RESEARCH PAPER

Straight-through microchannel devices for generating monodisperse emulsion droplets several microns in size

**Isao Kobayashi · Takayuki Takano · Ryutaro Maeda ·
Yoshihiro Wada · Kunihiko Uemura ·
Mitsutoshi Nakajima**

Abstract The authors recently proposed a promising technique for producing monodisperse emulsions using a straight-through microchannel (MC) device composed of an array of microfabricated oblong holes. This research developed new straight-through MC devices with tens of thousands of oblong channels of several microns in size on a silicon-on-insulator plate, and investigated the emulsification characteristics using the microfabricated straight-through MC devices. Monodisperse oil-in-water (O/W) and W/O emulsions with average droplet diameters of 4.4–9.8 μm and coefficients of variation of less than 6% were stably produced using surface-treated straight-through MC devices that included uniformly sized oblong channels with equivalent diameters of 1.7–5.4 μm. The droplet size of the resultant emulsions depended greatly on the size of the preceding oblong channels. The emulsification process using the straight-through MC devices developed in this research had very high apparent energy efficiencies of 47–60%, defined as (actual energy input applied to droplet generation/theoretical minimum energy input necessary for making droplets) × 100. Straight-through MC devices with numerous oblong microfluidic channels also have great potential for increasing the productivity of monodisperse fine emulsions.

I. Kobayashi (✉) · K. Uemura · M. Nakajima
Food Engineering Division, National Food Research Institute,
2-1-12 Kannondai, Tsukuba, Ibaraki 305-8642, Japan
e-mail: isaok@affrc.go.jp

M. Nakajima (✉)
e-mail: mnaka@affrc.go.jp

T. Takano · R. Maeda
Advanced Manufacturing Research Institute,
National Institute of Advanced Industrial Science
and Technology, 1-2-1 Namiki, Tsukuba,
Ibaraki 305-8564, Japan

Y. Wada
EP. Tech Co., Ltd, 2-4-1, Shiroganecho, Hitachi,
Ibaraki 317-0056, Japan

1 Introduction

Emulsions are thermodynamically metastable dispersions of two immiscible liquids in the presence of surfactants and/or micro/nano-particles, with one of the liquids dispersed as droplets in the continuous phase of the other. Emulsion droplets with diameters of 0.1–10 μm have diverse applications, including foods, cosmetics, pharmaceuticals, and chemicals. Monodisperse emulsions with very narrow size distributions can improve stability against droplet coalescence, enabling us to more easily clarify many important emulsion properties (Mason et al. 1996; McClements 2004). In addition, many important potential emulsion uses require monodisperse emulsions that are precisely controlled in size. However, conventional emulsification techniques, which use extensional and shear stress or impact to disrupt droplets, usually produce polydisperse emulsions with broad droplet size distributions (Karbstain and Schubert 1995; McClements 2004). Nakashima et al. (1991) proposed (crossflow) membrane emulsification to generate quasi-monodisperse emulsion droplets with coefficients of variation ''CV (standard deviation/average droplet diameter × 100)'' of approximately 10%. Several techniques have also been proposed to reduce the droplet size distribution of polydisperse emul-

sions (Bibette 1991; Mason and Bibette 1996; Suzuki et al. 1996; Vladisavljević et al. 2006).

Interest in microfluidic techniques for generating individual emulsion droplets has increased in the last decade. Kawakatsu et al. (1997) proposed microchannel (MC) emulsification to generate monodisperse emulsion droplets with CVs of less than 5% using a microfluidic channel array with a slit-like terrace. MC emulsification enables generation of monodisperse droplets by forcing a to-be-dispersed phase through channels into a deeply etched well filled with a continuous phase. Droplet generation for MC emulsification exploits the instability and nonlinearity of the flow of two liquid phases driven by interfacial tension, which is the dominant force on a micrometer scale; the to-be-dispersed phase that broke through the channels is transformed spontaneously into droplets (Sugiura et al. 2001a). Kobayashi et al. (2002, 2004a) proposed a microporous device with ten thousand oblong microfluidic holes ''a straight-through MC device'' and demonstrated the production of monodisperse oil-in-water (O/W) emulsions at increased droplet generation rates of up to 10^5 s^{-1}. Further scale-up of the straight-through MC device is in progress.

Throsen et al. (2001) and Nishisako et al. (2002) reported a T-junction of microfluidic channels at which monodisperse emulsion droplets can be generated by shearing off a to-be-dispersed phase using the crossflow of a continuous phase. Anna et al. (2003) designed a flow-focusing geometry composed of microfluidic channels and a narrow gap, which generated emulsion droplets by breaking up a to-be-dispersed-phase stream between two continuous-phase streams through the narrow gap. Xu and Nakajima (2004) generated monodisperse emulsion droplets using simple flow-focusing geometry. Several research groups have used three-dimensional flow-focusing devices with a microfluidic capillary or orifice to generate monodisperse droplets of single and double emulsions (Lorenceau et al. 2005; Takeuchi et al. 2005; Utada et al. 2005; Yobas et al. 2006). Priest et al. (2006) designed a device composed of a T-junction of microfluidic channels and a step downstream of the T-junction and generated monodisperse emulsion droplets near the T-junction and at the step, controlled by the flow rates of the two phases. Vladisavljević and Williams (2006) proposed a rotating device with microfabricated pores and produced monodisperse emulsions. Link et al. (2004) generated monodisperse emulsion droplets by passive breakup of uniformly sized larger droplets at a T-junction of microfluidic channels. However, one microfluidic channel for the to-be-dispersed phase was typically positioned in the preceding devices, leading to a limited droplet production rate per device, generally <10^3 s^{-1} for O/W emulsions and <10^4 s^{-1} for W/O emulsions.

Monodisperse emulsion droplets generated using microfluidic techniques have been used to produce advanced micro-materials, such as monodisperse solid and gel microparticles (e.g. Sugiura et al. 2000, 2001b; Iwamoto et al. 2002; Nishisako et al. 2004; De Geest et al. 2005; Xu et al. 2005), monodisperse microcapsules (e.g. Nakagawa et al. 2004; Takeuchi et al. 2005; Utada et al. 2005), monodisperse double emulsions (e.g. Kawakatsu et al. 2001a; Okushima et al. 2004; Utada et al. 2005), and monodisperse colloidal assemblies (e.g. Yi et al. 2003a, b). Many practical applications of monodisperse emulsions require small droplets several microns in size; for example, column microparticles for HPLC, spacer microparticles in electronic devices, and carrier microparticles and microcapsules for drug delivery systems (DDS). However, microfluidic techniques using a T-junction or flow-focusing geometry usually generate monodisperse emulsion droplets larger than 10 μm. MC emulsification using a microfluidic channel array is capable of generating monodisperse emulsion droplets several microns in size (Kobayashi et al. 2001, 2006), although this technique has a low droplet throughput problem due to the arrangement of the channels. In contrast, existing straight-through MC devices consisting of deep oblong channels over 10 μm in size can generate only monodisperse emulsion droplets several tens of microns in size at high production scales. However, the previous microfabrication process of straight-through MC devices made of single-crystal silicon (Kobayashi et al. 2002) had a difficulty in precisely manufacturing deep oblong channels several microns in size. It is therefore necessary to develop a new microfabrication process for manufacturing straight-through MC devices consisting of deep oblong channels several microns in size.

The first purpose of this research was to manufacture straight-through MC devices with deep oblong channels several microns in size using a new microfabrication process and a silicon-on-insulator (SOI) wafer. The oblong channels microfabricated in this research was microscopically evaluated, which are vertical to the top surface of straight-through MC devices. Our second objective was to examine the generation characteristics of O/W and W/O emulsion droplets several microns in size using the surface-treated straight-through MC devices with different channel sizes developed in this research.

2 Fundamentals of emulsification using straight-through MC devices

Figure 1 schematically illustrates the production of an O/W emulsion using a straight-through MC device. The channel cross-sectional shape is the most important factor affecting the emulsification process using straight-through MC devices. Straight-through MC devices consisting of oblong channels with aspect ratios exceeding a threshold value of
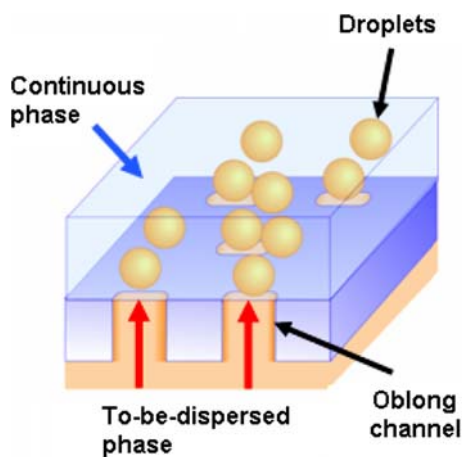
Fig. 1 Schematic illustration of production of an O/W emulsion from the outlet of the oblong channels

approximately three are capable of producing monodisperse emulsions (Kobayashi et al. 2002, 2004a). However, the use of straight-through MC devices consisting of circular channels or oblong channels with small aspect ratios results in continuous outflow of the to-be-dispersed phase or the generation of polydisperse emulsions (Kobayashi et al. 2002, 2004a). An analysis of these emulsification processes using the computational fluid dynamics (CFD) method demonstrates that a sufficient space for the continuous phase at the channel outlet must be maintained to generate uniformly sized droplets (Kobayashi et al. 2004b). The oblong channels with a large aspect ratio, which can maintain the preceding sufficient space during the droplet generation process, enables rapid shrinkage and instantaneous cutoff of the neck formed inside the channel, leading to the production of monodisperse emulsions. The droplet generation process is driven by the difference in Laplace pressure between the to-be-dispersed phase inside the channel ($\Delta P_{\text{Laplace},1}$) and the to-be-dispersed phase expanding from the channel outlet ($\Delta P_{\text{Laplace},2}$), expressed as $\Delta P_{\text{Laplace},1}$–$\Delta P_{\text{Laplace},2}$ (Sugiura et al. 2001a). In contrast, the circular channels or oblong channels with a small aspect ratio cause blockage of the channel outlet by the to-be-dispersed phase, hindering droplet generation from the channel outlet.

In MC emulsification, droplets are generated from channels with a slit-like terrace at applied pressures of a to-be-dispersed phase exceeding a breakthrough pressure. The breakthrough pressure, at which the to-be-dispersed phase can pass through the channels, is estimated by the Young–Laplace equation considering the contact angle effect

$$\Delta P_{\text{BT}} = 4\gamma \cos \theta / d_{\text{ch}} \tag{1}$$

where ΔP_{BT} is the Young–Laplace pressure between the two phases, γ is the interfacial tension, θ is the contact

angle of the to-be-dispersed phase to the channel surface, and d_{ch} is the channel diameter (Sugiura et al. 2000).

3 Experimental

### 3.1 Device design and microfabrication

A new straight-through MC plate was designed to produce monodisperse emulsion droplets several microns in size (Fig. 2). The straight-through MC plate had a size of 15×15-mm^2 top surface area and 525-μm thickness. The 8.4×8.4-mm channel area was placed in the center of the plate, and the 30-μm deep vulnerable channel area was reinforced by a latticed support layer. Oblong microfluidic channels several microns in size and vertical to the top surface of the plate were positioned at specific intervals sufficient to prevent droplets that expand from adjacent channel outlet to come into contact. The aspect ratio value of the channel outlet was also designed based on the
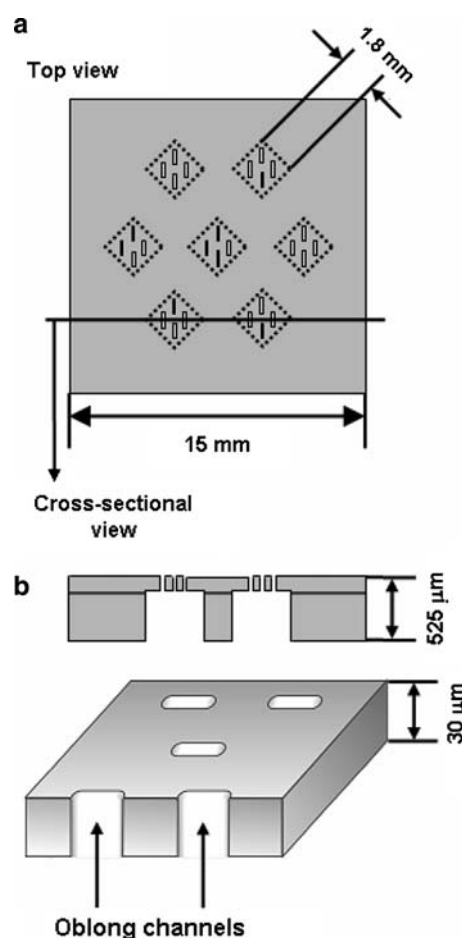
Fig. 2 Schematic illustration of a silicon straight-through MC plate: **a** top and cross-sectional views of the plate. **b** The straight-through MC with oblong channels designated by *dotted lines* in **a**

previous findings for emulsification using straight-through MC devices described in Sect. 2.

A straight-through MC plate made of silicon was manufactured via the process illustrated in Fig. 3. A 4-in.-diameter SOI wafer containing a 1-μm thick box layer (Toshiba Ceramics Co. Ltd., Tokyo, Japan) was used in this research (Fig. 3a). A thin layer of a photoresist (TSMR-V90, Tokyo Ohka Kogyo Co. Ltd., Kawasaki, Japan) was formed on the top surface of the SOI wafer by spincoating (1H-DX2, Mikasa Co. Ltd., Tokyo, Japan) (Fig. 3b). This layer was patterned by photolithography using a double-view mask aligner (PEM-800, Union Optical Co. Ltd., Tokyo, Japan) and a photomask for oblong channels and its subsequent development (NMD-3, Tokyo Ohka Kogyo Co. Ltd., Kawasaki, Japan) at room temperature (Fig. 3c, d). A second spincoating formed a thin layer of a photoresist (AZP4903, Clariant (Japan) K.K., Tokyo, Japan) on the bottom surface of the SOI wafer (Fig. 3e). The subsequent photolithography and development (AZ 400K, Clariant (Japan) K.K., Tokyo, Japan) of this layer patterned wells in it (Fig. 3f). Deep oblong channels were fabricated in the top silicon layer using the Alcatel A601E deep reactive ion etching (DRIE) etcher (Alcatel Vacuum Technology France, Annecy, France) (Fig. 3g), and DRIE was subsequently preformed in the bottom silicon layer to create deep wells (Fig. 3h).
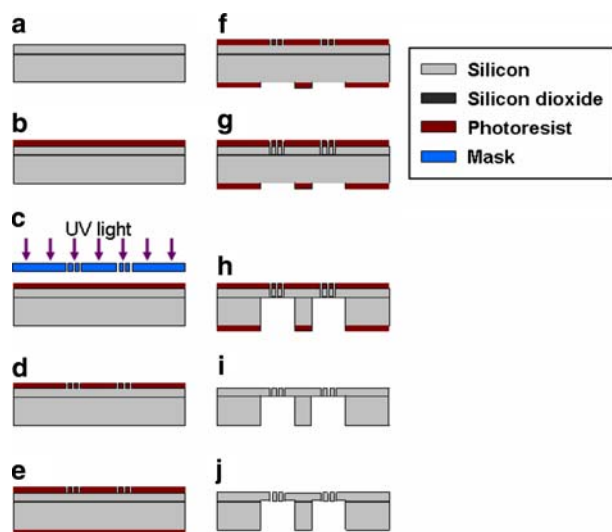
Fig. 3 Microfabrication process scheme for a silicon straight-through MC plate: **a** 4-in.-diameter silicon-on-insulator (SOI) wafer composed of a 30-μm thick top layer, a 1-μm thick box layer, and a 494-μm thick bottom layer. **b** Spincoating of a photoresist on the top surface. **c**, **d** Patterning the oblong channels in the top photoresist layer by photolithography **c** and development **d**. **e** Spincoating of a photoresist on the bottom surface. **f** Patterning the wells in the bottom photoresist layer by photolithography and development. **g** Deep reactive ion etching (DRIE) of the top silicon layer. **h** DRIE of the bottom silicon layer. **i** RIE of the box layer between the oblong channels and wells. **j** Removal of the photoresist layers on the wafer surfaces

The patterned photoresist layers were removed from the SOI wafer by soaking in acetone for 1 min. plus in iso-propyl alcohol for 1 min., and then washing using purified water (Fig. 3i). Finally, the box layer was dry-etched using a RIE system (Model RIE-10NRS, Samco Inc., Kyoto, Japan) in order to connect the oblong channels to the wells (Fig. 3j). The microfabricated SOI wafer was cut into individual straight-through MC plates using an automatic dicing saw (DAD522, Disco Co., Tokyo, Japan).

3.2 Surface treatment of straight-through MC devices

Stable production of monodisperse emulsions for MC emulsification can only be achieved when the continuous phase preferentially wets the channel surface (Tong et al. 2000; Kawakatsu et al. 2001b; Kobayashi et al. 2003). The channel surface must be hydrophilic for O/W emulsifica-tion experiments and hydrophobic for W/O emulsification experiments. Straight-through MC plates for producing O/W emulsions were obtained after a hydrophilic silicon-dioxide layer was grown on the plate surface using an oxygen plasma reactor. The straight-through MC plate for producing W/O emulsions was obtained by the following silanization treatment based on procedures in published literature (Kawakatsu et al. 1997; Sugiura et al. 2001c). A straight-through MC plate treated by plasma oxidation was silanized by immersion into a toluene solution containing 5.0 wt.% octadecyltriethoxysilane (LS-6970, Shin-Etsu Chemical Co., Ltd., Tokyo, Japan) at 110°C for 1 h. The silanized plate was ultrasonicated in toluene for 40 min and subsequently in hexane for 40 min to wash out unreacted materials. The static contact angles of MilliQ water drop-lets to the top surface of the oxidized straight-through MC plate and silanized straight-through MC plate in the air were measured.

3.3 Chemicals for droplet generation

In the O/W emulsification experiments, refined soybean oil (Wako Pure Chemical Ind., Osaka, Japan) was used as the to-be-dispersed phase, and MilliQ water solution containing 1.0 wt.% sodium dodecyl sulfate (SDS, Wako Pure Chemical Ind.) was used as the continuous phase. In the W/O emulsification experiments, a mixture of 25 wt.% MilliQ water solution containing 5.0 wt.% so-dium chloride (Wako Pure Chemical Ind.) and 75 wt.% glycerol (Wako Pure Chemical Ind.) was used as the to-be-dispersed phase, and decane (Wako Pure Chemical Ind.) solution containing 3.0 wt.% tetraglycerin monola-urate condensed ricinoleic acid esters (CR-310, Sakamoto Yakuhin Kogyo Co. Ltd., Osaka, Japan) was used as the continuous phase.

3.4 Density, viscosity, and interfacial tension measurements

A density meter (DA-130N, Kyoto Electronics Manufacturing Co. Ltd., Kyoto, Japan) to determine the densities of the liquid phases was used in this research. Their viscosity was measured using glass capillary viscometers (SO, Shibata Scientific Technology Ltd., Tokyo, Japan). The equilibrium interfacial tension at the interface between two immiscible liquid phases was determined using a pendant drop interfacial tensiometer (PD-W, Kyowa Interface Science Co. Ltd., Saitama, Japan). The density and viscosity measurements were repeated at least twice and interfacial tension measurements at least ten times and calculated the mean values. All the preceding measurements were conducted at 25°C. Table 1 presents the measured density, viscosity, and equilibrium interfacial tension of the O/W and W/O systems used in this study.

3.5 Emulsification setup and procedures

Figure 4 schematically illustrates the emulsification setup equipped with a straight-through MC plate. Two emulsification modules were designed to produce O/W emulsions (Fig. 4) and W/O emulsions. The plate was degassed in the continuous phase with ultrasonification at 100 kHz for 20 min prior to the emulsification experiments. The degassed plate was installed in the module, which is filled with the continuous phase. The continuous phase was introduced into the module via flexible tubing connected to
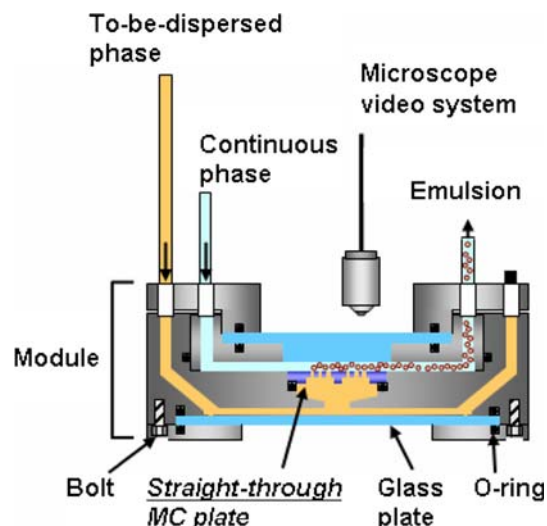

Fig. 4 Schematic representation of the emulsification module equipped with a straight-through MC plate for O/W emulsions

a syringe installed in a syringe pump (Model 11, Harvard Apparatus. Inc., MA, USA). The to-be-dispersed phase was introduced into the module via flexible tubing connected to a liquid chamber or a syringe installed in the syringe pump, filling the wells underneath the channel area. The to-be-dispersed phase was forced to break through the oblong channels to generate emulsion droplets, as schematically illustrated in Fig. 1. Droplet generation from the channel outlet was microscopically visualized and recorded using a microscope video system described elsewhere (Kobayashi et al. 2002). Emulsion droplets generated from the outlet of oblong channels moved away from the top surface of the straight-through MC plate by the gentle crossflow of the continuous phase in the clearance between the upper glass plate and the straight-through MC plate (1.0 mm). The emulsification experiments were conducted at about at 25°C.

3.6 Droplet size analysis

WinRoof software (Mitani Co. Ltd., Fukui, Japan) was used to measure the diameters of droplets in the images obtained by the microscope video system. The number-average diameter (d_{av}) of the generated droplets was determined using the measured data of 200 droplets.

3.7 Cleaning of straight-through MC devices

Straight-through MC devices used in emulsification experiments were cleaned using an ultrasonic cleaner with a high frequency of 100 kHz (VS-100III, As One Co., Osaka, Japan). Straight-through MC plates for producing O/W emulsions were cleaned in MilliQ water solution

Table 1 Measured density, viscosity, and equilibrium interfacial tension of O/W and W/O systems used in this study

Type	O/W System	W/O System
To-be-dispersed phase	Refined soybean oil	Mixture of 25 wt.% MilliQ water solution[a] and 75 wt.% glycerol
Continuous phase	1.0 wt.% SDS[b] aqueous solution	3.0 wt.% CR-310[c] decane solution
To-be-dispersed phase density (kg/m³)	920	1,203
Continuous phase density (kg/m³)	999	736
To-be-dispersed phase viscosity (mPa s)	50.4	33.9
Continuous phase viscosity (mPa s)	0.98	1.00
Interfacial tension (mN/m)	4.5	2.2

[a] MilliQ water solution contains sodium chloride at a concentration of 5.0 wt.%

[b] Sodium dodecyl sulfate

[c] Tetraglycerin monolaurate condensed ricinoleic acid esters

containing a detergent for 40 min and in MilliQ water for 20 mim. Straight-through MC plates for producing W/O emulsions were cleaned in hexane for 20 min, MilliQ water solution containing a detergent for 40 min, and in MilliQ water for 20 min. Straight-through MC plates did not break during the ultrasonic cleaning process.

4 Results

4.1 Device characterization

Table 2 lists dimensions of the oblong channels microfabricated in the silicon straight-through MCs, TMS11-1, –2, and –3. Figure 5a presents a scanning electron micrograph of part of a channel area for TMS11-2. Microfabricated channels were regularly positioned in this channel area. There is a deeply-etched well in the support layer beneath the channel area. Figure 5b, c presents scanning electron micrographs of the inlet and outlet of the microfabricated channels with an oblong section. Oblong channels of uniform size were successfully obtained, although the channel inlet was somewhat larger than the channel outlet. A scanning electron micrograph of an oblong channel, provided in Fig. 5d, also demonstrates that a deep through-hole with an oblong section was successfully microfabricated in the channel area. Figures 5e–g depict three straight-through MCs with different channel sizes microfabricated in this study. These straight-through MCs had fine oblong channels with a shorter line down to 1.0 μm and a longer line down to 4.6 μm (see Table 2). Their equivalent diameters, also presented in Table 2, indicate successful microfabrication of the straight-through MCs with oblong channels several microns in size. The oblong channels microfabricated in each straight-through MC had a narrow size distribution with CVs of <2% for TMS11-2 and -3 and <4% for TMS11-1. Their uniform size satisfied a necessary condition for producing monodisperse emulsions in MC emulsfication. The aspect ratio at the channel outlet, which is defined as the channel shorter line divided by the channel longer line,

Table 2 Dimensions of oblong channels microfabricated in silicon straight-through MCs

No.	Channel shorter line (μm)	Channel longer line (μm)	Channel depth (μm)	Channel equivalent diameter[a] (μm)	Total channel number (–)
TMS11-1	1.0	4.6	30.0	1.7	92,575
TMS11-2	2.3	10.0	30.0	3.7	23,548
TMS11-3	3.3	15.0	30.0	5.4	10,647

[a] Channel equivalent diameter is defined as four times the cross-sectional area divided by the wetted perimeter of the channel

was 4.6 for TMS11-1, 4.3 for TMS11-2, and 4.5 for TMS11-3. It is thus considered that the oblong channels several microns in size microfabricated in this study have the potential to produce monodisperse emulsions. The total channel number for TMS11 plates ranged between 1.1×10^4 and 9.3×10^4 and increased with an decrease in the channel size (see Table 2). The surface porosity in the channel area, which is defined as the ratio of the total cross-sectional area of the oblong channels to the total channel area, was calculated to be 1.9% for TMS11-1, 2.4% for TMS11-2, and 2.3% for TMS11-3. The measured ratio of the channel shorter line to the channel depth was between 9.1 (TMS11-3) and 30.0 (TMS11-1). These straight-through MCs with deep (or long) channels are advantageous for stable production of monodisperse emulsions at high droplet generation rates, which is attributable to the sizeable pressure drop of the to-be-dispersed phase that flows inside the channel (Sugiura et al. 2002).

The plasma-oxidized TMS11 plates had a static water/air contact angle of <5°, indicating that their surface had become fully hydrophilic. In contrast, a TMS11-2 plate silanized using the procedure described in section 3.2 had a static water/air contact angle of 94°, indicating that its surface had become hydrophobic. These results demonstrate that TMS11 plates with controlled surface wettability for producing O/W and W/O emulsions were obtained.

4.2 Production of O/W emulsions using straight-through MC devices

Emulsification experiments using hydrophilic TMS11 plates were conducted at pressures applied to a to-be-dispersed phase (P_d) of 4.6–13.1 kPa, which was dependent on the channel size (Table 3). These P_d values were just above the breakthrough pressure at which droplet generation commences from the channel outlet, which is inversely proportional to $d_{eq,channel}$ (Kawakatsu et al. 1997; Tong et al. 2001). It is noticed that no breakage of TMS11 plates occurred during the emulsification operations. The flow velocity of the continuous phase (U_c) along the plate surface was varied in a range of 0.0–0.56 mm/s to better observe the droplet generation behavior from the outlet of oblong channels and the generated droplets.

Figure 6a, b depicts typical examples of the production of an O/W emulsion using TMS11 plates. Figure 6c presents a typical optical micrograph of the produced O/W emulsion. The to-be-dispersed phase that broke through the oblong channels in the TMS11 plates expanded in the area over the plate surface and was then cut into uniformly sized oil droplets. The average droplet generation rates at the active channels that generated droplets increased significantly with decreases in the channel size, i.e., >30 s⁻¹ for TMS11-1, 5.8 s⁻¹ for TMS11-2, and 3.8 s⁻¹ for TMS11-3.

Table 3 Apparent energy efficiency of production of O/W emulsions using TMS11 plates

| No.	P_d^a(a)	$\Delta E_{in,exp}^b$ (J/kg)	$\Delta E_{in,min}^c$ (J/kg)	Energy efficiency[d] ($\Delta E_{in,min}/\Delta E_{in,exp}$) [%]
TMS11-1	13.1	14.2	6.7	47.2
TMS11-2	6.7	7.3	4.4	60.2
TMS11-3	4.6	5.0	3.0	60.0

[a] The pressure applied to the to-be-dispersed phase

[b] The actual energy input for a unit to-be-dispersed phase necessary for droplet generation from oblong channels

[c] The theoretical minimum energy input to make emulsion droplets of d_{av} values shown in Fig. 6

[d] Apparant energy efficiency of production of O/W emulsions using TMS11 plates

The percentage of the active micro holes was <1% for TMS11-1, 5.2% for TMS11-2, and 12.3% for TMS11-3, which may be affected by the fabrication accuracy of the oblong channels and the flow condition and pressure dis-tribution in the region near the micro holes. Most of the oil droplets generated from the outlet of each active channel formed quasi-linear patterns consisting of several layers of droplets for TMS11-1 (Fig. 6a) and quasi-linear patterns consisting of jointed droplets for TMS11-2 and -3 (Fig. 6b). The droplets just after generation under these conditions were pushed out by the following droplets generated from the same channel outlet. It is assumed that the droplet patterns depicted in Fig. 6a, b were determined by the balance of the floating-up velocity of the generated droplets and the expansion velocity of the to-be-dispersed phase that broke through the channel outlet. No droplet coalescence for the droplets generated using TMS11 plates was observed during emulsification operations.

Figure 7a–c shows the droplet size distributions of the O/W emulsions produced using TMS11 plates. Figure 7d shows the effect of $d_{eq,channel}$ on the d_{av} and CV of the generated oil droplets. All of the emulsions formulated using TMS11 plates had very narrow droplet size distributions with CVs of less than 6% (Fig. 7a–c). Their d_{av}

Fig. 5 **a** Scanning electron micrograph of part of a silicon straight-through MC (TMS11-2) microfabricated in this study. **b**, **c** Scanning electron micrographs of the inlet **b** and outlet **c** of oblong channels microfabricated in TMS11-2. **d** Scanning electron micrograph of a cross section of an oblong channel microfabricated in TMS11-2. **e–g** Optical micrographs of oblong channels microfabricated in TMS11-1 **e**, TMS11-2 **f**, and TMS11-3 **g**
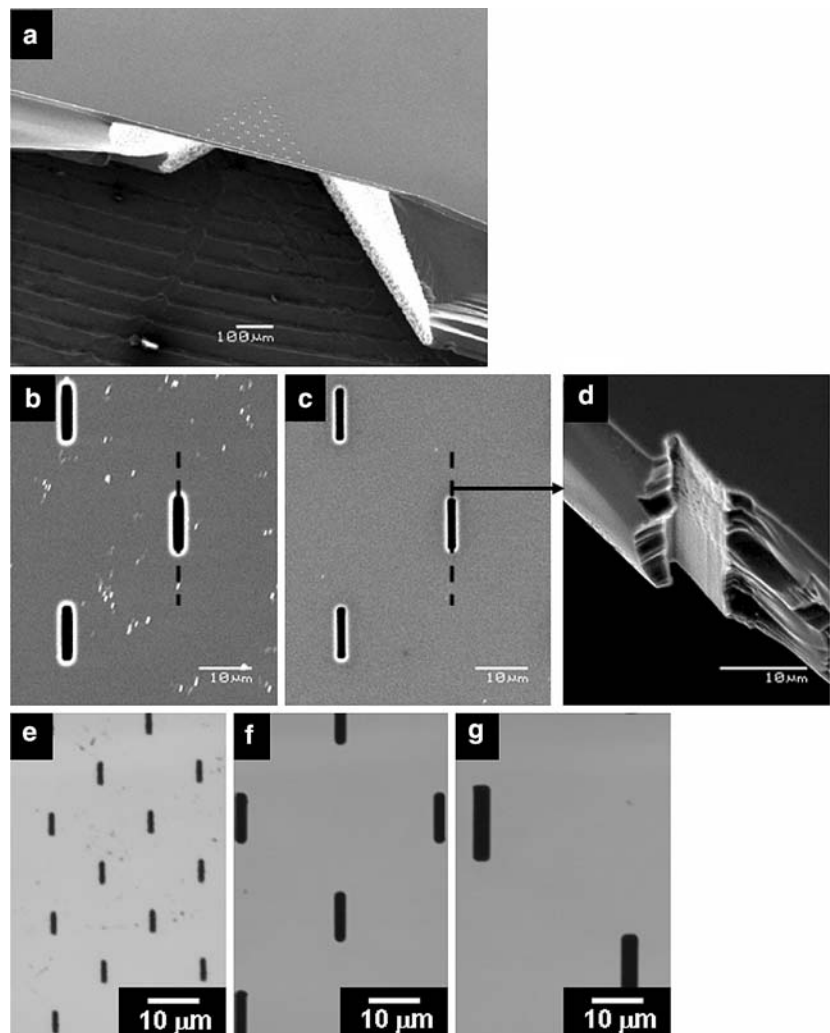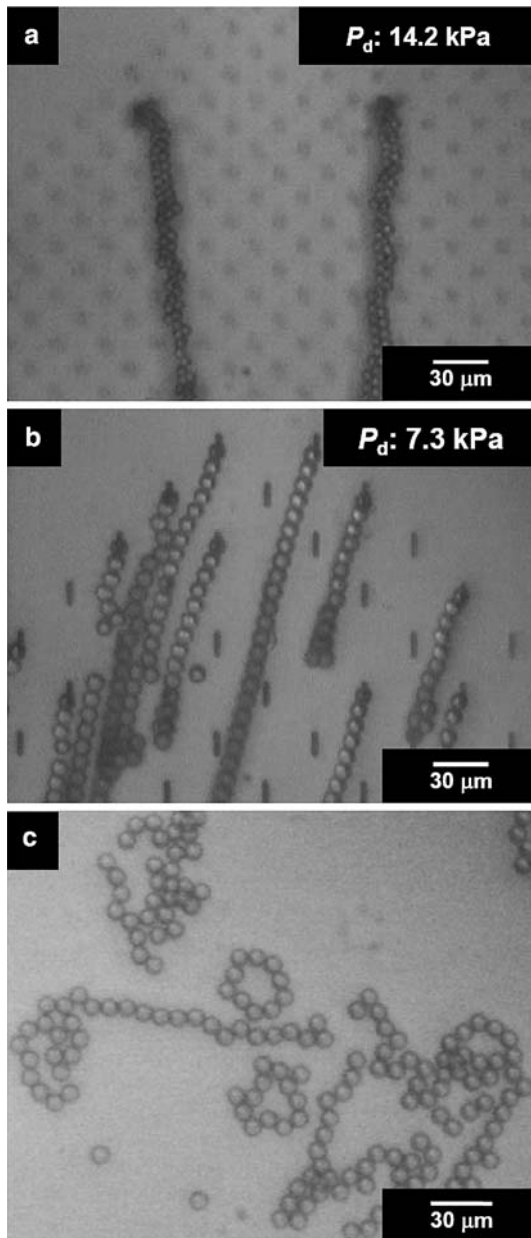
Fig. 6 Typical optical micrographs of successful production of an O/W emulsion using hydrophilic TMS11 plates: TMS11-1 **a** and TMS11-2 **b**. The flow velocity of the continuous phase along the plate surface was 0.56 mm/s. **c** Optical micrograph of a monodisperse O/W emulsion produced using the TMS11-2 plate

ranged from 4.4 to 9.8 μm as presented in Fig. 7a–c. Their d_{av} was also independent of the crossflow velocity in the range of U_c applied in this study. The minimum d_{av} value obtained in this study was considerably smaller, approximately one-seventh, than for existing straight-through MC devices (Kobayashi et al. 2002). The d_{av} of the droplets generated using TMS11 plates depended significantly on the channel size, gradually increasing with an increase in $d_{eq,channel}$ (Fig. 7d). Those d_{av} values exceeded the channel

shorter line by 2.9–4.4 times and $d_{eq,channel}$ by 1.8–2.6 times, which is analogous to those for existing straight-through MC devices (Kobayashi et al. 2002, 2003, 2004a). Therefore, the results reported in this section demonstrate that hydrophilic TMS11 plates are capable of stably generating monodisperse O/W emulsion droplets several microns in size.

The energy efficiency of emulsification using the TMS11 plates was also investigated in this research. It was possible to generate fine oil droplets from the outlet of oblong channels in TMS11 plates at the low P_d values presented in Table 3. The energy input for this droplet generation can be estimated by the following equation:

$$\Delta E_{in, exp} = P_d/\rho_d \tag{2}$$

where $\Delta E_{in,exp}$ is the actual energy input for the unit to-be-dispersed phase necessary for droplet generation from the outlet of oblong channels and ρ_d is the density of the to-be-dispersed phase. The calculated $\Delta E_{in,exp}$ was only 5.0–14.2 J/kg (Table 3) and gradually increased with an decrease in channel size. The energy input to be applied to the to-be-dispersed phase must exceed the theoretical minimum energy input calculated from the following equation to generate emulsion droplets (Walstra 1983; McClements 2004)

$$\Delta E_{in, min} = \Delta A \gamma_{ow} = 6\gamma_{ow}/\rho_d d_{dr} \tag{3}$$

where $\Delta E_{in,min}$ is the theoretical minimum energy input, ΔA is the increase in the interface area, γ_{ow} is the interfacial tension between the two phases, and d_{dr} is the droplet diameter. The $\Delta E_{in,min}$ at 25°C, for which calculation the d_{av} values obtained in this study was used as d_{dr}, ranged between 3.0 and 6.7 J/kg (Table 3). The apparent energy efficiencies of emulsion droplet generation using TMS11 plates, defined as $(\Delta E_{in,min}/\Delta E_{in,exp}) \times 100$, were 47–60% (Table 3). These calculated energy efficiencies, particularly for TMS11-2 and -3, are comparable to that of emulsification using a grooved MC device, which was reported to be 65% (Sugiura et al. 2001a). The estimated apparent maximum energy efficiency in droplet generation using a T-shape microchannel was about 10% (e.g. Nishisako et al. 2002). The energy loss in the above methods was due primarily to the pressure drop inside the channel for the to-be-dispersed phase, which is proportional to the channel length. The channel length (or depth) in MC emulsification devices is usually much shorter than the channel length in a T-shape microchannel. It is considered that the lower pressure drop inside the oblong channels in the TMS11 plates led to the higher apparent energy efficiency. In contrast, the apparent energy efficiency of a high-pressure homogenizer was significantly

Fig. 7 Droplet diameter
distributions of O/W emulsions
formulated using TMS11-1
a, TMS11-2 b, and TMS11-3
c. d Effect of the channel
equivalent diameter ($d_{eq,channel}$)
on the average droplet diameter
(d_{av}) and coefficient of variation
(CV) of the O/W emulsions
formulated using TMS11 plates
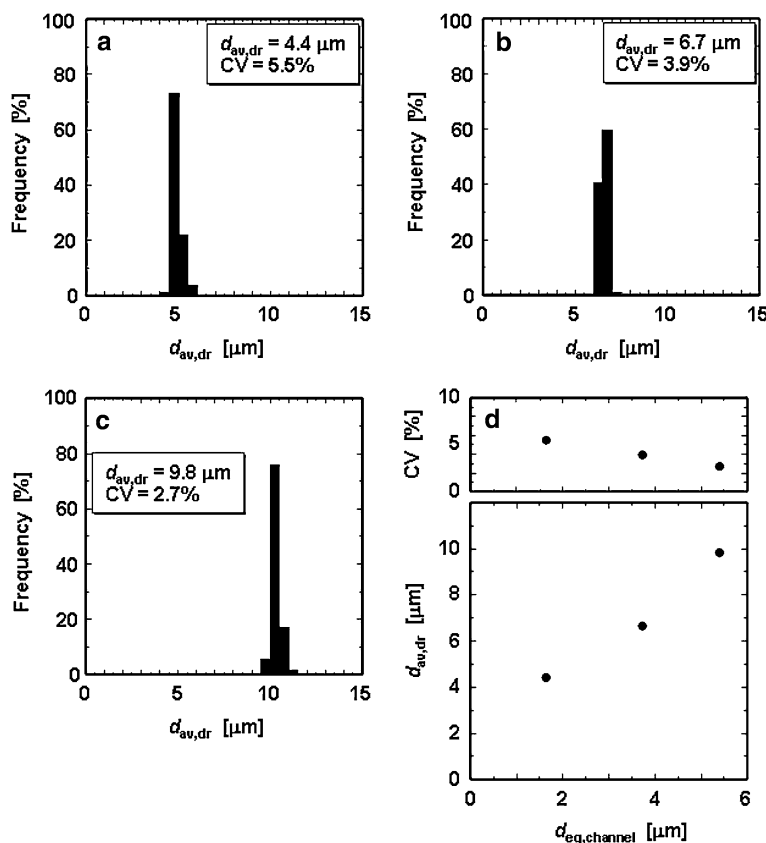
low with a typical value of less than 0.2%, since most of
the energy supplied is converted into heat (Gijsbertsen-
Abrahamse 2003). A comparison of the above apparent
energy efficiencies indicated the considerable efficiency of
emulsification using straight-through MC devices.

Monodisperse O/W emulsions with a d_{av} of approxi-
mately 7 μm were successfully formulated at a maximum
flow rate of the to-be-dispersed phase of 5×10^{-2} mL/h,
corresponding to a droplet generation rate of $1.8 \times 10^4 \, s^{-1}$,
when a syringe pump to supply the to-be-dispersed phase
was used for the emulsification experiments using a
TMS11-2 plate. Our previous paper demonstrated that the
droplet production rate of each channel in a straight-
through MC increases with decreases in the to-be-dispersed
phase viscosity (Kobayashi et al. 2005). While soybean oil
was used as the model to-be-dispersed phase in this study,
it is believed that the use of low-viscosity oils (e.g. alkanes)
as the to-be-dispersed phase can lead to enhanced emulsion
droplet productivity from a straight-through MC device.

### 4.3 Production of a W/O emulsion using a straight-through MC device

As schematically illustrated in Fig. 8a, the generation of
W/O emulsion droplets was carried out using a hydro-
phobic straight-through MC. The compositions of the two

phases in this case are presented in Table 1. Figure 8b
depicts a typical example of production of a W/O emulsion
using a hydrophobic TMS11-2 plate. Monodisperse water
droplets with a d_{av} of 7.1 μm and a CV of 2.8% were stably
generated from the outlet of the oblong channels at a P_d of
2.7 kPa. The droplet size distribution presented in Fig. 8c
confirms their narrow size distribution. The average droplet
generation rate at active channels was 1.1 s^{-1}. The optical
micrograph in Fig. 8b also shows that the generated water
droplets formed patterns consisting of jointed droplets,
whereas some of the generated water droplets stuck to the
channel outlet and were pushed out by the following
droplets with no droplet coalescence. It is presumed that
the droplet adhesion to the channel outlet was caused by
the weak repulsion between the water droplets and the plate
surface. Thus, the results obtained in this section demon-
strate that the hydrophobic straight-through MC device
(TMS11-2) was capable of generating monodisperse W/O
emulsion droplets several microns in size.

5 Conclusions

Monodisperse emulsions with fine droplets several microns
in size were successfully formulated using straight-through
MC devices developed in this research. The microfabri-

Fig. 8 a Schematic illustration of production of a W/O emulsion from the outlet of oblong channels. **b** Optical micrograph of successful production of a W/O emulsion using a hydrophobic TMS11-2 plate. The flow velocity of the continuous phase along the plate surface was 0.83 mm/s. **c** Droplet diameter distribution of the W/O emulsion produced using the TMS11-2 plate
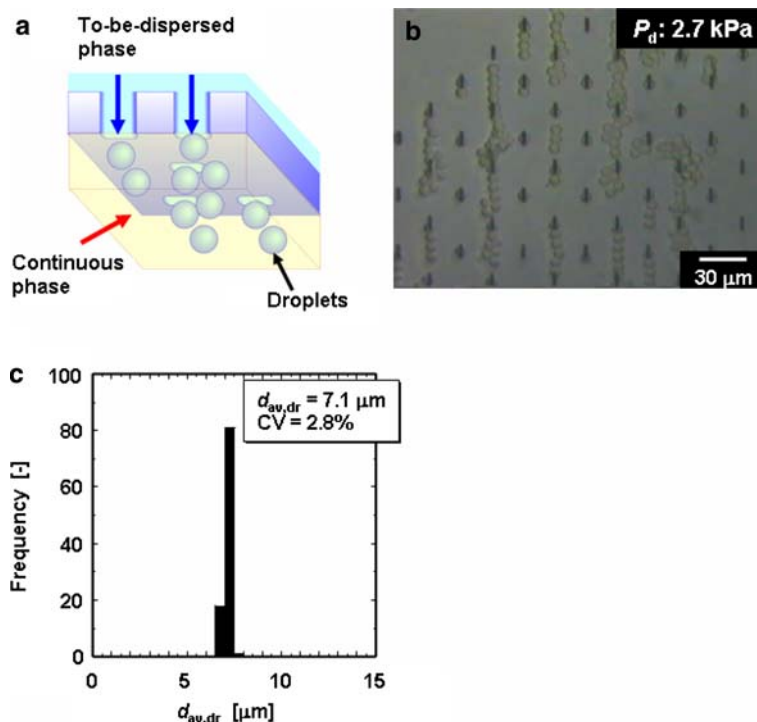
cation process including DRIE was successfully used to fabricate numerous oblong channels several micron in size in straight-through MC devices. Microscopic observations also demonstrated that the microfabricated deep oblong channels had very narrow size distributions and were regularly positioned in the channel area. Hydrophilic and hydrophobic TMS 11 plates were successfully used to generate monodisperse O/W and W/O emulsion droplets several microns in size even without applying the forced flow of the continuous phase. The size of the emulsion droplets could be controlled by the size of the oblong channels in the TMS11 plates. The energy efficiency for the emulsification process using straight-through MC devices was significantly higher than that of other emulsification techniques. In addition, straight-through MC devices with lots of microfluidic oblong channels have great potential for increasing the productivity of monodisperse fine emulsion droplets. In further experiments it is foreseen to apply the device to the production of monodisperse microparticles and microcapsules.

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