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Relationship between the continuous phase viscosity and the membrane permeation rate in premix membrane emulsification using Shirasu porous glass membranes

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ABSTRACT

Emulsion viscosity and membrane permeation rate (MPR) are the major factors influencing the wall shear stresses in the membrane pores. However, the effect of the discrete phase viscosities and their relationship with the MPR is not well elaborated. The relationship between droplet size and the continuous phase viscosity at different MPRs was investigated. At higher MPR, the emulsion droplet size was observed to decrease with continuous phase viscosity due to the increase in the wall shear stress in the pores, as the thickness of the lubrication layer between the droplets and the pore walls increases with viscosity. While at low MPR, the droplet size increased with viscosity due to coalescing as the rate of surfactant diffusion onto the interface of the new droplets reduced. The MPR was observed to complement or contrast the effects of the continuous phase viscosity on droplet formation in the membrane pores, depending on its relative magnitude.

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1. Introduction

Emulsification is the process of finely dispersing one or more immiscible fluids into each other. This process is widely used in the formulation of many kinds of products in the process industry, especially in the food, cosmetic, pharmaceutical, and chemical industries (Joscelyne and Trägårdh, 1999). The design of high-quality dispersions (emulsions), in terms of user-controlled droplet size and higher uniformity, is essential and yet still a key challenge in the industry (Piacentini

et al., 2017). High throughput devices (such as colloid mills, high-pressure homogenizers, and rotor-stator systems) are traditionally used to prepare emulsions (Berton-Carabin et al., 2018). However, the unpredictability of the emulsion quality from batch to batch is a big challenge for such devices. The turbulent eddies that generate shear stresses responsible for droplet disruption in the pre-emulsion are usually high and uncontrollable (Piacentini et al., 2017; Santana et al., 2013). This results in emulsions with a wide droplet size distribution (low-quality emulsion) and besides, the energy inputs and losses in form of heat are usually high.

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The high shear stress, combined with heat generation in the system can lead to the loss of functional properties of shear and heat-sensitive components, and the disintegration of multiple emulsions (Charcosset et al., 2004; Surh et al., 2007).

The use of microporous structures (membranes, micro-channels, microsieves, and packed bed of particles) to prepare droplet-size controlled emulsions, at low shear stress and energy input, has been proposed in several studies (Spyropoulos et al., 2014). Among the proposed methods, the premix ME (membrane emulsification) method has been demonstrated as a suitable method for preparing droplet-size controlled emulsions at production throughputs comparable to those achieved while using the traditional high throughput devices (Nazir et al., 2010; Sahin et al., 2014). A coarse pre-emulsion is permeated through a microporous membrane several times (about 3–10 times) to refine the emulsion droplet size and monodispersity. The large droplets of the coarse pre-emulsion are disrupted into fine droplets by utilizing the pores of the microporous membrane as a special kind of low-pressure, low-shear homogenizing valves. This has enabled the generation of user-tailored and reproducible fine emulsions with a narrow droplet size distribution, under low shear and energy input. And much higher production fluxes of 1 m³/m²/h and above, can be achieved (Vladisavljević et al., 2004). The microporous membrane is the most critical element of the Premix ME method and the selection of a suitable membrane is essential for achieving the desired emulsification results. Various kinds of microporous structures have been used in premix ME however the Shirasu porous glass (SPG) membrane is the most preferred membrane, due to its unique characteristics and superiority for preparing emulsions with controlled droplets size and uniformity (Vladisavljević et al., 2004). Other kinds of microporous structures and porous media have been discussed and reviewed by Nazir et al. Nazir and Vladisavljević (2021).

Preparation of concentrated emulsions had been a big challenge in the membrane emulsification method due to a decrease in the emulsion throughput and susceptibility to membrane fouling, at higher emulsion viscosity or when dispersed phase content was higher (50 vol% or more). However, by introducing the premix ME method, SUZUKI et al. (1999) could prepare highly concentrated emulsions through phase inversion of low concentrated forms of oil-in-water (O/W) or W/O pre-emulsion using PTFE membranes. Concentrated emulsions are of substantial interest in process industries such as pharmaceutical, cosmetics, personal care, and food, where many kinds of viscous emulsions are handled. The emulsion viscosity greatly affects the appearance, texture, surface gloss, and storage stability of the emulsion products (Li et al., 2018; Osborn and Akoh, 2004). Thickeners are usually added to the continuous phase to increase its viscosity and prevent rapid phase separation, thereby delaying the onset of droplet creaming or sedimentation (Berton-Carabin et al., 2018).

In premix ME, the emulsion viscosity is one of the major factors that influence the wall shear stresses in pores responsible for droplet disruption (Nazir et al., 2010). Vladisavljević et al. (2006) and Zhou et al. (2008) have reported that the product of the mean emulsion viscosity and the membrane permeation rate (or flux), largely influence the wall shear stresses inside the membrane pores. The wall shear stresses can either be adjusted by varying the membrane permeation rate or the emulsion viscosity. However, this relationship between the MPR and emulsion

viscosity has not been studied extensively. Especially the effect of the viscosity of the discrete phases. In this study, we investigated the relationship of droplet size with the continuous phase viscosity and viscosity ratio at various membrane permeation rates. A high-pressure non-pulsating, high-flow type plunger pump, with a maximum pressure of 50 MPa was used to control a steady flow rate through the membrane pores.

2. Materials and equipment

2.1. Materials

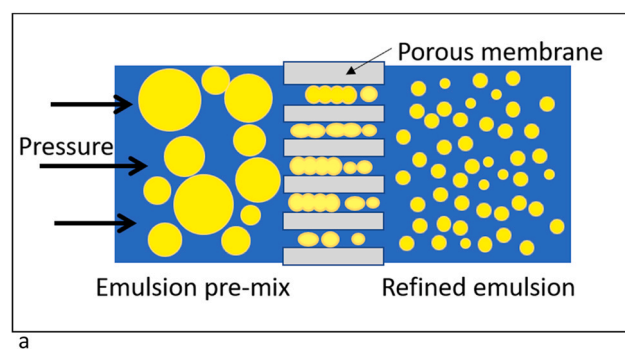
The microporous structure was a shirasu porous glass (SPG) membrane (SPG Techno Co. Ltd, Miyazaki, Japan) with an inner diameter of 9.0 mm, length of 150 mm, a thickness of 0.50 mm, and mean pore size of 5.0 μm. It was cut and polished according to the module length. The effective membrane area was 2.6 cm². The continuous phase was water containing a non-ionic surfactant Polyoxyethylene (20) sorbitan monolaurate (Tween 20) (Wako Pure Chemical Industries, Ltd, Osaka, Japan). Sodium alginate thickener (Nacalai Tesque, Inc., Kyoto, Japan) was used for adjusting the continuous phase viscosity. The dispersed phase was low and high viscosity Liquid paraffin (Nacalai Tesque, Inc.), mixed at appropriate viscosities. The viscosities of the continuous phase and the dispersed phase were measured at room temperature using a vibration type viscometer VM-10A-L (Sekonic Corporation, Tokyo, Japan).

2.2. Membrane emulsification setup

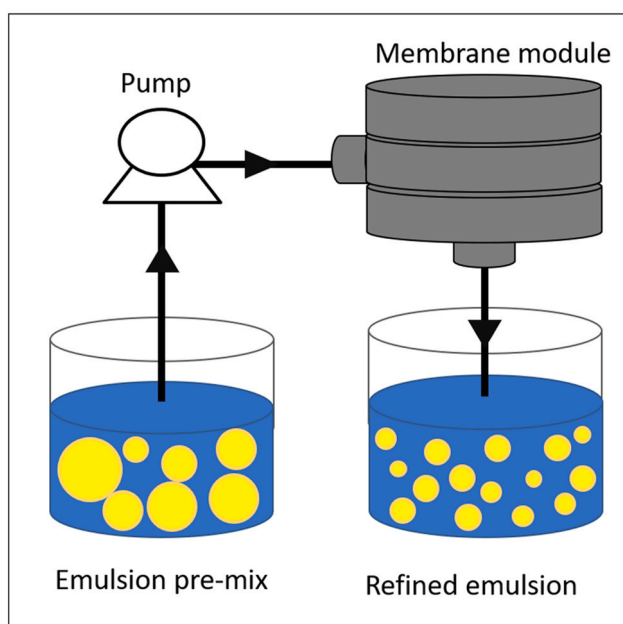
The membrane module was made from stainless steel material, Fig. 1. The SPG membrane was housed in 3 disassembled parts, connected through O-rings. A high-pressure non-pulsating, high-flow-type plunger pump, NPGXL 1000 (NS Nihon Seimitsu Kagaku Co., Ltd, Tokyo, Japan), was used to permeate the preliminary emulsion through the membrane at flow rates of 10–1000 ml/min and the maximum pump pressure was 50 MPa. A high-pressure non-pulsating, high-flow type plunger pump, with a maximum pressure of 50 MPa was used, to control a steady flow velocity in the membrane pores.

2.3. Emulsion preparation

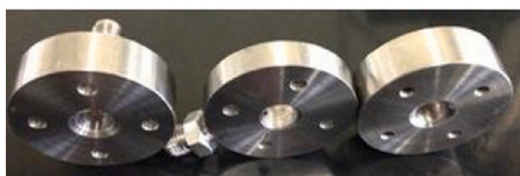
Oil-in-water (O/W) emulsions were prepared using the apparatus illustrated in figure1. The continuous water phase was 0.5 wt% Tween 20, with viscosity adjusted from 1 to 55 mPa·s and the dispersed phase was liquid paraffin. The pre-emulsion was prepared by mixing 4: 1 part of the continuous phase and the dispersed phase using an AHG-160D homogenizer (AS ONE Corporation, Osaka Japan) at 5000 rpm for 45 s. The pre-emulsion was then permeated through a 5.0 μm SPG membrane at a permeation flux of 2.4–240 m³/m²·h. The droplet size of the prepared emulsion was analyzed using a laser diffraction particle size distribution analyzer, SALD-200 V (Shimadzu Co. Ltd., Kyoto, Japan). Before emulsification, the SPG membrane was cleaned with ethanol and with ion-exchanged water for 5 min each, in an ultrasonic washer, and then wetted with a continuous phase solution. After emulsification, the membrane was ultrasonic cleaned in a detergent solution in hot water (about 60 °C) for about 30 min, followed by rinsing in ethanol and ion-exchanged water. In



a



b



c

Fig. 1 – a. Simple illustration of the droplet breakup formation process in premix membrane emulsification method. Fig. 1b. Simple illustration of the experimental setup of the premix membrane emulsification method. Fig. 1c. Stainless steel module and the disassembled parts housing the shirasu porous glass (SPG) membrane.

cases when fouling was observed, the membrane was heated at 500 °C for about 6 h in an Advantec model KM-420 electric muffle furnace, to restore the inherent properties of the membranes.

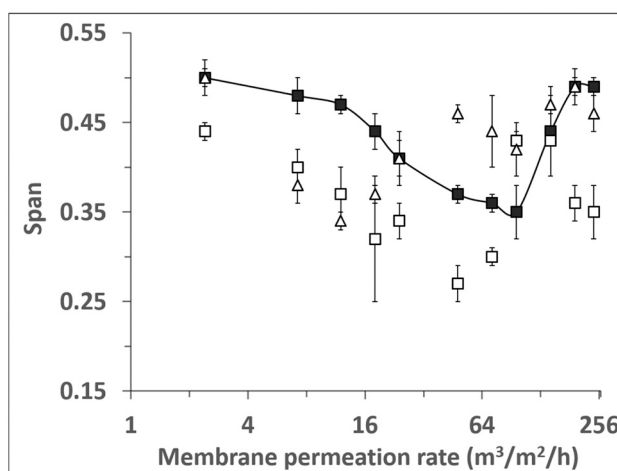
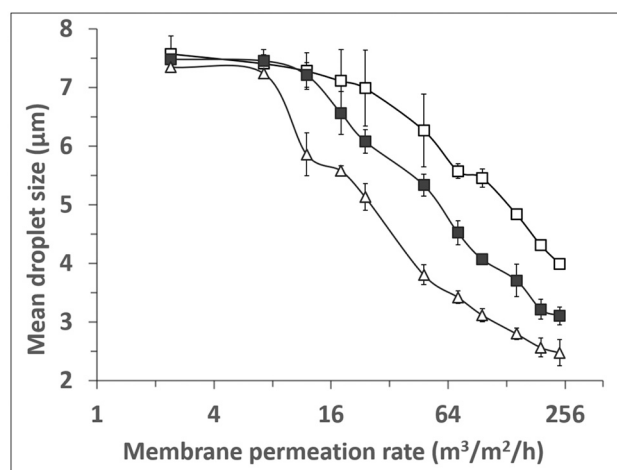


Fig. 2 – Effect of membrane permeation rate and continuous phase viscosity (□ 5, ■ 20, and △ 55 mPa·s) on mean droplet size and droplet size distribution (span). Dispersed phase viscosity fixed at 20 mPa·s.

3. Results and discussions

3.1. Effect of membrane permeation rate on droplet size

In this study, a coarse pre-emulsion (average droplet size of 43 µm) was permeated through a 5.0 µm SPG membrane at permeation rates (flux) of 2.4–240 m³/m²h, to refine the emulsion droplet size and uniformity. The effect of the membrane permeation rate (MPR) on droplet size at low and higher continuous phase viscosities, was investigated. The dispersed phase viscosity was fixed at 20 mPa·s. From Fig. 2, the mean droplet size was observed to decrease with an increase in the MPR due to the higher wall shear stresses generated in the pores at higher flow velocities. All the emulsions prepared were monodispersed with a droplet size distribution (span) of 0.27 – 0.5. When the emulsion pre-emulsion is permeated through the microporous membrane, the wall shear stresses ($\sigma_{w,p}$) inside the pores, which are responsible for the breakup of the droplets (Nazir et al., 2010; Vladisavljević et al., 2004) are defined as;

$$\sigma_{w,p} = \frac{8\eta_e J \xi}{\varepsilon d_p} \quad (1)$$

Where, η_e is the mean emulsion viscosity, J is the MPR, ξ the mean tortuosity factor of the pores, ε the mean membrane porosity and d_p the mean pore size. From Eq. 1, the wall shear

stresses in the membrane pores are directly proportional to the MPR, therefore the decrease in the droplet size at higher MPR is a result of the higher shear stresses in the pores. Though, at lower MPR, there was no significant effect on the droplet size due to the low shear stress generated. The droplet breakup formation inside the pores was mostly governed by the snap-off mechanism due to localized shear and deformation occurring at the pore constrictions, pore inlets, and the pore outlets, (Van Der Zwan et al., 2006), however, as the shear stress increased with MPR the size of the droplets became smaller and span narrower. At higher MPR (about $96 \text{ m}^3/\text{m}^2\text{h}$ and above), although the droplet size decreased further, the span values tended to increase due to irregular droplet breakdown as Rayleigh instabilities developed in the elongating droplet, when the shear stresses in the system are intense (Mugabi et al., 2019). Therefore, during premix ME, the MPR should be controlled to prevent widening of the span, although still, the span values in this study (of 0.27 – 0.5) were in the allowable range for monodispersed emulsions.

3.2. Effect of continuous phase viscosity on droplet size

The viscosity of the continuous phase was adjusted from 1 to 55 mPa·s by adding appropriate amounts of sodium alginate and its effect on the mean emulsion droplet size was investigated. From Fig. 3, we observed that the mean droplet size decreased with continuous phase viscosity and the dispersed phase viscosity had no significant effect on the relationship. The wall shear stress in the membrane pores increased with continuous phase viscosity, resulting in the generation of smaller droplets (Vladislavljević et al., 2006). From Eq. 1, the mean emulsion viscosity is proportional to the wall shear stress.

When the dispersed phase volume fraction is low, the mean emulsion viscosity is close to the viscosity of the continuous phase viscosity (Nazir et al., 2010), therefore increasing the continuous phase viscosity, led to an increase in the wall shear stress in the pores. In this study, the dispersed phase volume fraction was maintained at 20 vol%, and increasing its viscosity did not significantly affect the relationship between the continuous phase viscosity and droplet size, as observed in Fig. 3. The continuous phase viscosity also affects the thickness of the thin lubricating fluid layer which surrounds the stretching droplets in the pores. The thickness (h) is related to the radius (R_c) of the droplets as (Kukizaki, 2009);

$$h = R_c (\eta_c U_d / \gamma)^{2/3} \quad (2)$$

where, η_c is the viscosity of the continuous phase, γ interfacial tension and U_d the velocity of droplets. The droplets are disintegrated into smaller droplets when the thickness (h) of the lubricating layer is sufficiently large, that $R_c < \lambda/(2\pi)$, where, λ is the Plateau-Rayleigh wavelength. Since (h) is proportional to η_c , the emulsion droplet size decreases with the increase of the continuous phase viscosity (Vladislavljević et al., 2006; Kukizaki, 2009).

3.3. Effect of MPR on the relationship of continuous phase viscosity and droplet size

The relationship between the continuous phase viscosity and droplet size was also studied at various membrane permeation rates (MPR) of 2.4, 7.2, 48, 144, and $240 \text{ m}^3/\text{m}^2\text{h}$. From

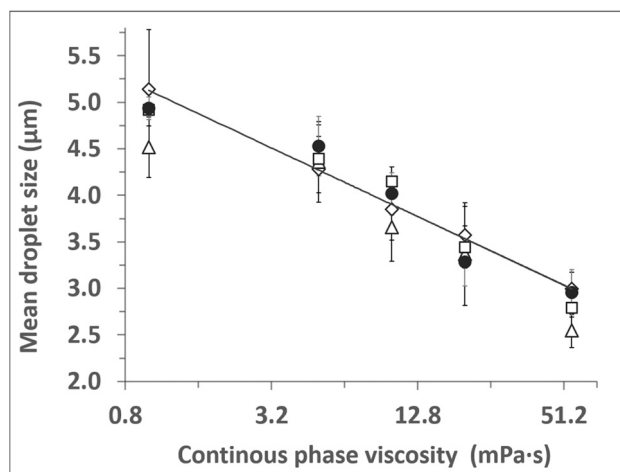


Fig. 3 – Effect of continuous phase viscosity on droplet size at MPR of $192 \text{ m}^3/\text{m}^2\text{h}$ and dispersed phase viscosity of $\diamond$ 5, $\triangle$ 20, $\square$ 55 and $\bullet$ 110 mPa·s.

Fig. 4, it can be observed that the relationship between the continuous phase viscosity and mean droplet size differed at high and low MPR. At lower MPR (2.4 and $7.2 \text{ m}^3/\text{m}^2\text{h}$) the mean droplet size increased with an increase in the continuous phase viscosity, although the extent was relatively small. Whereas at higher MPR (about $48 \text{ m}^3/\text{m}^2\text{h}$ and above), the mean droplet size decreased gradually with an increase in the continuous phase viscosity. Although, the droplet size was expected to decrease with increased continuous phase viscosity, based on Eqs. 1 and 2, the increase in droplet size at lower MPR was thought to be a result of droplet coalesce as the rate of diffusion of the surfactant molecules to the newly created interface was reduced at lower flow velocities in the pores (Nazir et al., 2014).

However, at higher MPR, the rate of adsorption of the surfactant molecules to the newly created interface after droplet disruption was faster. The magnitude of shear stresses generated in the pores also increased due to an increase in the flow velocity at higher MPR, leading to the formation of smaller droplets. Therefore, the relative variations of the MPR and emulsion viscosity may increase or decrease the shear stresses acting on the droplets of the pre-emulsion flowing through the membrane pores, and conditions for emulsification should be chosen carefully for optimum emulsification results.

3.4. Effect of viscosity ratio on emulsion droplet size

The influence of the emulsion viscosity ratio on droplet size was also investigated. The viscosity ratio (δ) was defined as in Eq. 3, where, η_c was the continuous phase viscosity and η_d the dispersed phase viscosity.

$$\delta = \eta_d / \eta_c \quad (3)$$

The continuous phase viscosity was adjusted from 1 mPa·s to 5, 10, 20, 55 mPa·s by adding appropriate amounts of Sodium alginate while the dispersed phase viscosity was adjusted from 5 mPa·s to 10, 20, 55, 110, 220 mPa·s by mixing appropriate proportions of low and high viscosity liquid paraffin. First, the effects of δ were analyzed at a lower MPR of $2.4 \text{ m}^3/\text{m}^2\text{h}$. The droplet size was observed to decrease with an increase in the viscosity ratio as shown in Fig. 5a. However, when the MPR was moderately high ($72 \text{ m}^3/\text{m}^2\text{h}$), the droplet size was observed to increase with viscosity ratio,

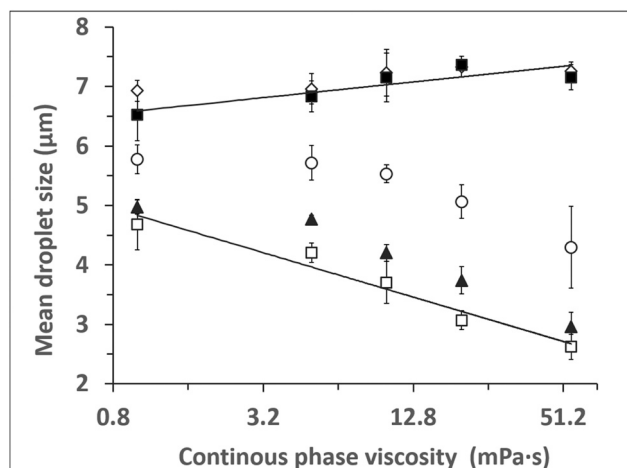


Fig. 4 – Effect of continuous phase viscosity on droplet size at dispersed phase viscosity of 55 mPa·s and MPR of $\diamond$ 2.4, $\blacksquare$ 7.2, $\circ$ 48, $\blacktriangle$ 144, and $\square$ 240 $\text{m}^3/\text{m}^2/\text{h}$.

until a certain ratio beyond which the droplet size became independent of the viscosity ratio (Fig. 5b), and with further increase in MPR (beyond 192 $\text{m}^3/\text{m}^2/\text{h}$), the droplet size increased steadily with viscosity ratio (Fig. 5c). The difference in the relationship between viscosity ratio and droplet size with MPR was thought to be a result of changes in the droplet disruption mechanism occurring at different flow velocities in the membrane pores. In Section 3.2, Eq. 2, it was observed that an increase in the continuous phase viscosity led to the formation of the smaller droplet. Therefore, it would be expected that increasing the viscosity ratio (that is, reducing the continuous phase viscosity); augmented by the low wall shear stress in the pores due to the low MPR, would increase the emulsion droplet size.

Certainly, the droplet size at a lower viscosity ratio seemed to be increasing when observed individually, but when observed as a whole, the trend is decreasing with an increase in viscosity ratio. Some studies have shown that, at lower flow velocities, increasing the viscosity of the stretching fluid (dispersed phase) can profoundly delay the development of perturbing ripples (Rayleigh-Plateau instabilities), thus allowing the droplets to stretch into very long and thin threads, before breaking up into smaller droplets (Javadi et al., 2013; Senchenko and Bohr, 2005). This could explain why smaller droplets were prepared with an increasing viscosity ratio at a lower MPR (Fig. 5a). However, as the flow velocity in the pores increase, the stretching droplet becomes more susceptible to the flow instabilities and this phenomenon is lost.

At moderately high MPR (72 $\text{m}^3/\text{m}^2/\text{h}$), the droplet size increased with viscosity ratio, but after a certain viscosity ratio, the droplet size became more or less independent of the viscosity ratio (Fig. 5b). This kind of relationship was also previously reported by Nazir et al. (2014). They observed that from a viscosity ratio of 3 and above the droplet size became more independent of the viscosity ratio. They deduced that this was a result of the change of droplet formation mechanism as the flux through the pore became low with increasing emulsion viscosity. In this study, a high-pressure non-pulsating, high flow type plunger pump, which can comparatively control a steady flux, was used to permeate the preliminary emulsion through the membrane. The droplet size became less dependent on the viscosity ratio at

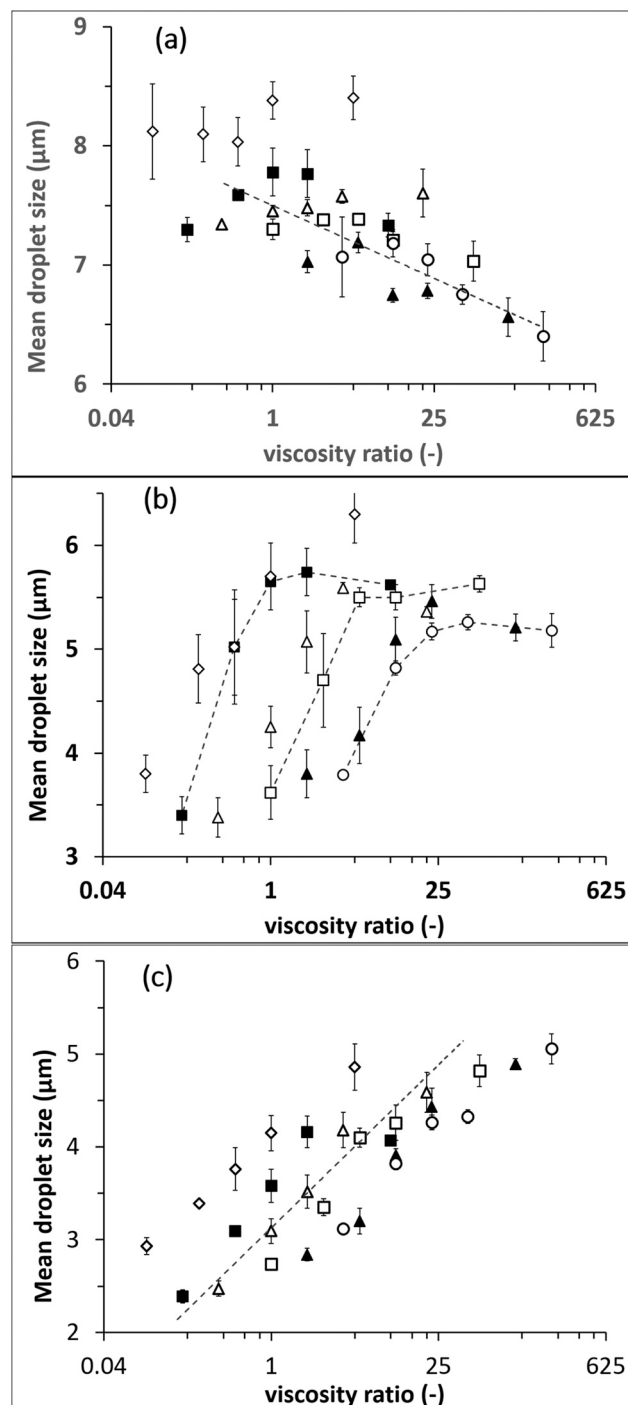


Fig. 5 – Effect of viscosity ratio on mean droplet size at (a) lower (2.4 $\text{m}^3/\text{m}^2/\text{h}$), (b) moderate (72 $\text{m}^3/\text{m}^2/\text{h}$) and (c) higher (192 $\text{m}^3/\text{m}^2/\text{h}$) membrane permeation rates. The dispersed phase viscosities were $\diamond$ 5, $\blacksquare$ 10, $\triangle$ 20, $\square$ 55, $\blacktriangle$ 110 and $\circ$ 220 mPa·s.

different ratios depending on the emulsion formulation (that is, dispersed phase viscosities). At lower viscosity ratios, the viscosity of the continuous phase relative to the disperse phase is higher. Therefore, the continuous phase can effectively stretch the droplet into thinner liquid threads/cylinders before breaking it up into smaller droplets. Komrakova et al. (2015) also observed that droplets become more elongated as the viscosity ratio is decreased. However, as the viscosity ratio increased, the lubricating fluid of the continuous phase become thinner while the dispersed droplets became more rigid and more difficult to deform or

fragmented by the continuous phase lubricating film (Nekouei and Vanapalli, 2017). At some point, the influence of the continuous phase on droplet deformation becomes minimal and the droplet breakup process becomes more governed by the snap-off mechanism due to localized shear and deformation occurring at the constrictions and channels in the membrane pores (Van Der Zwan et al., 2006). At this point, the droplet size becomes less dependent on the viscosity ratio. Since Nazir et al. (2014) deduced that the change in droplet formation mechanism was a result of the flux through the membrane, the relationship between viscosity ratio was also studied at a higher MPR ($192 \text{ m}^3/\text{m}^2/\text{h}$). The tendency of the droplet size to become less dependent on viscosity ratio was lost at higher MPR, and the droplet size increased more linearly as shown in Fig. 5c, due to an increase in the shear stress in the pores. This statement may be quite misleading when observed apart because the increase in shear stress is expected to result in smaller droplets. When we compare Figs. 5a, 5b, and 5c, the emulsion droplet sizes decreased in the order of $5a > 5b > 5c$, as the MPR was increased. In this study, we observed that the relationship between the emulsion viscosity ratio and the emulsion droplet size is highly affected by the membrane permeation rate.

4. Conclusion

The premix membrane emulsification method is a suitable method for preparing viscous emulsions with a controlled droplet-size distribution and at production throughputs comparable to the traditional high throughput devices. The emulsion viscosity and the membrane permeation rates (MPR) are the major factors influencing the wall shear stresses in pores responsible for droplet disruption. However, the effect of the discrete phase viscosities on droplet size and their relationship with the MPR is not well elaborated. In this study, the relationship between droplet size and the continuous phase viscosity ($1\text{--}55 \text{ mPa}\cdot\text{s}$) at different MPRs was investigated. The emulsion droplet size was observed to decrease with continuous phase viscosity due to the increase in the wall shear stress in the pores. The relationship between the continuous phase viscosity and droplet size changed with MPR. At lower MPR, the droplet size was observed to increase with viscosity due to coalescing as the rate of surfactant diffusion onto the interface of the new droplet reduced, but at higher MPR the droplet size decreased with viscosity. The wall shear stresses in the membrane pore may be low or high depending on the relative variation of the emulsion viscosity and the MPR.

The viscosity ratio was also observed to affect the droplet size differently depending on the MPR. The droplet size was observed to decrease with an increase in the viscosity ratio, at lower MPR, due to the prolonged stretching of the droplets, as the development of Rayleigh-Plateau instabilities in the stretching droplet is delayed by the dispersed phase viscosity, at low flow fluxes. The stretching droplets become more susceptible to instabilities at higher MPR. When the MPR was moderated, the droplet size was observed to increase with viscosity ratio, until a certain ratio beyond which the droplet size became independent of the viscosity ratio, although with further increase in MPR, this region was lost and droplet size increased steadily with viscosity ratio.

In this study, we observed that the relationship between the mean emulsion viscosity and the membrane permeation

rate (or flux) on droplet formation in the pores of a micro-porous membrane, may be complementary or contrasting depending on their relative magnitude and the effect of the viscosities of the discrete phases is vital.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Prof. Jae-Ho Jeong reports financial support was provided by Ministry of Trade, Industry and Energy (MOTIE, Korea). Prof. Jae-Ho Jeong reports financial support was provided by Korea Institute of Energy Technology Evaluation and Planning (KETEP).

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