ELSEVIER

Food Chemistry 66 (1999) 327–331

Food Chemistry

www.elsevier.com/locate/foodchem

Viscosity change in oil/water food emulsions prepared using a membrane emulsification system

Y. Asano*, K. Sotoyama

Food Research and Development Laboratory, Morinaga Milk Industry Co., Ltd, 1-83, 5-Chome Higashihara, Zama-shi, Kanagawa-ken 228-8583, Japan

Received 18 November 1998; accepted 26 January 1999

Abstract

This paper reports viscosity measurements of oil/water (O/W) monodispersed emulsions of different droplet diameters obtained in a membrane emulsification system. Hydrophilic microporous glass membranes of different pore diameters were used to prepare O/W emulsions. The results showed that the droplet diameter of the emulsions varied with the average pore diameter of the membrane. The average droplet diameter was found to be about five times greater than the average membrane pore diameter. A correlation was found for the relationship between the average droplet diameter and the emulsion viscosity. As the dispersed droplet size became smaller, the total surface area of the droplets increased. Therefore, the emulsion viscosity and the relative viscosity increased. Few studies have reported the viscosity of O/W emulsions with droplet diameter of 5 μm or more and an oil phase concentration of 10 vol% or less. In the present study a correlation between the droplet diameter and the emulsion viscosity was statistically established. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Viscosity; Oil/water (O/W); Emulsion; Membrane; Droplet diameter

1. Introduction

It is known that the condition of dispersion particles affects the physical properties of food emulsions. The size of the dispersion particles influences the emulsion viscosity, taste, hardness and other properties (Sherman, 1971). The viscosity of liquid emulsions has already been studied (Matsumoto & Sherman, 1969; Parkinson, Matsumoto & Sherman, 1970). These studies have reported on the relationship of concentration and viscosity of emulsions with small droplet diameters of 5 μm or less. In this paper, we have studied the relationship of the viscosity of an oil/water (O/W) emulsion with droplet diameter of 5 μm or more at low concentration. To prepare these monodispersed emulsions, we used a membrane emulsification system (Katoh, Asano, Furuya, Sotoyama & Tomita, 1996), a new emulsification technology. The physical properties of these emulsions were measured and analyzed statistically.

2. Materials and methods

2.1. Membrane emulsification apparatus

In the membrane emulsification lab-apparatus (Fig. 1; Kiyomoto Iron & Machinery Works Co., Ltd), the continuous phase circulates outside of the Shirasu porous glass membrane tube (MPG, Ise Chemical Industries Co., Ltd.) in the vessel and the dispersion phase permeates from the interior of the MPG tubes under N_2 gas pressure (Katoh, Asano, Furuya & Tomita, 1995). The dimensions of the MPG tubes used were 10 mm outer diameter, 0.75 mm thickness, and 15 mm length. The average pore diameter of the membrane (Dm) was measured using a mercury porosimeter (Shimadzu Corporation, PC-9300).

2.2. Materials

For the oil phase, corn oil was used. For the water phase, deionized water was used. For the emulsifier, polyglycerol esters (PGE) were used. For the stabilizer, carnageenan was used. The additive amounts of each emulsifier and stabilizer were represented as wt% in the added phase.

---

* Corresponding author. Tel.: +81-462-52-3029; fax: +81-462-52-3017.

0308-8146/99/$ - see front matter © 1999 Elsevier Science Ltd. All rights reserved.

PII: S0308-8146(99)00068-0

Nomenclature

Dm	average pore diameter of membrane
Dp	average droplet diameter of emulsion
k	hydrodynamic interaction coefficient
n	total number of dispersed droplets
S	total surface area of dispersed droplets
Sd	standard deviation of droplet diameter
v	volume of a dispersed droplet
V	total volume of dispersion phase
α	coefficient of dispersion
η	emulsion viscosity
η/η_0	relative viscosity of emulsions
η_0	continuous phase viscosity
η_r	relative viscosity calculated by Moony's equation
ϕ	volume concentration of dispersion phase

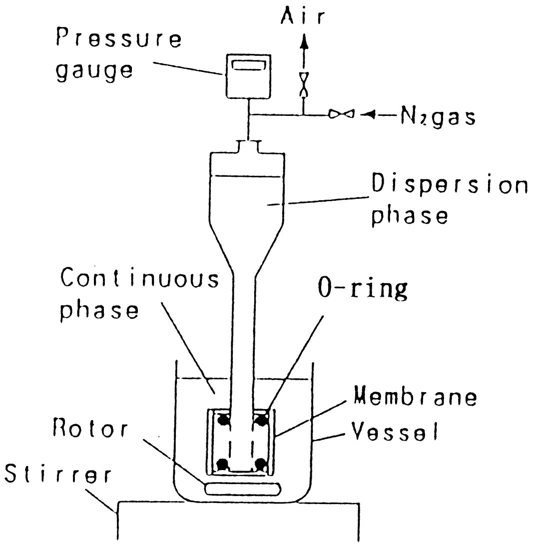

Fig. 1. Lab-scale emulsification apparatus.

2.3. Preparation of the emulsion

Before emulsification, the MPG tube was pre-soaked in the continuous phase in an ultrasonic bath for 30 min (Katoh et al., 1997). Then the MPG tube was set in the apparatus. Next, the dispersion phase was delivered into the MPG tube and, after extraction of the air from the interior of the MPG tube, the continuous phase was circulated in the vessel (Nakashima, Shimizu & Kawano, 1987). The permeant pressure of the dispersion phase was gradually increased and the dispersion phase was emulsified into the continuous phase by passage through the membrane. This operation was continued until the dispersion phase concentration of 10 vol% at the emulsifying temperature of 25°C was reached. The dispersion phase volume was calculated by measuring the total weight of the vessel. The permeant pressure of the dispersion phase at the time of emulsification was considered to be the emulsifying pressure. In the case of normal membrane emulsification, the emulsifying pressure was 10∼20 kPa higher than the emulsifying critical pressure (Nakashima, Shimizu & Kukizaki, 1991).

The average droplet diameter and the droplet distribution were measured using an image analyzer system (Nikon, Co. Ltd., Cosmozone R-500). The viscosity of the emulsion was measured using a vibration viscometer (Chichibu Onoda Co. Ltd, DJV-5000).

2.4. Assessment of monodispersed emulsion

As an index of monodispersion, a coefficient of dispersion (α) was adopted (Katoh et al., 1995).

$\alpha = \text{Sd}/\text{Dp}$ Sd, a standard deviation of the droplet diameter
Dp, the average droplet diameter of an emulsion

In the relationship between α values and the distribution of dispersed droplets of O/W emulsions observed under a microscope (Fig. 2), the smaller the α values, the more monodispersed the emulsions. We defined monodispersed emulsions for α equal or smaller than 0.35.

2.5. The water phase composition of O/W emulsions

When we consider the composition of the water phase as a continuous phase, it is important that its viscosity does not vary in the course of measuring as it provides an interfacial active force required for preparing O/W emulsions. For the emulsifier and stabilizer, PGE at 0.5% and carnageenan of 0.04% were added to the ionized water, and a solution with viscosity of 3.7 mPa s at 25°C was prepared as the water phase. Then its viscosity was confirmed constant by measuring after a day.

3. Results and discussion

3.1. Preparation of O/W emulsions using a membrane emulsification system

In the O/W emulsions, composed of deionized water/PGE/corn oil, prepared using membranes with average pore diameter of Dm = 1.1, 2.9, 4.5, 5.7 µm (Table 1) and in the microscopic observations [Fig. 3(a)], the value of α was 0.2∼0.3, so it was evident that each emulsion was monodispersed. From the results of Dp in the emulsions

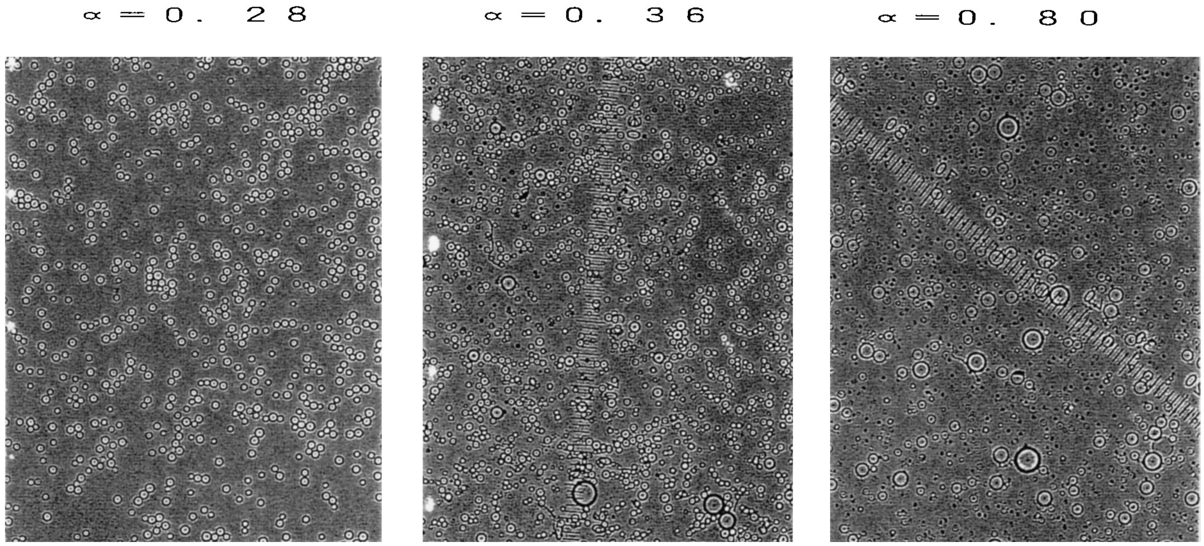

Fig. 2. Micrographs of O/W emulsions and α.

Table 1
Data of O/W emulsions prepared using membrane emulsification[a]

	Directly after emulsification				After a month at room temperature			
D m[b] (μm)	1.1	2.9	4.5	5.7	1.1	2.9	4.5	5.7
D p (μm)	5.48	14.19	21.82	27.71	5.92	14.21	21.57	29.97
α (–)	0.20	0.27	0.23	0.25	0.23	0.29	0.23	0.26
η (mPa.s)	4.33	4.11	3.98	3.93	4.40	4.21	4.13	3.97
η_0 (mPa.s)	3.7	3.7	3.7	3.7				
η/η_0 (–)	1.17	1.11	1.08	1.06				
Moony's equation k (–)	3.083	3.080	3.079	3.079				
η_r (–)	1.435	1.435	1.435	1.435				
V ($\times 10^{-6}$ m^3)	38.89	38.89	38.89	38.89				
ϕ (–)	0.1	0.1	0.1	0.1				
v ($\times 10^{-21}$ m^3)	0.086	1.496	5.440	11.141				
n ($\times 10^{19}$/m^3)	116.30	6.68	1.84	0.897				
S ($\times 10^{10}$ m^2/m^3)	10.97	4.23	2.75	2.18				

[a] v, n an S were calculated using D p, V and ϕ.
[b] See Nomenclature for explanation of symbols.

prepared with each Dm, there was found to be a significant difference between each Dp, at a level of significance of 1%. Therefore, these results were used to study and compare the physical properties of each emulsion.

3.2. Relationship between Dm and Dp

In the relationship between Dm and Dp (Table 2 and Fig. 4), a straight line ($Y = 4.8\ X + 0.17$) with a correlation coefficient of 0.999 was plotted and Dp was found to be about 5 times greater than Dm in these compositions and conditions. The results demonstrated that the dispersion droplet diameter basically depends upon the membrane pore diameter.

3.3. Stability of the O/W emulsions prepared by membrane emulsification

In the micrographs obtained directly after emulsification and 1 month after emulsification [Fig. 3(a) and (b)], there were no differences between these emulsions. So, this indicated that a stable monodispersed emulsion could be prepared even with the droplet diameter of 5 μm or more. Generally, emulsions with a large dispersed droplet size have inferior stability. However, within the scope of this experiment, this trend was not observed and it was confirmed to be possible, using the membrane emulsification method, to prepare O/W emulsions

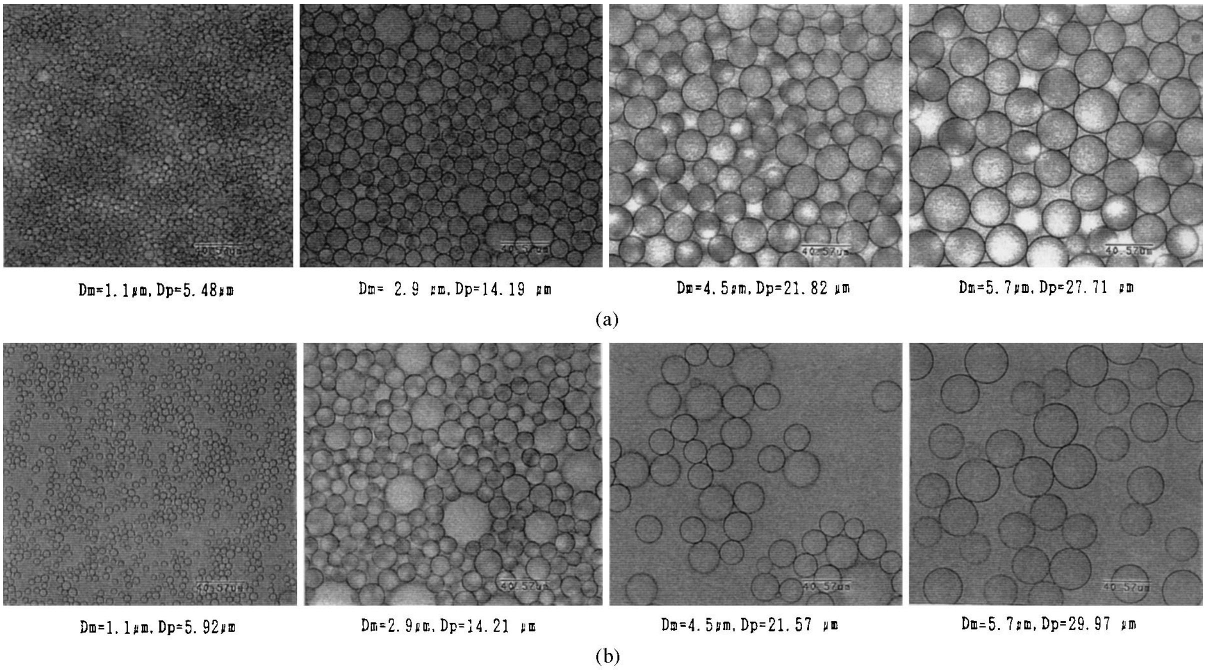

Fig. 3. (a) Micrographs of O/W emulsions by membrane emulsification; (b) micrographs of O/W emulsions after a month at room temperature.

Table 2
Correlation coefficients among each data

	Dm	Dp	η	η/η_0	S
Dm	1				
Dp	0.9999	1			
η	−0.9815	−0.9811	1		
η/η_0	−0.9855	−0.9854	0.9983	1	
S	−0.9208	−0.9206	0.9752	0.9735	1

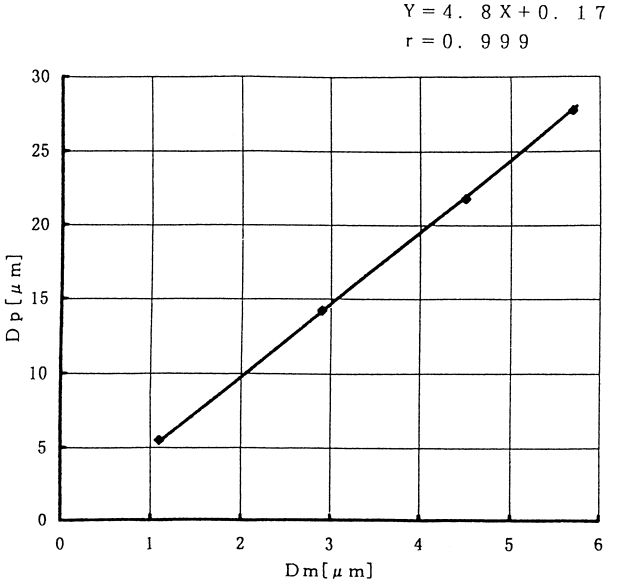

Fig. 4. Relationship between Dm and Dp.

of almost the same level of stability with a Dp range from 5 μm to 30 μm.

3.4. The effect of Dp and the total surface area of the droplets (S) on emulsion viscosity

S was calculated using the total volume of the dispersion phase (V) and the total number of droplets (n). In the relationship between Dp and the relative viscosity of emulsions (η/η_0), and between S and η/η_0 (Figs. 5 and 6) it was found that as Dp increased, η/η_0 decreased and, as S increased, η/η_0 increased. These results were similar to those of Mori and Ototake (1956) and Saunders (1961) who reported that, as S increased, the emulsion viscosity increased. Sherman (1961) did not find a correlation between the droplet diameter and viscosity of ice cream mixtures at 1.5 μm or less at an oil phase concentration of 10 vol%. However, in the present study, a correlation between droplet diameter of 5 μm or more and the emulsion viscosity at an oil phase concentration of 10 vol%, was statistically established.

The difference of η/η_0 and the relative viscosity (η_r) was calculated by Moony's equation (Moony, 1951):

$$\eta_r = \exp(a.\phi/(1 - k\cdot\phi))$$

where α is 2.5 when the droplets are stable and no droplet aggregate is present, and ϕ is the volume concentration of the dispersion phase and is 0.1 in this experiment, and that of Matsumoto et al. (1969). k is a hydrodynamic interaction coefficient which empirically depends on Dp according to

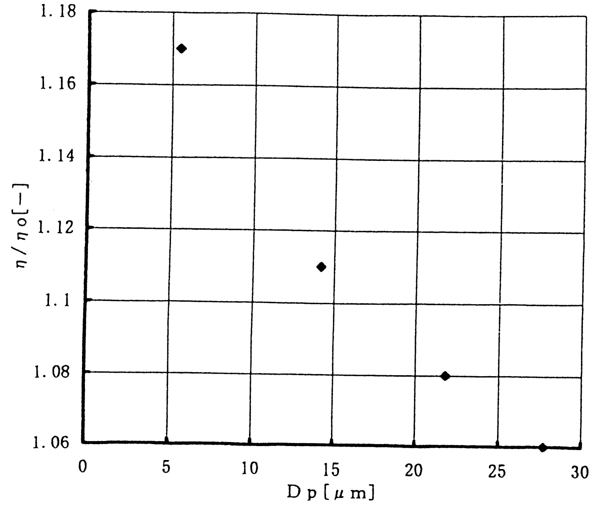

Fig. 5. Relationship between Dp and η/η_0.

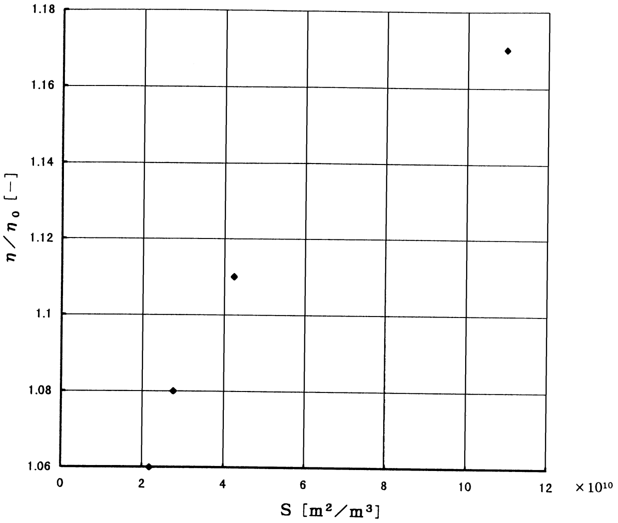

Fig. 6. Relationship between S and η/η_0.

$$k = 1.079 + \exp(0.01008/\mathrm{Dp}) + \exp(0.00290/\mathrm{Dp}^2).$$

η_r calculated using these equations is shown in Table 1. These η_r were constant, even if Dp changed from 5 to 30 μm. Then there was no fit between η/η_0 and η_r.

References

Katoh, R., Asano, Y., Furuya, A., Sotoyama, K., & Tomita, M. (1996). Preparation of food emulsions using a membrane emulsification system. *Journal of Membrane Science, 113*, 131–135.

Katoh, R., Asano, Y., Furuya, A., Sotoyama, K., Tomita, M., & Okonogi, N. (1997). A method of preventing dispersed droplets from crushing using a membrane. *Journal of the Japanese Society for Food Science and Technology, 44*, 233–237.

Katoh, R., Asano, Y., Furuya, K., & Tomita, M. (1995). Conditions for preparation of O/W food emulsions using a membrane emulsion system. *Journal of the Japanese Society for Food Science and Technology, 42*, 548–555.

Matsumoto, S., & Sherman, P. (1969). The viscosity of microemulsions. *Journal of Colloid Science, 30*, 525–536.

Moony, M. (1951). The viscosity of a concentrated suspension of spherical particles. *Journal of Colloid Science, 6*, 162–171.

Mori, Y., & Ototake, T. (1956). On the viscosity of suspensions. *Chemical Engineering of Japan, 20*, 488–494.

Nakashima, T., Shimizu, M., & Kawano, M. (1987). Articles of porous glass and process for preparing the same. US patent, 4657875.

Nakashima, T., Shimizu, M., & Kukizaki, M. (1991) *Membrane emulsification operation manual.* Miyazaki, Japan: Industrial Research Institute of Miyazaki.

Parkinson, C., Matsumoto, S., & Sherman, P. (1970). The influence of particle-size distribution on apparent viscosity of non-Newtonian dispersed system. *Journal of Colloid Science, 33*, 150–160.

Saunders, F. L. (1961). Rheological properties of monodisperse latex systems. *Journal of Colloid Science, 16*, 13–22.

Sherman, P. (1961). Rheological methods for studying the physical of emulsifier films at the oil–water interface in ice cream. *Food Technology*, September, 394–399.

Sherman, P. (1971). *Emarusyon no Kagaku (Emulsion science)* (p. 60). Tokyo: Asakura Press.