

Production characteristics of oil-in-water emulsions using microchannel homogenisation

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Abstract

Microchannel homogenisation (MCH) is a highly efficient technique for producing uniform emulsions in a one-pass process. This involves passing premix emulsions through a microchannel (MC) array chip. The preparation of oil-in-water (O/W) emulsions using MCH has not been thoroughly explored compared to other premix emulsification methods. Therefore, in the present work, we aimed to clarify the production characteristics and productivity of O/W emulsions in a one-pass process using MCH with an asymmetric straight-through MC array. Numerous asymmetric straight-through MCs, each comprising a microslot and circular microhole (diameter: 10 μm), were microfabricated on a silicon-on-insulator chip. The continuous phase was a Milli-Q water solution containing 9.0% (w/w) polyethylene glycol and 1.0% (w/w) polyoxyethylene 20 sorbitan monolaurate. The dispersed phase comprised 10 - 30% (v/v) soybean oil or medium-chain triacylglycerol oil. The volume-length mean diameter ($d_{4,3}$) of the O/W emulsions prepared *via* MCH was approximately 15 μm below the critical pressure. Emulsion fluxes reached $> 1.0 \text{ m}^3/(\text{m}^2 \text{ h})$ even with 30% (v/v) dispersed phase. These results demonstrated that MCH with an asymmetric MC array could stably prepare O/W emulsions with a high dispersed-phase concentration in a one-pass process. Furthermore, MCH achieved high throughput, comparable to that of premix membrane emulsification.

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Introduction

An emulsion is a dispersion of two or more immiscible liquids, such as water and oil, in which one liquid is dispersed as droplets or particles in the other. The process used to prepare such emulsion is called emulsification (McClements, 2015). The major types of emulsions include oil-in-water (O/W), water-in-oil (W/O), and double emulsions. O/W emulsions, such as milk, ice cream, and mayonnaise, are a part of our regular diet (Fredrick *et al.*, 2010). The droplet size and its distribution of food emulsions are important factors that affect the taste and storage stability of emulsified foods.

For the preparation of food emulsions, methods such as rotor-stator homogenisers, colloid mills, and high-pressure homogenisers have been used in the food industry. These methods disperse liquids in the dispersed phase (internal phase) as fine particles in the continuous phase (external phase) by applying strong mechanical shear. However, these emulsification methods are characterised by

difficulty in controlling droplet size (Håkansson, 2019). Therefore, controlling the droplet size and uniformity of the O/W emulsion is an important issue.

Research on the preparation of monodispersed emulsions has progressed over the past decade, and microfluidic emulsification, membrane emulsification (ME), and microchannel emulsification (MCE) methods have been developed (Vladislavljević *et al.*, 2012). In microfluidic emulsification systems, the dispersed and continuous phases flow through channels, mainly as liquids, forming droplets at the intersections of channels, such as T-junctions and microchannel systems (Maan *et al.*, 2015). ME using Shirasu porous glass (SPG) membranes was developed by Nakashima *et al.* (2000). Direct ME is a technique for producing monodispersed emulsions by injecting the dispersed phase under pressure through a microporous membrane into the continuous phase (Piacentini *et al.*, 2014). The main feature of direct ME is that the droplet diameter of the emulsion can be controlled by selecting the pore size of the porous membrane.

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However, the most commonly used SPG membranes with a labyrinthine internal structure require several cleaning operations, including ultrasonic cleaning with solvent. Moreover, membrane regeneration processes including heat cleaning with a muffle furnace are also required when the SPG membrane surface becomes less hydrophilic with repeated use. These operations are time-consuming, and have may damage the membrane devices.

MCE using a microchannel (MC) array chip enables the preparation of emulsions with precisely controlled droplet sizes (Sugiura *et al.*, 2001; Kobayashi *et al.*, 2008). Based on the microfabrication design, MC arrays can be classified as grooved MC array chips, consisting of uniform microgrooves and slit-like terraces, or straight-through MC array chips with microholes and microslots (Vladisavljević *et al.*, 2012). Grooved MC array plates are further classified into dead-end and cross-flow types. Cross-flow type grooved MC arrays are capable of producing monodispersed emulsions at low dispersed phase flow rates, and the droplet production process can be observed (Kobayashi *et al.*, 2010). Straight-through MC arrays are classified into symmetric and asymmetric MC array plates. Asymmetric MC array plates enable the stable production of uniform droplets of low-viscosity oil (Vladisavljević *et al.*, 2011). Recent MCE studies reported the development of a variety of monodisperse emulsions, including functional emulsions (Ma *et al.*, 2020) and structured lipid microparticles (Wang *et al.*, 2022). However, as the productivity of the monodisperse emulsions in MCE is still low, parallelisation and/or scale-up of the MCE devices are needed to achieve higher productivity.

Premix membrane emulsification (PME) is an emulsification method in which coarse emulsions are emulsified into fine droplets in a porous structure (Nazir and Vladisavljević, 2021). Surh *et al.* (2008) reported that PME using SPG membrane can prepare emulsions at high production rates (*e.g.*, $> 1.0 \text{ m}^3/(\text{m}^2 \text{ h})$). Monodisperse emulsions are difficult to obtain in a single emulsification process using PME, and have required multiple emulsification treatments (Vladisavljević *et al.*, 2006). However, Mugabi *et al.* (2021) recently showed that monodispersed O/W emulsions can be prepared in one pass by PME using SPG membranes. They reported that a considerably high membrane permeability ($10 - 200 \text{ m}^3/(\text{m}^2 \text{ h})$) was required as a preferred condition to obtain monodisperse emulsions by a single treatment. In this

technique, extremely high shear stress is applied when the premix emulsion passes through the SPG membrane. Therefore, when applying the single emulsification process using PME to food products, the nutritional quality of components may deteriorate due to high shear stress.

Our research group also proposed MC homogenisation (MCH) using an MC array, which can enable one-pass preparation of monodispersed emulsions. In a conventional MCE setup, the dispersed phase (oil) is delivered to the MC module using a syringe pump or a tank when preparing O/W emulsions. In contrast, in MCH, the premix emulsion in the supply vessel is delivered to the module using air pressure, allowing for simpler emulsion preparation. The O/W emulsions obtained after the one-pass operation were relatively monodispersed, with a coefficient of variation (CV) below 20% (Kobayashi *et al.*, 2008).

MCH with an asymmetric straight-through MC array can achieve the high throughput preparation of O/W emulsions with uniform droplet sizes in a single emulsification process. Vladisavljević *et al.* (2011) reported that the viscosity of the dispersed phases affected the throughput of O/W emulsion prepared *via* MCE using asymmetric MC array plate. Furthermore, Li *et al.* (2023) reported that in the production characteristics of W/O emulsions using MCH, the operating pressure affected the droplet size and flux of the W/O emulsion. Therefore, the composition and operating conditions may be important factors in the production characteristics of O/W emulsions using MCH, whereas there is little knowledge about them. Therefore, in the present work, we investigated the effects of the type and volume fraction of the dispersed phase on the particle size distribution, flux, and viscosity of the O/W emulsions using MCH with an asymmetric straight-through MC array.

Materials and methods

Chemicals

Refined soybean oil (SO), polyoxyethylene 20 sorbitan monolaurate (Tween 20), and polyethylene glycol (M.W. 20,000, PEG) were purchased from FUJIFILM Wako Pure Chemical Co. (Osaka, Japan). Medium-chain triglyceride oil (MCT, SUNOIL MCT-7) was purchased from Taiyo Kagaku Co., Ltd. (Yokkaichi, Japan).

MCH setup

Asymmetric MC array chip

Emulsification experiments were conducted using an asymmetric straight-through MC array chip (Model 11-1, EP Tec. Co., Ltd., Hitachi, Japan) with different shapes on the inlet and outlet sides (Figure 1a). The size of the MC array chip was 24×24 mm, and contained 13,824 MCs in an active area of approximately 100 mm^2 in which asymmetric MCs were arranged. Each asymmetric MC microfabricated on a silicon-on-insulator (SOI) chip comprised a $10 \times 80 \mu\text{m}$ microslot with a depth of $40 \mu\text{m}$, and cylindrical straight microholes with a diameter (d_{MC})

of $10 \mu\text{m}$ and a depth of $160 \mu\text{m}$. To increase the strength of the silicon MC array, a $300 \mu\text{m}$ supporting quartz glass layer was anodically bonded to the microfabricated SOI chip.

Before starting the emulsification experiments, the MC plates were plasma-oxidized using a plasma exciter (PR500, Yamato Science Co., Ltd., Tokyo, Japan) for 15 min. Plasma oxidation treatment forms a hydrophilic layer on the surface of the MC array. After each experiment, the MC array was cleaned using an ultrasonic bath (VS-100 III, As One Co.) at a frequency of 100 kHz for 20 min in a mixture of neutral detergent, ethanol, and Milli-Q water.

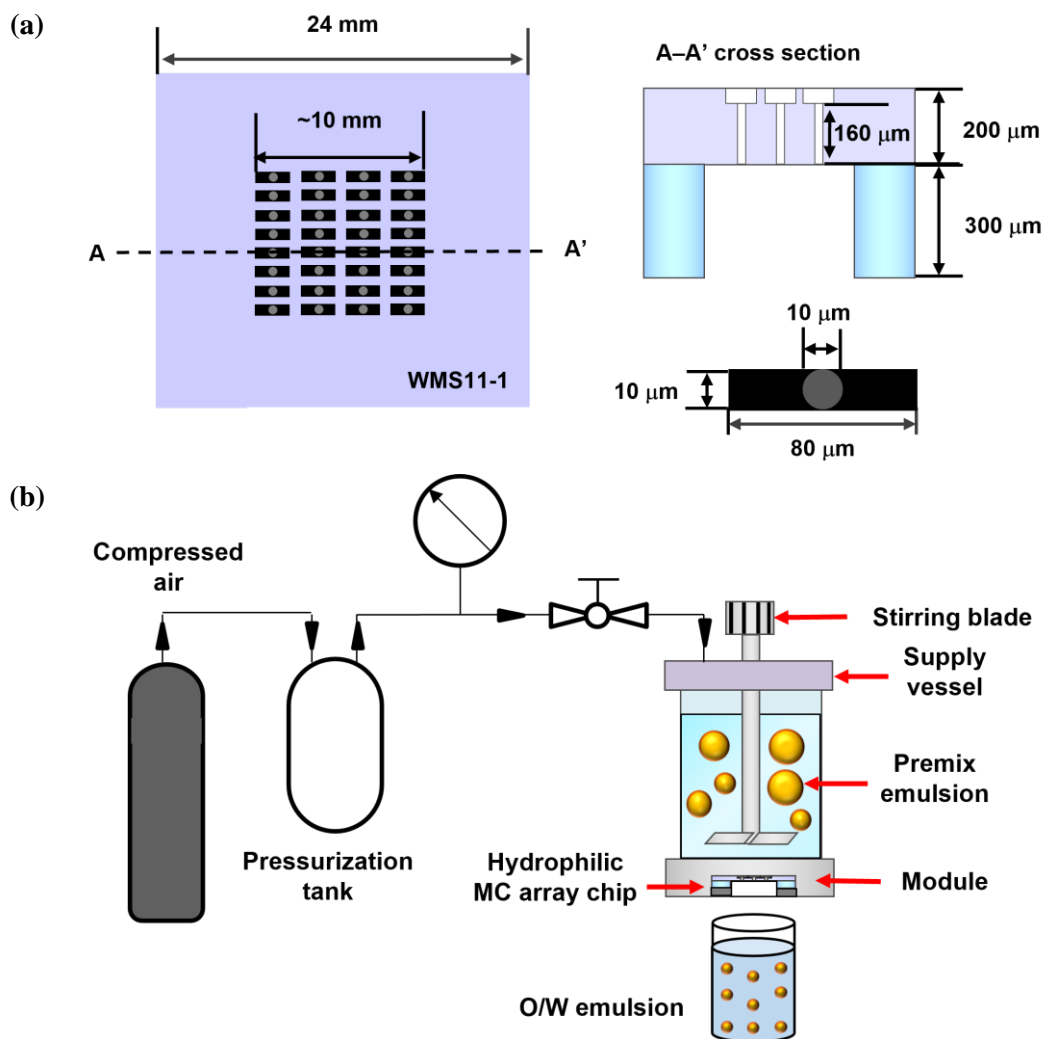


Figure 1. (a) Schematic representation of asymmetric straight-through MC array chip (model WMS11-1). (b) Schematic representation of MCH setup with asymmetric straight-through MC array chip. MC: microchannel; and MCH: microchannel homogenisation.

Microchannel homogeniser

The microchannel homogeniser consisted of an air cylinder, a 5 L pressurisation tank, a pressure gauge, a 100 mL feed vessel with stirring blades, and an emulsification module (Figure 1b). The feed vessel

with stirring blades was connected by a belt to a mechanical stirrer (Eurostar 40 digital, IKA-Werke GmbH and Co., Staufen, Germany), which could mix the premix emulsion, even when sealed. During the emulsification process, the stirring blades were

rotated to prevent the oil phase of the premix from creaming. The supply vessel was connected to a pressurisation tank filled with air gas, which was then pressurised into a module equipped with an MC array. The pressurised air gas was controlled using a pressure gauge.

Formulation of continuous and dispersed phases

The continuous phase was prepared with 9.0% (w/w) PEG as a thickener. In addition, 1.0% (w/w) Tween 20 was used as an emulsifier, and formulated with Milli-Q water. For the dispersed phase, SO and MCT with a dispersed phase volume fraction (ϕ_d) of 10 - 30% (v/v) were used.

Emulsification procedure

The MC array plate was degassed in the continuous phase using an ultrasonic bath at a frequency of 100 kHz for 20 min. Subsequently, the MC array was placed in the module.

The premix emulsion was prepared by placing the continuous phase in an Erlenmeyer flask containing the dispersed phase, and by stirring them (900 rpm, 1 min) using a magnetic stirrer. The prepared premix emulsion was immediately placed into the feeding vessel in its entire volume. The emulsification process was performed under pressure, and the container was sealed. The stirring blade was rotated at 200 rpm during the experiment.

Measurements and analysis

The throughput of emulsions prepared by MCH was calculated using Eq. 1:

$$Q_e = W_e/t \quad (\text{Eq. 1})$$

where W_e = weight of the O/W emulsion, and t = emulsification time. The emulsion flux J_e was calculated using Eq. 2:

$$J_e = Q_e/A_{MCA} \quad (\text{Eq. 2})$$

where A_{MCA} = total area of the MC array.

The O/W emulsion droplets were analysed using a laser light-scattering size analyser (Beckman Coulter LS 13 320, Miami, USA). The mean droplet size was expressed in terms of the volume-length mean diameter ($d_{4,3}$), and determined using Eq. 3:

$$d_{4,3} = \sum n_i d_i^4 / \sum n_i d_i^3 \quad (\text{Eq. 3})$$

where d_i = diameter of the i -th droplet measured, and n = number of droplets measured ($n = 100$).

The droplet size distribution of the O/W emulsion was presented as the relative span factor (RSF), and determined by Eq. 4:

$$RSF = (d_{90} - d_{10})/d_{50} \quad (\text{Eq. 4})$$

The particle size distribution was measured four times for each condition.

The O/W emulsion using MCH was observed using an optical microscope (DM-IRM Leica Microsystems, Weizler, Germany).

The viscosity of the O/W emulsions was measured using a viscometer (Brookfield DV-II+ Pro, Middleboro, USA). Briefly, 6.7 mL of O/W emulsion was put into the chamber, and the spindle (SC4-18; diameter: 17.5 mm, height: 31.7 mm) was rotated at 1 - 40 rpm. The viscosity value was obtained and recorded 30 s after the spindle was rotated. As emulsion viscosity is sensitive to temperature, a water circulating thermostatic bath was used to maintain the emulsion temperature at 25 °C. Viscosity measurements were performed in triplicate.

Results and discussion

Effect of operating pressure and type of dispersed phase

The effects of the operating pressure (ΔP) and the type of dispersed phase on the particle size distribution of O/W emulsions produced by MCH were investigated. The dispersed phase was either 10% (v/v) SO or MCT. When the dispersed phase was 10% (v/v) SO, the $d_{4,3}$ of the emulsions under the ΔP of 10 - 30 kPa was 14.2 - 16.9 μm , and remained almost constant regardless of the ΔP (Figure 2a). The ratio of the emulsions to the microhole diameter ($d_{4,3}/d_{MC}$) was approximately 1.5 at ΔP below 30 kPa. In contrast, $d_{4,3}$ at 40 kPa increased to 23.1 μm , and $d_{4,3}/d_{MC}$ was approximately 2.3. Li *et al.* (2023) reported that above a critical operating pressure, the droplet size of W/O emulsions prepared *via* MCH with the same MC array model (WMS11-1) increased. This correlated well with that obtained in the present work.

Direct observation of oil-droplet breakup processes *via* a silicon MC array revealed that premix emulsions began to penetrate into the MC at a very

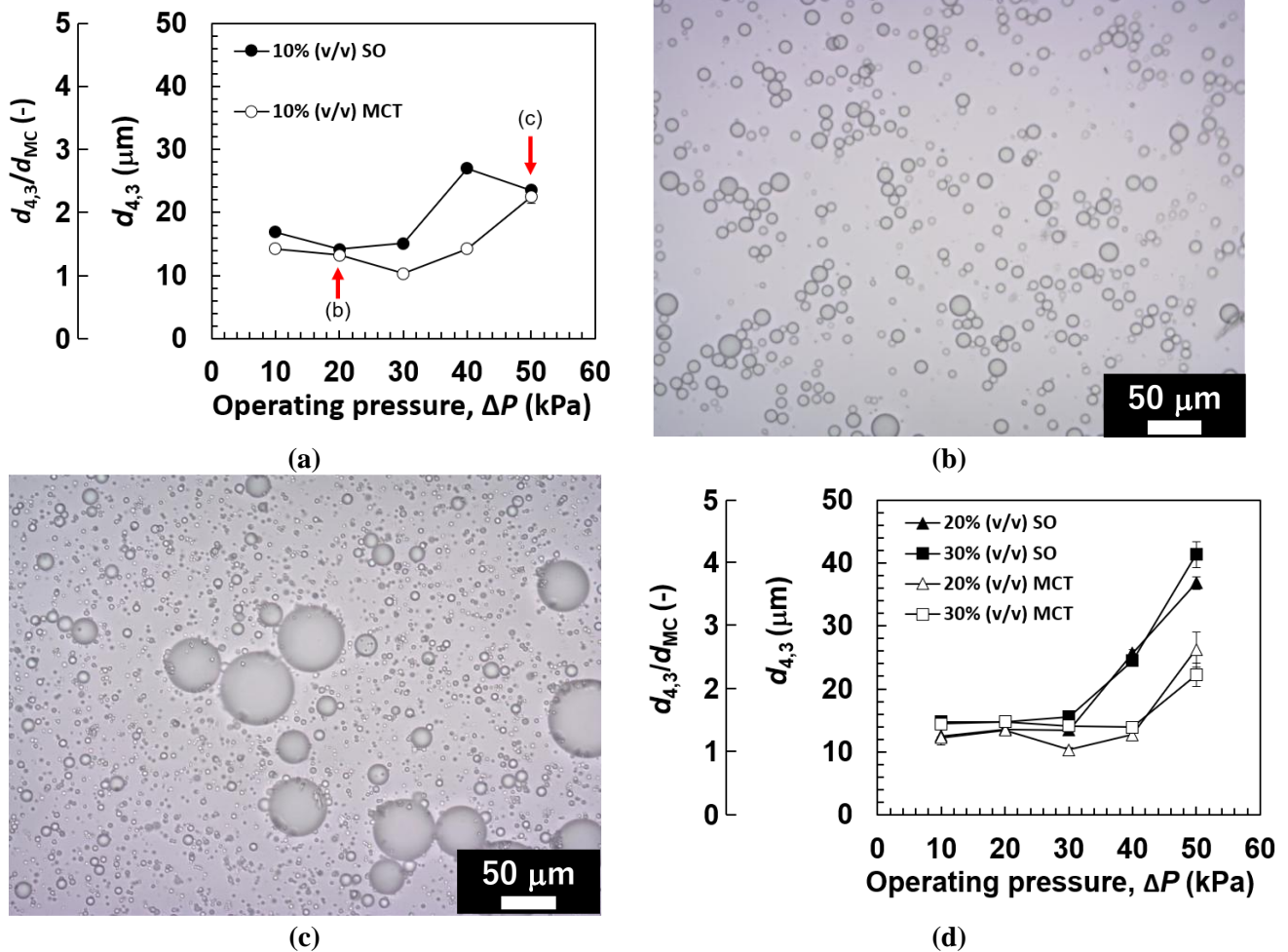


Figure 2. (a) Effect of ΔP and type of dispersed phase on $d_{4,3}$ of O/W emulsions prepared via MCH. (●): 10% (v/v) SO; and (○): 10% (v/v) MCT. (b) Optical micrograph of 10% (v/v) MCT oil O/W emulsion prepared via MCH at 20 kPa, and (c) 50 kPa. (d) Effect of volume fraction of dispersed phase on $d_{4,3}$ of O/W emulsions prepared via MCH. (▲): 20% (v/v) SO; (■): 30% (v/v) SO; (△): 20% (v/v) MCT; and (□): 30% (v/v) MCT. ΔP : operating pressure; $d_{4,3}$: average diameter; O/W: oil-in-water; SO: soybean oil; and MCT: medium-chain triglyceride oil.

low operating pressure, and were broken up in the MC within 3 ms (Kobayashi *et al.*, 2008). Furthermore, droplet breakup processes by MCE leads to a rapid increase in emulsion droplet diameter at conditions above a critical flow velocity (Sugiura *et al.*, 2002). Although the asymmetric straight-through MC used in the present work did not allow direct observation of the breakup behaviour, the premix emulsions passed through the MCs without elongation or splitting above the critical pressure (Figure 3).

When the dispersed phase was 10% (v/v) MCT, the $d_{4,3}$ of the O/W emulsion prepared at ΔP of 20 kPa was 13.3 μm , and the *RSF* value was 1.8 (Figures 2a and 2b). The major peak of the emulsion was at 10 μm , but a second low peak was identified

at around 30 μm . The *RSF* value for the particle size distribution expressed in terms of volume was large because the *RSF* was affected more by the larger droplet size. The $d_{4,3}$ of emulsions at ΔP of 50 kPa increased to 23.1 μm , and microscopic observations confirmed that some droplets were larger than 50 μm (Figures 2a and 2c). Thus, the critical pressure of the MCT was 40 kPa, which depends on the type of dispersed phase. Below the critical pressure, the droplet size of the prepared emulsions did not vary with the type of dispersed phase. In contrast, the critical pressure threshold was higher for MCT than for SO. This result might have been influenced by the viscosity of the dispersed phase and the interfacial tension. Several studies on droplet refinement simulations in premix membrane emulsification have

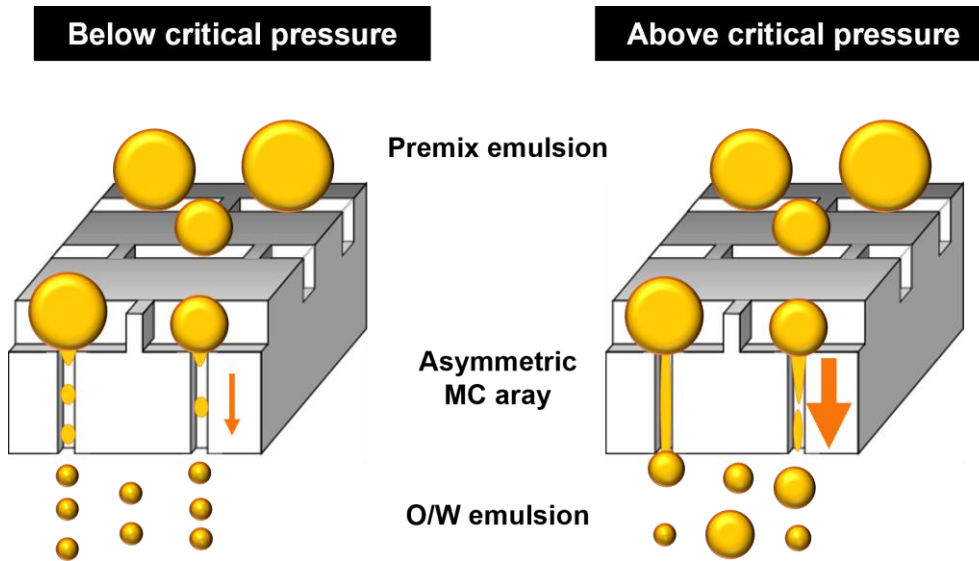


Figure 3. Schematic of mechanism of droplet refining as premix emulsion passes through asymmetric straight-through MC array. Premix emulsion passes through MC array without breakup when operating pressure is above critical pressure.

analysed the processes of droplet deformation, breakup, and permeation within pores using the capillary number (Ca) (van der Zwan *et al.*, 2006; Wollborn *et al.*, 2019). Ca is a dimensionless number representing the ratio of viscous forces to interfacial tension, and determined using Eq. 5:

$$Ca = \eta_d U / \gamma \quad (\text{Eq. 5})$$

where η_d = viscosity of the dispersed phase, U = velocity of the liquid, and γ = interfacial tension. From this equation, the balance between dispersed phase viscosity and interfacial tension is one of the important physical properties considering premix emulsion breakup behaviour in MCH. The interfacial tensions (γ_{SO} , γ_{MCT}) of the continuous phase and dispersed phase used in the present work were 5.0 and 5.9 mN/m, respectively, whereas η_{SO} was more than twice that of η_{MCT} . Thus, the dispersed phase viscosity had greater influence than the interfacial tension, resulting in the penetration behaviour of coarse droplets into the MC, and the elongation and breakup behaviour in the MC. This suggested that the critical pressure thresholds depended on the type of dispersed phase.

Effect of volume fraction of dispersed phase

The effect of ϕ_d on the particle size distribution of the O/W emulsions prepared by MCH was investigated (Figure 2d). Regardless of the increase in ϕ_d , the critical pressures of SO and MCT

were 30 and 40 kPa, respectively. The $d_{4,3}/d_{MC}$ below the critical pressure was approximately 1.5. This indicated that MCH with asymmetric straight-through MC arrays could produce O/W emulsions with ϕ_d of 10 - 30% (v/v) in a single pass at conditions below the critical pressure.

Under constant operating conditions, the average droplet size of emulsions produced by PME did not change (1 - 60% (v/v)) with varying dispersed phase concentration (Vladislavljević *et al.*, 2004). This was because the average diameter of the emulsions produced by PME is mainly determined by the average pore size and the pore shear stress. The shear stress in the pores is determined by the balance between emulsion viscosity and flux. As the dispersed phase concentration increases, the emulsion viscosity increases, and the flux decreases. Therefore, under certain experimental conditions, the shear stress in the pore may not affect the concentration of the dispersed phase. In contrast, in high-pressure emulsification, the droplet size depends on the proportion of the dispersed phase under certain operating conditions (Brösel and Schubert, 1999). This is because the surface area produced as it passes through the device is not covered by the surfactant in a timely manner, making the produced emulsion unstable.

O/W emulsions prepared *via* MCH were not affected by changes in the volume fraction of the dispersed phase (10 - 30% (v/v)) below the critical pressure. Our results were similar to those obtained

with PME, suggesting that below the critical pressure, the shear stress in the MC was independent of the increase in the volume fraction of the dispersed phase.

Emulsion flux

The effects of the dispersed phase type and volume fraction on J_e were investigated. The J_e of the emulsions at ϕ_d 10% (v/v) increased linearly with increasing ΔP , regardless of the type of emulsion (Figure 4a). Altenbach-Rehm *et al.* (2002) found that in PME with polytetrafluoroethylene (PTFE) membranes, the permeate flux increased almost linearly with pressure difference when the dispersed phase concentration was 10 - 30% (v/v). Our results were consistent with these findings. There was a strong positive correlation observed between ΔP and J_e , where in SO emulsion, the linear approximation yielded,

$$J_e = 0.085 \Delta P \quad (R^2 = 0.98), \quad (\text{Eq. 6})$$

and in MCT,

$$J_e = 0.082 \Delta P \quad (R^2 = 0.99). \quad (\text{Eq. 7})$$

At the ϕ_d 10% (v/v), the flux of the emulsion at critical pressure reached $1.9 \text{ m}^3/(\text{m}^2 \text{ h})$ for SO and $3.4 \text{ m}^3/(\text{m}^2 \text{ h})$ for MCT. The slope of the approximate straight line between SO and MCT did not change, suggesting that the permeate velocity of the emulsion was affected by the operating pressure, rather than the type of dispersed phase.

At higher ϕ_d of 20% (v/v) and 30% (v/v), J_e increased linearly with increasing ΔP . However, the J_e decreased as ϕ_d increased (Figure 4b). At ΔP of 20 kPa, J_e for SO emulsion was $1.4 \text{ m}^3/(\text{m}^2 \text{ h})$ at ϕ_d 20% (v/v), and $1.3 \text{ m}^3/(\text{m}^2 \text{ h})$ at 30% (v/v), corresponding to a decrease in flux of 78.8 and 72.9%, respectively, compared to ϕ_d at 10% (v/v). Meanwhile, for MCT emulsion at ϕ_d of 20% (v/v) and 30% (v/v), the J_e values at ΔP of 20 kPa were 1.2 and $1.0 \text{ m}^3/(\text{m}^2 \text{ h})$, respectively. This showed that flux decrease rates were 86.9 and 77.3% compared to ϕ_d at 10% (v/v). The decrease rate in flux with increasing dispersed phase for MCT was lower than that for SO, which was likely due to the higher viscosity of SO as the dispersed phase. We considered that this resulted in a higher increase in viscosity of premix emulsion as the dispersed phase volume fraction increased. In a previous study, multiple water-in-oil-in-water (W/O/W) emulsions were prepared by PME with

SPG membranes, and as the droplet concentration increased, the transmembrane flux was reported to decrease (Vladislavljević *et al.*, 2011). These findings are consistent with those observed in the present study. Emulsion fluxes at critical pressure achieved $1.8 \text{ m}^3/(\text{m}^2 \text{ h})$ for 30% (v/v) SO, and $2.6 \text{ m}^3/(\text{m}^2 \text{ h})$ for 30% (v/v) MCT. Among the emulsification techniques that use microfabricated devices, premix membrane emulsification is known to be highly productive (Nazir and Vladislavljević, 2021). The flux of premix membrane emulsification was shown to be $> 1.0 \text{ m}^3/(\text{m}^2 \text{ h})$ (Surh *et al.*, 2008). Thus, the results obtained in the present work were of the same order as those obtained in previous studies, indicating that the throughput of MCH is comparable to that of premix membrane emulsification.

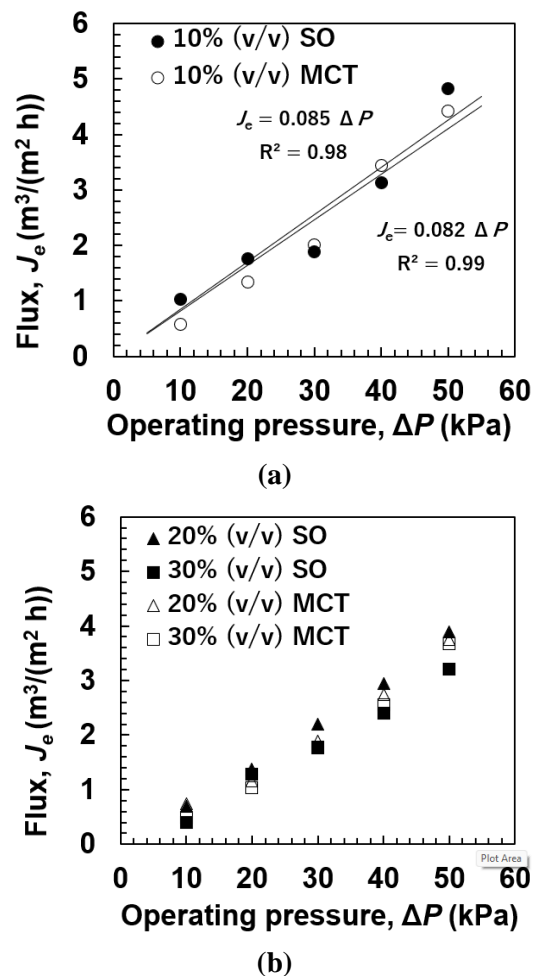


Figure 4. Effect of ΔP on J_e of O/W emulsions prepared via MCH. (a) 10% (v/v) SO and MCT. (b) 20 - 30% (v/v) SO and MCT. J_e : flux of emulsion.

Measurement of fluid properties

The viscosity of emulsions (η_e) prepared via MCH at ΔP 30 kPa was measured using a rotational viscometer (Figures 5a and 5b). The shear rates

ranged from 1.3 to 52.8 s⁻¹ when the spindles were rotated at 1 - 40 rpm. The SO emulsion at ϕ_d of 10% (v/v) exhibited significant shear-thinning behavior, with decreasing viscosity from 39.0 to 13.8 mPa s within the low shear rate range from 1.3 to 6.6 s⁻¹. At the shear rate of 52.8 s⁻¹, η_e was 11.4 mPa s. In the shear rate range from 6.6 to 52.8 s⁻¹, the viscosity decreased gradually. This SO emulsion behaved as non-Newtonian fluid. As ϕ_d increased to 20% (v/v) and 30% (v/v), this behaviour was still observed, and the viscosity curve shifted towards higher viscosities. This can be attributed to the increased proportion of SO droplets, which exhibited a higher viscosity than the continuous phase, in conjunction with the packing density of the droplets within the emulsion. Moreover, the rate of increase in the emulsion viscosity at each shear rate was greater when the dispersed phase volume fraction was increased from

20% (v/v) to 30% (v/v) than when it was increased from 10% (v/v) to 20% (v/v).

The fluid properties of the MCT emulsions prepared *via* MCH were similar to those of SO emulsions. Although the dispersed phase viscosity of MCT was lower than that of SO, the viscosities of the MCT emulsions at the same ϕ_d were almost the same as or even higher than those of the SO emulsions. A possible reason for this result is the difference in particle size distribution between SO and MCT emulsions prepared *via* MCH. Particle size affects the viscosity characteristics of emulsions, with smaller droplets increasing the viscosity of the emulsion (Ramirez *et al.*, 2002). That is, the decrease in droplet diameter increases the droplet surface area. All emulsions prepared *via* MCH at ΔP 30 kPa showed a small second peak approximately 30 μm , with the proportion of this second peak being higher in the SO emulsions than in the MCT emulsions. This indicated that the SO emulsions prepared *via* MCH contained a higher percentage of larger droplets, resulting in lower viscosity.

Conclusion

MCH with an asymmetric straight-through MC array chip produced fine oil droplets with a dispersed phase concentration of up to 30% (v/v) in a single emulsification process. This indicated that there was a critical pressure above which polydispersed emulsions were produced, which was affected by the viscosity of the dispersed phase. The average droplet size produced by MCH was approximately 1.5 times larger than that of d_{MC} below the critical pressure, and was not affected by the type and concentration of the dispersed phase. These results demonstrated that MCH can be used for stable droplet production regardless of the type and concentration of oil, which is applicable to more emulsified food products.

The flux of the O/W emulsions, which represents droplet throughput, increased in proportion to ΔP , indicating that the throughput was comparable to that of PME, even for emulsions with higher volume fractions. The viscosity of the emulsions showed a Newtonian fluid trend.

The fundamental findings from this study could be applicable to the highly efficient preparation of emulsified food products with uniform droplet sizes.

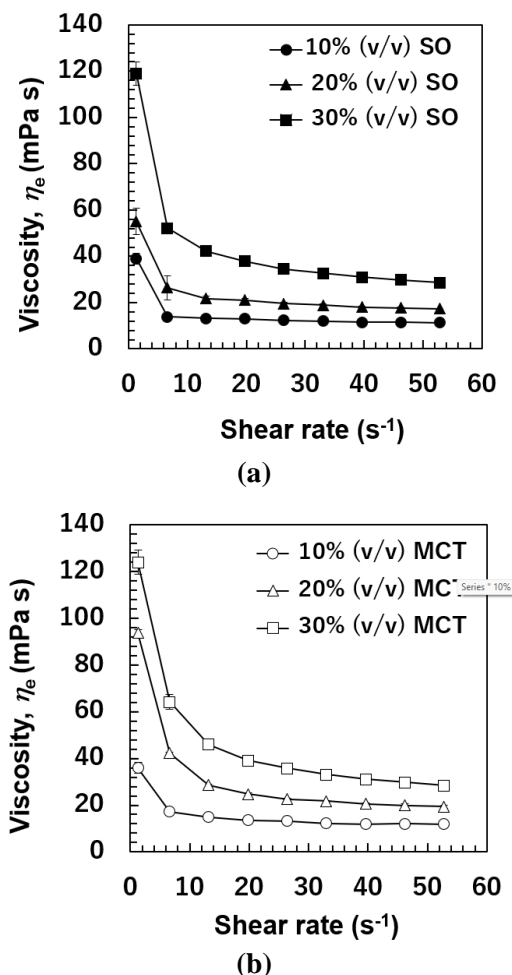


Figure 5. Effect of shear rate on viscosity of O/W emulsions prepared *via* MCH. (a) 10 - 30% (v/v) SO. (b) 10 - 30% (v/v) MCT. (Plots and error bars indicate mean and standard deviation of three repeated measurements, respectively.)

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References

- Altenbach-Rehm, J., Suzuki, K. and Schubert, H. 2002. Production of O/W-emulsions with narrow droplet size distribution by repeated premix membrane emulsification. In Proceedings of 3rd World Congress on Emulsions, p. 2-B-051. Lyon, France.
- Brösel, S. and Schubert, H. 1999. Investigations on the role of surfactants in mechanical emulsification using a high-pressure homogenizer with an orifice valve. *Chemical Engineering and Processing - Process Intensification* 38(4-6): 533-540.
- Fredrick, E., Walstra, P. and Dewettinck, K. 2010. Factors governing partial coalescence in oil-in-water emulsions. *Advances in Colloid and Interface Science* 153(1-2): 30-42.
- Håkansson, A. 2019. Emulsion formation by homogenization: Current understanding and future perspectives. *Annual Review of Food Science and Technology* 10: 239-258.
- Kobayashi, I., Uemura, K. and Nakajima, M. 2008. Breakup of oil-in-water emulsion droplets in microchannel array devices at low applied pressures. *Journal of Chemical Engineering of Japan* 41(5): 420-426.
- Kobayashi, I., Wada, Y., Uemura, K. and Nakajima, M. 2010. Microchannel emulsification for mass production of uniform fine droplets: Integration of microchannel arrays on a chip. *Microfluidics and Nanofluidics* 8(2): 255-262.
- Li, R., Kobayashi, I., Zhang, Y., Neves, M. A., Uemura, K. and Nakajima, M. 2023. Preparation of monodisperse water-in-oil emulsions using microchannel homogenization. *Particulate Science and Technology* 41(7): 930-939.
- Ma, Z., Zhao, Y., Khalid, N., Shu, G., Neves, M. A., Kobayashi, I. and Nakajima, M. 2020. Comparative study of oil-in-water emulsions encapsulating fucoxanthin formulated by microchannel emulsification and high-pressure homogenization. *Food Hydrocolloids* 108: 105977.
- Maan, A. A., Nazir, A., Khan, M. K. I., Boom, R. and Schroën, K. 2015. Microfluidic emulsification in food processing. *Journal of Food Engineering* 147: 1-7.
- McClements, D. J. 2015. *Food emulsions - Principles, practices, and techniques*. United States: CRC Press.
- Mugabi, J., Jeong, J. H., Igura, N. and Shimoda, M. 2021. Preparation of monodispersed emulsions by premix membrane emulsification without repetitive permeation: Influence of membrane permeation rate (flux) and emulsion viscosity. *Colloids and Surfaces A - Physicochemical and Engineering Aspects* 630: 127560.
- Nakashima, T., Shimizu, M. and Kukizaki, M. 2000. Particle control of emulsion by membrane emulsification and its applications. *Advanced Drug Delivery Reviews* 45 (1): 47-56.
- Nazir, A. and Vladisavljević, G. T. 2021. Droplet breakup mechanisms in premix membrane emulsification and related microfluidic channels. *Advances in Colloid and Interface Science* 290: 102393.
- Piacentini, E., Drioli, E. and Giorno, L. 2014. Membrane emulsification technology: Twenty-five years of inventions and research through patent survey. *Journal of Membrane Science* 468: 410-422.
- Rámirez, M., Bullón, J., Andérez, J., Mira, I. and Salager, J. L. 2002. Drop size distribution bimodality and its effect on O/W emulsion viscosity. *Journal of Dispersion Science and Technology* 23(1-3): 309-321.
- Sugiura, S., Nakajima, M. and Seki, M. 2002. Effect of channel structure on microchannel emulsification. *Langmuir* 18(15): 5708-5712.
- Sugiura, S., Nakajima, M., Ushijima, H., Yamamoto, K. and Seki, M. 2001. Preparation characteristics of monodispersed water-in-oil emulsions using microchannel emulsification. *Journal of Chemical Engineering of Japan* 34(6): 757-765.
- Surh, J., Jeong, Y. G. and Vladisavljević, G. T. 2008. On the preparation of lecithin-stabilized oil-in-water emulsions by multi-stage premix membrane emulsification. *Journal of Food Engineering* 89(2): 164-170.
- van der Zwan, E., Schroën, K., van Dijke, K. and Boom, R. 2006. Visualization of droplet breakup in pre-mix membrane emulsification using microfluidic devices. *Colloids and Surfaces*

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- Vladisavljević, G. T., Kobayashi, I. and Nakajima, M. 2011. Effect of dispersed phase viscosity on maximum droplet generation frequency in microchannel emulsification using asymmetric straight-through channels. *Microfluidics and Nanofluidics* 10(6): 1199-1209.
- Vladisavljević, G. T., Kobayashi, I. and Nakajima, M. 2012. Production of uniform droplets using membrane, microchannel and microfluidic emulsification devices. *Microfluidics and Nanofluidics* 13(1): 151-178.
- Vladisavljević, G. T., Shimizu, M. and Nakashima, T. 2004. Preparation of monodisperse multiple emulsions at high production rates by multi-stage premix membrane emulsification. *Journal of Membrane Science* 244(1-2): 97-106.
- Vladisavljević, G. T., Surh, J. and McClements, J. D. 2006. Effect of emulsifier type on droplet disruption in repeated Shirasu porous glass membrane homogenization. *Langmuir* 22(10): 4526-4533.
- Wang, H., Nakajima, M., Neves, M. A., Uemura, K., Todoriki, S. and Kobayashi, I. 2022. Formulation characteristics of monodisperse structured lipid microparticles using microchannel emulsification. *Particulate Science and Technology* 40(2): 196-206.
- Wollborn, T., Luhede, L. and Fritsching, U. 2019. Evaluating interfacial shear and strain stress during droplet deformation in micro-pores. *Physics of Fluids* 31(1): 012109.