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## Large microchannel emulsification device for producing monodisperse fine droplets

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### Abstract

In this paper, we present a novel microchannel emulsification (MCE) system for mass-producing uniform fine droplets. A 60 × 60-mm MCE chip made of single-crystal silicon has 14 microchannel (MC) arrays and  $1.2 \times 10^4$  MCs, and each MC array consists of many parallel MCs and a terrace. A holder with two inlet through-holes and one outlet through-hole was also developed for simply infusing each liquid and collecting emulsion products. The MCE chip was sealed well by physically attaching it to a flat glass plate in the holder during emulsification. Uniform fine droplets of soybean oil with an average diameter of 10 μm were reliably generated from all the MC arrays. The size of the resultant fine droplets was almost independent of the dispersed-phase flow rate below a critical value. The continuous-phase flow rate was unimportant for both the droplet generation and the droplet size. The MCE chip enabled mass-producing uniform fine droplets at  $1.5 \text{ mL h}^{-1}$  and  $1.9 \times 10^9 \text{ h}^{-1}$ , which could be further increased using a dispersed phase of low viscosity.

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### 1. Introduction

Microchannel emulsification (MCE) is a relatively new and promising technique to produce uniform droplets with a coefficient of variation (CV) of typically below 5% [1]. MCE chips consist of MC arrays with many parallel microgrooves and a terrace [1] or many through-holes [2]. In MCE, droplet generation is driven by spontaneously transformation of the dispersed phase that has passed through the channels [3].

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The flow of the dispersed phase is the most important process parameter affecting droplet generation for MCE, since the gentle flow of the continuous phase used to collect the resultant droplets is negligible in such droplet generation. Moreover, the size and uniformity of droplets generated by MCE are not sensitive to the velocity of the dispersed phase below the critical value [4], which is suitable for readily performing emulsification. The current MCE chips enable generating uniform (fine) droplets of 1 to 200  $\mu\text{m}$  [5,6]. Uniform droplets generated by MCE are also useful templates for producing uniform food-grade microspheres and microcapsules [7,8].

The previous MCE chips for generating small droplets had a very low throughput in terms of volume flow rate of the dispersed phase ( $Q_d$ ), since droplets are usually generated from one pair of short MC arrays [5]. For instance, uniform fine (10  $\mu\text{m}$ ) droplets of triglyceride oil are generated at  $Q_d$  of  $<10^{-1}$   $\text{mL h}^{-1}$  using an MCE chip. This throughput would be difficult to satisfy even in laboratory-scale production. Thus, the scaling up of MCE systems is vital for increasing the number of MCs.

Here, we present a new MCE chip consisting of integrated MC arrays, which is intended for reliably mass-producing uniform fine droplets. The MC geometry selected here is an MC array consisting of parallel microgrooves and a terrace. We also investigated droplet generation in the new MCE chip and the throughput capacity of uniform fine droplets for such a chip. The silicon MCE chip consisting of shallow microgrooves, terraces and deep channels is strong enough for handling. A new holder with two inlet through-holes and one outlet through-hole was also designed and used for simple MCE operation. Since an MCE chip only physically attaches to a transparent plate in the module without chemical bonding, the chip can be detached from the plate when necessary. This feature is practically advantageous when tiny particles that have entered the MCE chip disrupts droplet generation.

## 2. Materials and Methods

A 60  $\times$  60-mm MCE chip containing 14 MC arrays was designed for this study (Fig. 1). Each parallel MC array (length 34 mm) consisted of 850 MCs and terraces positioned at the inlet and outlet; 11,900 MCs were positioned on the chip. The steps were designed to be much deeper than MCs to enhance the dispersibility of the generated droplets. A channel with two through-holes was located at the outlet side of each MC array. In this channel, a continuous phase was infused from the inlet through-hole, and an emulsion was collected from the outlet through-hole. A channel for supplying a dispersed phase was located at the inlet side of each MC array. All the channels were connected at side edges of the chip, which opened to channels outside the chip. This geometry also contributes to reducing the chip size, since the channel formed outside the chip enables forming a laminar dispersed-phase flow. As a result, the MCE chip designed here increased the number of MCs by 17 times that of the previous crossflow-type MCE chip [9]. MC arrays and deeper channels were fabricated on a single-crystal silicon substrate with a thickness of 0.5 mm via two steps of anisotropic wet etching. Through-holes with a diameter of 1.0 mm were fabricated in an MCE chip by sandblast etching. The MC cross-section and terrace length in the fabricated MC arrays were highly uniform, which is essential for generating uniform droplets by MCE. The fabricated long MCs had a depth of 2  $\mu\text{m}$ , a width of 10  $\mu\text{m}$ , and a length of 104  $\mu\text{m}$ ; the terraces fabricated at the MC outlets had a depth of 2  $\mu\text{m}$  and a length of 19  $\mu\text{m}$ . These MC and terrace dimensions were determined to stably generate uniform fine droplets. The step depth was 100  $\mu\text{m}$ .

An MCE module designed for this study has a diameter of 15 cm and a height of 2.4 cm. The holder consists of a bottom lid, an artificial quartz glass plate, and an upper cover plate. Fluoro-rubber o-rings and a spacer are also attached inside the holder for appropriate separation of two liquids. The bottom lid, equipped with the glass plate and the spacer, forms a reservoir where an MCE chip is mounted. The large

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