction of active MCs, and N_0 = 23,348 is the number of MCs in the plate. In the size stable zone assuming k =1 one obtains U < 2.6 mm/s from Eq. (1), which agrees well with Fig. 2 and 3. In the size-expanding zone one obtains 2.6 < U < 9.1 mm/s. The critical velocity of 9.1 mm/s calculated from Eq. (1) was in excellent agreement with U_{cr} value from CFD results of 9 mm/s.

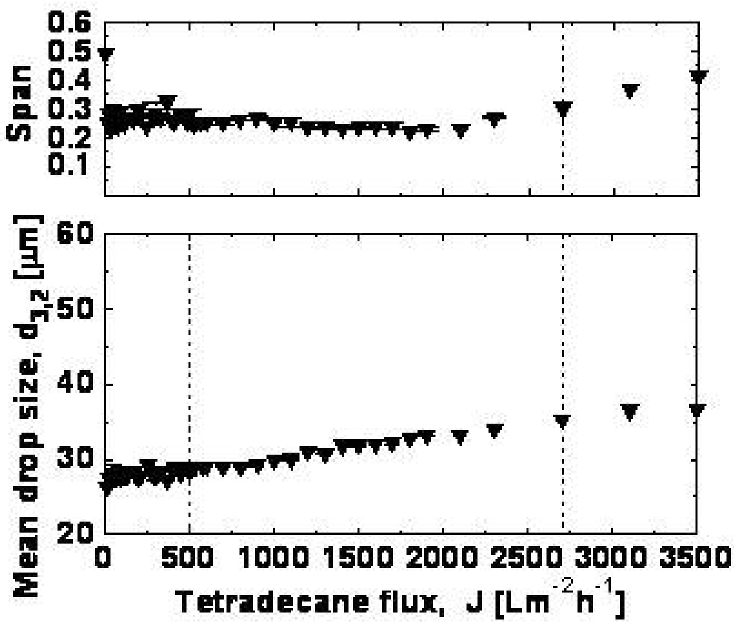

Fig. 5. The effect of flux through the MC plate on the mean Sauter diameter and the span of drop size distribution for n-tetradecane emulsions.

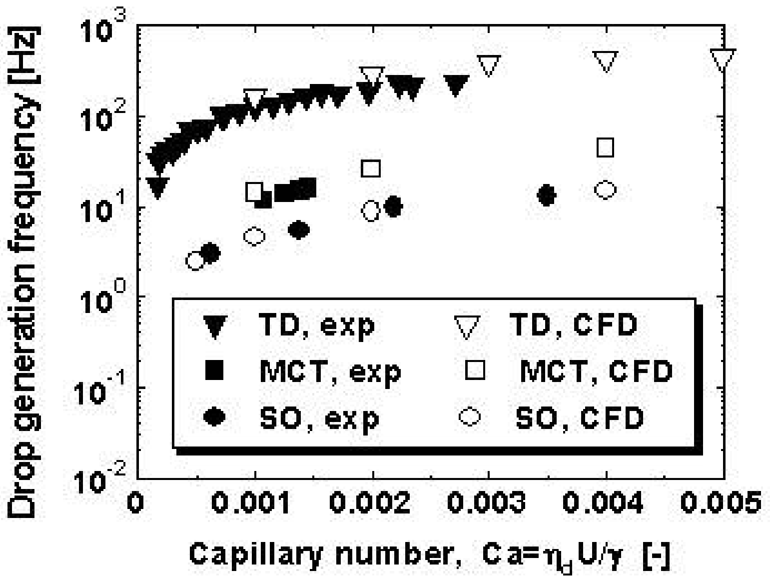

Fig. 6. Comparison between simulation (CFD) and experimental results for the drop generation from a single MC as a function of the capillary number. Dispersed phase: TD= n-tetradecane, MCT= MCT oil, SO= soybean oil.

Fig. 5 shows the effect of tetradecane flux on the size and monodispersity of tetradecane drops. At J < 500 $Lm^{-2}h^{-1}$ the drop size was independent on the tetradecane flux. A narrow particle size distribution with a span in the range of 0.22-0.30 persisted to J = 2700 $Lm^{-2}h^{-1}$. A transition from size-expanding to outflow regime was very gradual, so even at J = 3500 $Lm^{-2}h^{-1}$ the span was only 0.41. The fraction of active channels was found to approach 50% as the flux approached the critical value.

By putting k = 0.5 and U_{cr} = 92 mm/s (from Fig. 3) in Eq. (1), one obtains J_{cr}= 3040 $Lm^{-2}h^{-1}$. Fig. 6 shows a comparison between experimental and theoretical values for drop generation frequency. The agreement between the experimental and CFD results was excellent for soybean oil and less satisfactory for n-tetradecane. The maximum drop generation frequency for n-tetradecane was found to be 250 Hz per channel and nearly 3 million Hz per module on the assumption that 50% of the MCs were active. The high drop generation frequency for n-tetradecane as compared to soybean oil has been found to be a consequence of smaller number of active MCs.

4. Conclusions

Highly uniform droplets of soybean oil, MCT (medium-chain fatty acid triglyceride) oil and n-tetradecane have been generated using a silicon MC array consisting of 10×50 µm slots on top of circular 10-µm diameter channels. The critical flux of dispersed phase decreased with increasing the viscosity of dispersed phase. The critical flux for n-tetradecane was around 2700 $Lm^{-2}h^{-1}$ and the maximum drop generation frequency per single MC was 250 Hz and nearly 6 million per module. The correlation between experimental and CFD results was very good, particularly for soybean oil.

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