Potential of High-Powered Bath-Type Ultrasonicator for Manufacturing of Emulsifier-Free Emulsions

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Abstract

We report here on the manufacturing methods for emulsifier-free (EF-) oil-in-water (O/W) emulsions, in which oil and water were mechanically mixed in the absence of any emulsifiers such as surfactants. A commercially available bath-type ultrasonic cleaner (Bath-US; 42 kHz, 26 W) did not disperse dodecane (DD) completely in water even by the treatment for 15 min, and the average diameter of DD droplets in the EF-dodecane-in-water (DD/W) emulsions prepared was several micrometers. Using a commercially available horn-type ultrasonic homogenizer (Horn-US; 19.5 kHz, 600 W), the EF-DD/W emulsions were prepared by treatment for ~1 min, and the average diameter of DD droplets in EF-DD/W emulsions prepared was ~1 μm. Preparation of EF-DD/W emulsions using a high-powered bath-type ultrasonicator (HPBath-US; 28 kHz, 300 W) that we developed required the treatment period for ~1 min. The DD droplets in EF-DD/W emulsions prepared with HPBath-US became several hundred nanometers in diameter by treatment for ~10 min. The preparation of EF-DD/W emulsions with a commercially available rotor-stator homogenizer (RS-HG; 15,000 rpm, 800 W) required the treatment period for ~1 min, and DD droplets in DD/W emulsions prepared were several micrometers in diameter. Colloidal stability of EF-DD/W emulsions prepared with Horn-US and HPBath-US was higher than that of EF-DD/W emulsions prepared with Bath-US and RS-HG. These studies indicate that HPBath-US has advantages for the manufacturing of EF-O/W emulsions. Furthermore, the continuous manufacturing of EF-DD/W emulsions was achieved by attaching the flow chamber with HPBath-US.

Key-words: Manufacturing method, Emulsifier-free oil-in-water emulsion, High-powered bath-type ultrasonicator, Treatment period, Droplet size, Colloidal stability, Continuous manufacturing

1. Introduction

An emulsion is a transient mixture of two immiscible liquids such as oil and water prepared with the aid of mechanical mixers (e.g., homogenizers) and surface active agents¹⁻³). Since the interfacial free energy (G) of the emulsion (mixture of oil and water) becomes higher due to the increase in the interface area (A) between oil and water (see equation 1), oil and water naturally separate from each other with the elapsed time after preparation¹⁻³).

$$G = \gamma A \quad (1)$$

where γ is the interfacial tension between oil and water. Thus, the lower γ and the resulting lower G are required to retain the emulsion state for a longer period. Therefore, in general, the emulsifiers such as surfactants are required for the long-term colloidal stabilization of emulsions because the emulsifiers decrease the interfacial tension (γ) between

oil and water. On the other hand, we have investigated the colloidal stability of oil-in-water (O/W)⁴⁻¹²) and water-in-oil (W/O) emulsions¹³⁻¹⁵) prepared with the ultrasonicator in the absence of any emulsifiers such as surfactants, which were named as emulsifier-free (EF-) O/W and W/O emulsions, respectively. We found that the long-term colloidal stabilization of EF-O/W and EF-W/O emulsions prepared with an ultrasonicator was achieved by optimization of oil type and composition⁴⁻¹⁵). Moreover, in recent years, the EF-emulsion products are strongly demanded because of the biocompatibility of emulsion products and environmentally benign approaches for manufacturing emulsion products¹⁶⁻²⁴).

Then, in this work, the manufacturing methods of EF-O/W emulsions were examined for practical applications. A commercially available both-type ultrasonic cleaner (Bath-US; 42 kHz, 26 W), a commercially available horn-type ultrasonic homogenizer (Horn-US; 19.5 kHz, 600 W), a high-powered bath-type ultrasonicator (HPBath-US; 28 kHz, 300 W) that we developed and a

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commercially available rotor-stator homogenizer (RS-HG; 15,000 rpm, 800 W) were tested for the manufacturing of EF-O/W emulsions in terms of the treatment period for preparation, a minimum diameter of oil droplets in EF-O/W emulsions and colloidal stability of EF-O/W emulsions. Furthermore, the continuous manufacturing of EF-O/W emulsions was examined toward mass production of EF-O/W emulsions.

2. Experimental

2.1 Preparation of EF-O/W emulsions

EF-O/W emulsions were prepared by mechanically mixing 1.0 mL Dodecane (DD; $C_{12}H_{26}$, 99.5%, Tokyo Chemical Industry Co., Ltd.) and 250 mL ultrapure water (18.2 M Ω cm at 25 °C, Millipore-filtered water) at 25 °C. A commercially available bath-type ultrasonic cleaner (Bath-US, 42 kHz, 26 W), a commercially available horn-type ultrasonic homogenizer (Horn-US, 19.5 kHz, 600 W), a high-powered bath-type ultrasonicator (HPBath-US; 28 kHz, 300 W) that we developed or a commercially available rotor-stator

homogenizer (RS-HG; 15,000 rpm, 800 W) were used for preparation of EF-O/W emulsions (see Fig. 1).

2.2 Characterization of emulsifying process for preparation of EF-DD/W emulsions

Turbidity of the EF-DD/W emulsion during emulsification treatment was measured at room temperature with a UV-visible spectrophotometer (V-630, JASCO) to characterize the emulsifying process with Bath-US, Horn-US, HPBath-US, and RS-HG for preparation of EF-DD/W emulsions. The turbidity of the EF-DD/W emulsion was obtained from the difference of transmittances between EF-DD/W emulsion and continuous phase (water phase) at a wavelength of 700 nm measured with the UV-visible spectrophotometer. Size distribution of DD droplets in the EF-DD/W emulsion was also measured at room temperature during emulsification treatment by a laser diffraction method (LA-950, HORIBA) to characterize the emulsifying process with Bath-US, Horn-US, HPBath-US and RS-HG for preparation of EF-DD/W emulsions. For the size distribution measurement of DD droplets in the EF-DD/W emulsion prepared, the EF-DD/W emulsion was diluted by ultrapure water to adjust the transmittance of the EF-DD/W emulsion.

2.3 Characterization of colloidal stability of EF-DD/W emulsions prepared

Colloidal stability of the EF-DD/W emulsions was characterized by monitoring the turbidity of the EF-DD/W emulsions with the elapsed time after emulsification treatment for 5 min with Bath-US, Horn-US, HPBath-US, and RS-HG. The EF-DD/W emulsions prepared were stored in the glass cell with a screw cap for UV-visible spectroscopy at 25 °C during the characterization of colloidal stability of EF-DD/W emulsions. The turbidity of the EF-DD/W emulsion prepared was measured at room temperature with a UV-visible spectrophotometer (V-630, JASCO). The light passes

Fig. 1 Schematic illustration on emulsification system of EF-O/W emulsions with (a) Bath-US, (b) Horn-US, (c) HPBath-US and (d) RS-HG.

Fig. 2 Schematic illustration on Continuous emulsification setup, in which the flow chamber is attached to HPBath-US.

through the window (width of 8.0 mm and height of 20 mm) positioned at 5.0 mm from the bottom of the glass cell (inner width of 10 mm square and height of 48 mm). Thus, the change in the turbidity of EF-DD/W emulsion results from the creaming of DD droplets in the EF-DD/W emulsion.

2.4 Continuous manufacturing of EF-DD/W emulsions with HPBath-US

Continuous emulsification setup was built by attaching of flow chamber with HPBath-US. EF-DD/W emulsion preprepared by mixing of 2.0 mL DD and 500 mL ultrapure water with RS-HG (treatment period for 7 min) was carried to the flow chamber attached to the HPBath-US through silicone tubes with a pump (see Fig. 2).

3. Results and discussion

3.1 Emulsifying process for preparation of EF-DD/W emulsions with Bath-US, Horn-US, HPBath-US, and RS-HG

In order to evaluate the performance of Bath-US, Horn-US, HPBath-US, and RS-HG for the preparation of EF-DD/W emulsions, the change in the turbidity of mixture of DD and water was monitored during treatment. Dodecane (DD) and water are both originally transparent liquids, and the mixture of DD and water became opaque after treatment for 3 min

with Bath-US, which indicates the formation of EF-DD/W emulsion (see images of Bath-US in Fig. 3 (a)). On the other hand, the turbidity of EF-DD/W emulsion measured with the UV-visible spectrophotometer became ~50% after the treatment for 15 min with Bath-US (see plot maker ○ in Fig. 3 (b)). This indicates that DD was not dispersed completely in water even by the treatment for 15 min with Bath-US. In the case of emulsification with Horn-US, the mixture of DD and water changed to opaque after treatment for 0.5 min with Horn-US, which indicates the formation of EF-DD/W emulsion (see images of Horn-US in Fig. 3 (a)). Turbidity of EF-DD/W emulsion measured with the UV-visible spectrophotometer became 80% after treatment for 1 min with Horn-US (see plot maker □ in Fig. 3 (b)). The mixture of DD and water also changed to opaque after treatment for 0.5 min with HPBath-US, which indicates the formation of EF-DD/W emulsion (see images of HPBath-US in Fig. 3 (a)). Turbidity of EF-DD/W emulsion measured with the UV-visible spectrophotometer became 80% after treatment for 2 min with HPBath-US (see plot maker ● in Fig. 3 (b)). The mixture of DD and water changed to opaque after treatment for 0.5 min with RS-HG, which indicates the formation of EF-DD/W emulsion (see images of RS-HG in Fig. 3 (a)). Turbidity of EF-DD/W emulsion measured with the UV-visible spectrophotometer became 80% after treatment for 0.5 min with RS-HG (see plot maker ■ in Fig. 3 (b)). These indicate that the Horn-US, HPBath-US, and RS-HG are effective for preparation of EF-O/W emulsions (see Fig. 3 (c)).

Also, the change in the size of DD droplets in EF-DD/W emulsions was monitored during treatment to characterize the performance of Bath-US, Horn-US, HPBath-US, and RS-HG for the preparation of EF-DD/W emulsions. The average diameter of DD droplets in the EF-DD/W emulsion prepared with Bath-US was ~4 μm by treatment for 5 min and remained unchanged even for a longer treatment period (see Fig. 4 (a) and plot maker ○ in Fig. 4 (e)). For the EF-DD/W emulsion prepared with Horn-US, the average diameter of DD droplets in the EF-DD/W emulsion was ~1 μm by treatment for 1 min (see Fig. 4 (b) and plot maker □ in Fig. 4 (e)). In the case of EF-DD/W emulsion prepared with HPBath-US, the average diameter of DD droplets in the EF-DD/W emulsion gradually decreased up to 10 min in the treatment period and reached ~0.3 μm (see Fig. 4 (c) and plot maker ● in Fig. 4 (e)). The average diameter of DD droplets in the EF-DD/W emulsions prepared with RS-HG slightly decreased as treatment period and became ~6 μm by treatment for 7 min (see Fig. 4 (d) and plot maker ■ in Fig. 4 (e)). These results indicate that the HPBath-US has a benefit for production of smaller oil droplets in EF-O/W emulsions (see Fig. 4 (f)).

Fig. 3 (a) EF-DD/W emulsions during treatment with Bath-US, Horn-US, HPBath-US and RS-HG, (b) Turbidity of EF-DD/W emulsions prepared with (○) Bath-US, (□) Horn-US, (●) HPBath-US and (■) RS-HG plotted as a function of treatment period, and (c) Turbidity of EF-DD/W emulsions prepared after 15 min treatment with Bath-US, Horn-US, HPBath-US and RS-HG.

Fig. 4 Droplet size distributions of DD droplets in EF-DD/W emulsions prepared with (a) Bath-US, (b) Horn-US, (c) HPBath-US and (d) RS-HG in the treatment for 1, 5, 10 and 15 min, (e) Average diameter of DD droplets in EF-DD/W emulsions prepared with (○) Bath-US, (□) Horn-US, (●) HPBath-US and (■) RS-HG plotted as a function of treatment period, and (f) Average diameter of DD droplets in EF-DD/W emulsions prepared after 15 min treatment with Bath-US, Horn-US, HPBath-US and RS-HG.

3.2 Colloidal stability of EF-DD/W emulsions prepared with Bath-US, Horn-US, HPBath-US, and RS-HG

An emulsion is thermodynamically unstable, so it eventually separates into oil and water after preparation. Then, the colloidal stability of EF-DD/W emulsions prepared with Bath-US, Horn-US, HPBath-US, and RS-HG were checked in terms of turbidity change of EF-DD/W emulsions with the elapsed time after preparation. The opaqueness of EF-DD/W emulsions prepared with Bath-US and RS-HG changed to transparent at 1 d after preparation (see images of Bath-US and RS-HG in **Fig. 5** (a)). In the case of EF-DD/W emulsions prepared with Horn-US and HPBath-US, the opaque region of EF-DD/W emulsions gradually moved to upside, which is the creaming of DD droplets in the EF-DD/W emulsions (see images of Horn-US and HPBath-US in **Fig. 5** (a)). Turbidity of EF-DD/W emulsions prepared with Bath-US and RS-HG measured with the UV-visible

Fig. 5 (a) EF-DD/W emulsions after preparation with Bath-US, Horn-US, HPBath-US and RS-HG observed with elapsed time, (b) Turbidity of EF-DD/W emulsions after preparation with (○) Bath-US, (□) Horn-US, (●) HPBath-US and (■) RS-HG plotted as a function of elapsed time, and (c) Turbidity of EF-DD/W emulsions at 2 d after preparation with Bath-US, Horn-US, HPBath-US and RS-HG.

spectrophotometer became lower than 10% at 1 d after preparation (see plot maker ○ and ■ in **Fig. 5** (b)). Turbidity of EF-DD/W emulsions prepared with Horn-US and HPBath-US measured with the UV-visible spectrophotometer gradually decreased with the elapsed time after preparation and became ~20% at 6 d after preparation (see plot maker □ and ● in **Fig. 5** (b)). This is most likely related to the size of DD droplets in EF-DD/W emulsions prepared. As defined by Stokes equation (see equation 2)²⁵⁾, the creaming rate (u) of oil droplets in O/W emulsion is proportional to the square of droplet radius (r).

$$u = \frac{2r^2(\rho_0 - \rho)g}{9\eta} \dots\dots\dots (2)$$

where $(\rho_0 - \rho)$ is the density difference between continuous phase (water) and dispersed phase (oil), η is the viscosity of continuous phase (water) and g is the gravitational acceleration. The creaming rate (u) of DD droplets in EF-

DD/W emulsions prepared with Horn-US and HPBath-US became lower because of the smaller DD droplets in EF-DD/W emulsions prepared with Horn-US and HPBath-US (see Fig. 5 (c)).

As mentioned so far, the ultrasonic emulsification has advantage for EF-emulsification. This implies that the cavitation effect generated by ultrasonication is effective for EF-emulsification. In particular, the cavitation effect generated by ultrasonication is effective for the decrease in the size of oil droplets in EF-O/W emulsions and leads to the increase in the colloidal stability of EF-O/W emulsions (see Figs. 4 and 5). Among ultrasonic emulsification using Bath-US, Horn-US, and HPBath-US, the ultrasound with higher power (Horn-US and HPBath-US) is efficient for EF-emulsification (see Fig. 3). For comparison between Horn-US and HPBath-US, the diameter of DD droplets in EF-DD/W emulsions prepared with HPBath-US is smaller than that of DD droplets in EF-DD/W emulsions prepared with Horn-US (see Fig. 4). This is most likely related to the volume ratio of irradiation to non-irradiation of ultrasound to EF-DD/W emulsion. In the case of Horn-US, the ultrasonic oscillator is located inside of the EF-DD/W emulsion itself for emulsification, and the ultrasound generates from the top of the horn of Horn-US. As a result, DD droplets would be smaller around the top of the horn of Horn-US, while DD droplets would be larger at other locations in the EF-DD/W emulsion than the top of the horn of Horn-US because of the non-irradiation of ultrasound to EF-DD/W emulsion. On the other hand, in the case of HPBath-US, the ultrasound irradiates the whole emulsion from outside of the EF-DD/W emulsion for emulsification, so DD droplets would effectively be smaller.

3.3 Continuous manufacturing of EF-DD/W emulsion by attaching the flow chamber to HPBath-US

Having established the advantage of HPBath-US for EF-emulsification, the mass production of EF-O/W emulsion using HPBath-US was examined for practical application of EF-O/W emulsion. HPBath-US is not able to process the large volume of EF-O/W emulsion at once. Namely, there is an optimum volume of EF-O/W emulsion for EF-emulsification using HPBath-US. So, the iteration of batch emulsification and scale-up of HPBath-US are required in order to achieve the mass production of EF-O/W emulsion using HPBath-US. On the other hand, if the appropriate volume of EF-O/W emulsion for EF-emulsification carries to the HPBath-US continuously, it is possible to produce the massive EF-O/W emulsion at once. Then, the continuous manufacturing of EF-O/W emulsion using HPBath-US was examined for mass production of EF-O/W emulsion. EF-DD/W emulsion prepared with RS-HG was carried to the flow chamber attached to the HPBath-US through

Fig. 6 (a) Average diameter of DD droplets in EF-DD/W emulsions prepared by continuous emulsification with a flow chamber attached to HPBath-US plotted as a function of pass number. Flow rate of EF-DD/W emulsion; (▲) 1.21, (◆) 2.56, (▼) 3.18, (◀) 3.97 and (◆) 4.71 mL s⁻¹. (b) Average diameter of DD droplets in the EF-DD/W emulsion obtained after 5 times passage through the flow chamber attached to the HPBath-US plotted as a function of flow rate of EF-DD/W emulsion.

silicone tubes with a pump (see Fig. 2). The DD droplets with average diameter of $\sim 10 \mu\text{m}$ in the DD/W emulsion prepared with RS-HG decreased by passage through the flow chamber attached to the HPBath-US (see Fig. 6 (a)). In the case of flow rate of 1.21 mL s^{-1} in the flow chamber, the DD droplets with average diameter of $\sim 10 \mu\text{m}$ in the DD/W emulsion prepared with RS-HG decreased to $\sim 0.4 \mu\text{m}$ after 5 times passage through the flow chamber attached to the HPBath-US (see plot maker ▲ in Figs. 6 (a) and (b)). The average diameter of DD droplets in the DD/W emulsion obtained after 5 times passage through the flow chamber attached to the HPBath-US was similar to that of DD droplets in the DD/W emulsion prepared by batch emulsification with HPBath-US (as shown in Fig. 1 (c)). On the other hand, the increase in the flow rate up to 4.71 mL s^{-1} in the flow chamber led to the increase in the size of DD droplets in EF-DD/W emulsion even after 5 times passage through the flow chamber attached to the HPBath-US, in which the average diameter of DD droplets in the EF-DD/W emulsion was $\sim 1.5 \mu\text{m}$ (see plot maker ◆ in Figs. 6 (a) and (b)). This indicates that the size of oil droplets in O/W emulsions is controlled by the number of passages and flow rate of O/W emulsion in the flow chamber attached to the HPBath-US. Colloidal stability of EF-DD/W emulsion obtained after 5 times passage with flow rate of 1.21 mL s^{-1} through the flow chamber attached to the HPBath-US was consistent with that of the EF-DD/W emulsion obtained by batch emulsification with HPBath-US (see Fig. 7). These results suggest that mass production of EF-O/W emulsions is achieved by the combination of flow chamber with HPBath-US.

4. Conclusion

Manufacturing of emulsifier-free (EF-) dodecane-in-water (DD/W) emulsions was examined using commercially

Fig. 7 (a) EF-DD/W emulsions prepared by Continuous emulsification (5 times passage with flow rate of 1.21 mL s^{-1} through the flow chamber attached to the HPBath-US) and Batch emulsification (with HPBath-US) observed with elapsed time, (b) Turbidity of ($\diamond$) EF-DD/W emulsion prepared by Continuous emulsification and ($\bullet$) EF-DD/W emulsion prepared by Batch emulsification recorded with elapsed time, and (c) Turbidity of EF-DD/W emulsions at 2 d after preparation by Continuous emulsification and Batch emulsification.

available bath-type ultrasonic cleaner (Bath-US), horn-type ultrasonic homogenizer (Horn-US), high-powered bath-type ultrasonicator (HPBath-US) that we developed, and rotor-stator homogenizer (RS-HG). We revealed that HPBath-US has advantages for the manufacturing of EF-O/W emulsions. Then, the continuous manufacturing system of EF-O/W emulsions was developed by attaching the flow chamber to

HPBath-US. These studies indicate that HPBath-US itself and the continuous manufacturing system with HPBath-US are applicable to the manufacturing of EF-emulsion products.

References

- 1) Food emulsions (4th ed.), S. E. Friberg, K. Larsson, J. Sjöblom, Marcel Dekker, New York 2004.
- 2) Food emulsions: Principles, practices, and techniques (2nd ed.), D. J. McClements, CRC Press, Boca Raton (2005).
- 3) Pharmaceutical Emulsions, D. K. Sarker, WILEY Blackwell, Oxford (2013).
- 4) K. Kamogawa, T. Sakai, N. Momozawa, M. Shimazaki, M. Enomura, H. Sakai, M. Abe: *J. Jpn. Oil Chem. Soc.*, **47**, 159 (1998).
- 5) T. Sakai, K. Kamogawa, F. Harusawa, N. Momozawa, H. Sakai, M. Abe: *Langmuir*, **17**, 255 (2001).
- 6) T. Sakai, K. Kamogawa, K. Nishiyama, H. Sakai, M. Abe: *Langmuir*, **18**, 1985 (2002).
- 7) T. Sakai, Y. Takeda, F. Mafune, M. Abe, T. Kondow: *J. Phys. Chem. B*, **107**, 2921 (2003).
- 8) T. Sakai: *Curr. Opin. Colloid Interface Sci.*, **13**, 228 (2008).
- 9) K. Kamogawa, M. Matsumoto, T. Kobayashi, T. Sakai, H. Sakai, M. Abe: *Langmuir*, **15**, 1913 (1999).
- 10) K. Kamogawa, H. Akatsuka, M. Matsumoto, S. Yokoyama, T. Sakai, H. Sakai, M. Abe: *Colloid Surf. A*, **180**, 41 (2001).
- 11) K. Kamogawa, N. Kuwayama, T. Katagiri, H. Akatsuka, T. Sakai, H. Sakai, M. Abe: *Langmuir*, **19**, 4063 (2003).
- 12) K. Kamogawa, G. Okudaira, M. Matsumoto, T. Sakai, H. Sakai, M. Abe: *Langmuir*, **20**, 2043 (2004).
- 13) T. Sakai, K. Seo: *J. Jpn. Soc. Colour Mater.*, **87**, 387 (2014).
- 14) T. Sakai, S. Urabe, K. Seo: *J. Jpn. Soc. Colour Mater.*, **89**, 333 (2016).
- 15) T. Sakai, T. Oishi: *J. Taiwan Inst. Chem. Eng.*, **92**, 123 (2018).
- 16) R. M. Pashley: *J. Phys. Chem. B*, **107**, 1714 (2003).
- 17) N. Maeda, K. J. Rosenberg, J. N. Israelachvili, R. M. Pashley: *Langmuir*, **20**, 3129 (2004).
- 18) M. J. Francis, R. M. Pashley: *Colloid Surf. A*, **260**, 7 (2005).
- 19) K. Nakabayashi, F. Amemiya, T. Fuchigami, K. Machida, S. Takeda, K. Tamamitsu, M. Atobe: *Chem. Commun.*, **47**, 5765 (2011).
- 20) K. Nakabayashi, T. Fuchigami, M. Atobe: *Electrochimica Acta*, **110**, 593 (2013).
- 21) K. Nakabayashi, M. Kojima, S. Inagi, Y. Hirai, M. Atobe: *ACS Macro Lett.*, **2**, 482 (2013).
- 22) K. Nakabayashi, T. Fuchigami, M. Atobe: *Rsc Adv.*, **4**, 22938 (2014).
- 23) K. Nakabayashi, H. Yanagi, M. Atobe: *Rsc Adv.*, **4**, 57608 (2014).
- 24) M. Kumpugdee-Vollrath, J.-P. Krause, S. Bürk: *Inter. J. Pharmacological and Pharmaceutical Sci.*, **8**, 574 (2014).
- 25) THE COLLOIDAL DOMAIN Where Physics, Chemistry, Biology, and Technology Meet Second Edition, D. F. Evans, H. Wennerstrom, WILEY-VCH, New York (1999).

乳化剤フリーエマルジョンの調製における高出力バス型超音波照射機の可能性

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要 旨

本稿では、界面活性剤などの乳化剤を使用せず油と水を機械力により混合した乳化剤フリー (EF-) 水中油滴型 (O/W) エマルジョンの調製方法について報告する。市販の超音波洗浄機 (Bath-US; 42 kHz, 26 W) では15分間処理を行ってもドデカン (DD) を水中に完全に分散させることはできなかった。また、調製されたEF-DD/Wエマルジョン中のDD滴の平均直径は数マイクロメートル (μm) であった。市販のホーン型超音波ホモジナイザー (Horn-US; 19.5 kHz, 600 W) を用いてEF-DD/Wエマルジョンを調製した場合、EF-DD/Wエマルジョンは1分間程度の処理で調製され、調製されたEF-DD/Wエマルジョン中のDD滴の平均直径は1マイクロメートル (μm) 程度であった。著者らが開発した高出力バス型超音波照射機 (HPBath-US; 28 kHz, 300 W) を用いてEF-DD/Wエマルジョンを調製した場合も1分間程度の処理でEF-DD/Wエマルジョンは調製された。調製されたEF-DD/Wエマルジョン中のDD滴の平均直径は10分間の処理で数百ナノメートル (nm) となった。ローター式ホモジナイザー (RS-HG; 15,000 rpm, 800 W) を用いてEF-DD/Wエマルジョンを調製した場合は1分間程度の処理でEF-DD/Wエマルジョンは調製されたが、調製されたEF-DD/Wエマルジョン中のDD滴の平均直径は数マイクロメートル (μm) であった。さらに、Horn-USとHPBath-USを用いて調製されたEF-DD/Wエマルジョンの分散安定性は、Bath-USとRS-HGを用いて調製されたEF-DD/Wエマルジョンよりも高いことがわかった。これらのことから、HPBath-USはEF-O/Wエマルジョンの調製に有効であることが明らかとなった。さらに、流通用チャンバーをHPBath-USに装着するとEF-DD/Wエマルジョンを連続的に調製できることが明らかとなった。

キーワード：調製方法, 乳化剤フリー水中油滴型エマルジョン, 高出力バス型超音波照射機, 処理時間, 液滴径, 分散安定性, 連続調製