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Studies on an Emulsion Formation by Flow Jet Mixer. Comparison of Flow Jet Mixer with Agitator and Colloid-Mill on Emulsion Formation[1)]

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In this study, one of the purpose is the comparison of the capabilities of machines as emulsifying equipments. The other purpose is to evaluate the stability of emulsion prepared above equipments, from the changes of turbidities in the course of time.

The Flow Jet Mixer, the Agitator, and the Colloid-Mill were used as emulsifying equipments. Mixtures of n-C_7H_{16} and CCl_4 were employed as the dispersed phase. Each sample of emulsion was quickly photographed under an optical microscope, in succession, the mean length diameter and the particle size distribution were calculated. Furthermore, the shear velocity du/dx could be calculated at three emulsifying equipments.

The following results were obtained:

(1) The size distribution curves of emulsions prepared by Colloid-Mill and Flow Jet Mixer had a sharper and narrower shape.

(2) The values of shear velocity were 6.37×10^5 at Agitator, 1.94×10^6 at Flow Jet Mixer, and 5.70×10^6 (1/sec) at Colloid-Mill, respectively, when the concentration of dispersed phase was kept at 3.55% (v/v).

Keywords——Agitator; Colloid-Mill; emulsion; energy efficiency; Flow Jet Mixer; mean length diameter; microphotographic method; oil-in-water; Turbidimeter

The selection of the kind of emulsifying equipments fitting to the emulsion system is considered as one of the most important factors affecting the formation of emulsions.[3)] The available commercial machines, *viz.*, mixing,[4)] colloid milling,[5)] and homogenizing,[6)] cover a wide range of capacities, from small laboratory models to large industrial units. These machines serve to obtain a fundamental information for some of the guiding principles of emulsification.[7)]

In this study, we select the typical machines such as the Flow Jet Mixer, the Agitator,[8)] and the Colloid-Mill[9)] as an emulsifying equipment. Flow Jet Mixer as a new apparatus for preparing emulsion was based on a revolutionary principle.

In this paper, therefore, the particle size distribution of oil droplets in the oil-in-water type emulsion formation by Flow Jet Mixer will be experimentally examined in relation to the revolution numbers of stator and the concentration of the dispersed phase to continuous phase.

Futhermore, the comparison of the capabilities of three machines on the emulsion formation is done and discussed.

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3) W.A. Rodger, V.G. Trice, and J.H. Rushton, *Chem. Eng. Progr.*, **52**, 515 (1956).
4) G.E. Langlois and T. Vermeulen, *Chem. Eng. Progr.*, **51**, 81 (1955).
5) J.A. Mc. Donough, J.W. Tomme, and C.D. Holland, *A. I. Ch. J.*, **6**, 615 (1960).
6) H. Kamura, *Kogyo Zairyo*, **19**, 25 (1971).
7) P. Sherman, "Emulsion Science," Academic Press, New York, p. 6.
8) S. Tsukiyama, A. Takamura, and N. Nakura, *Yakugaku Zasshi*, **94**, 490 (1974).
9) S. Tsukiyama, A. Takamura, and N. Nakura, *Yakugaku Zasshi*, **93**, 231 (1973).

Experimental

Equipment——A sketch of the apparatus employed is shown in Fig. 1. The system comprises from Flow Jet Mixer Model MW-B-150-S (Funken Co., Ltd.) for emulsification and Feeder Model CR-5D (Elepon Chem. Eng. Co., Ltd.) for liquids.

For instance, in preparation of oil-in-water type emulsion, water having a emulsifier, as a continuous phase, is feeded into mixing disk continuously by overflow cone, indicated as 3 in figure, of the mixer. At the same time, the oil, as dispersed phase, which is feeded continuously towards center of the mixer is developed into liquid film by spreader cone, indicated as 2 in figure, of the mixer and mix the overflowing thin water film through over the flow cone. Both liquids are emulsified continuously and in seconds due to a shear force generated by mixing disk accompanied with a resonance vibration of induced air.

A sketch of Agitator[10] employed is shown in Fig. 2. The clear acrylated resin agitation tank with 4-baffles was 150 mmϕ in diameter and 210 mm in depth. A stainless-steel agitation impeller of 49.00 mmϕ was of the standard Rushton type with 6-blades. The tank was surrounded by a water jacket in order to keep the temperature of the liquid within it at 20.0 °. Then, the liquids were agitated at 660 rpm for 60 min.

Figure 3 illustrates the apparatus of Colloid-Mill. This apparatus was of SL-type of Asahi Homogenizer Co., Ltd. and the diameter of stator was 76.90 mmϕ.[11] The liquids entered continuously at the top through the tubes in the stator frame, flow through the narrow clearance between the stator and rotor, and finally make their exit. The rotor is dynamically balanced and can rotate at speeds of 13600 rpm.

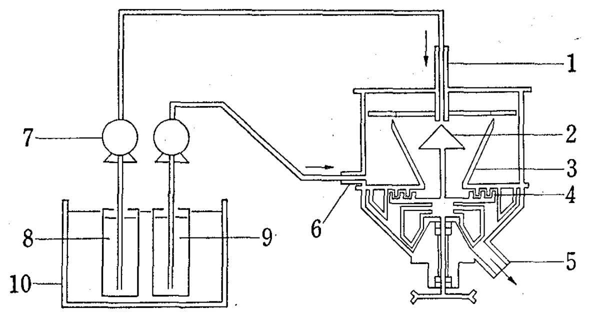

Fig. 1. Apparatus of Flow Jet Mixer

1: entrance, 2: conical spreader 3: over flow cone, 4: pin, 5: exit, 6: entrance, 7: pump, 8: vessel(dispersed phase) 9: vessel(continuous phase) 10: thermostat.

Experimental Procedures of Emulsification——Distilled water was used as the continuous phase. Mixtures of n-C_7H_{16} and CCl_4 were used as the dispersed phase. The total concentration of dispersed phase ranged from 3.55 to 27.50% (v/v). The density of dispersed phase was adjusted so as to be same as that of the continuous phase (1.000 gr/cm^3).[12] The emulsifying agent was not used in this study. Interfacial tensions were measured by the ring method (Kyōwa Chemical Co., Ltd.).[13] And the viscosities of the liquid were measured by a rotating viscometer (Shibaura Electric Co., Ltd.) at $20.0+0.1$°.[14] The concentration of dispersed phase, the density ratio of two phases, the interfacial tensions between two phases, the range of revolution number, and the range of shearing power are summarized in Table I. These experimentals were examined under the same physical conditions without the differences of the shear velocity.

Each sample was photographed immediately with a Nikon AFM camera equipped with a microscope. The diameters of the droplets were measured with a micrometer scale which had been photographed and

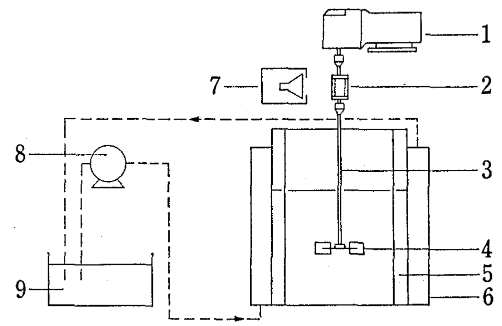

Fig. 2. Apparatus of Agitator

1: motor 2: torque torque meter 3: shaft 4: impeller 5: baffle 6: tank 7: storobo scope 8: pump 9: thermostat.

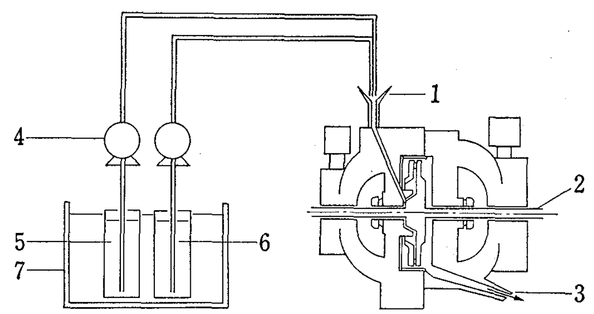

Fig. 3. Apparatus of Colloid-Mill

1: entrance 2: shaft 3: exit 4: pump 5: vessel(dispersed phase) 6: vessel(continuous phase) 7: thermostat.

10) S. Tsukiyama, H. Takahashi, I. Takashima, and S. Hatano, *Yakugaku Zasshi*, **91**, 305 (1971).
11) S. Tsukiyama, A. Takamura, and M. Nakano, *Yakugaku Zasshi*, **93**, 231 (1973).
12) S. Tsukiyama and A. Takamura, *Shikizai Kyōkaishi*, **48**, 151 (1975).
13) S. Tsukiyama and A. Takamura, *Yakugaku Zasshi*, **93**, 785 (1973).
14) *a*) S. Tsukiyama, A. Takamura, Y. Fukuda, and M. Koishi, *Bull. Chem. Soc. Japan*, **48**, 3561 (1975); *b*) S. Tsukiyama, A. Takamura, Y. Fukuda, and M. Koishi, *Chem. Pharm. Bull.* (Tokyo), **24**, 414 (1976).

TABLE I. Physical Properties of Emulsion and Mechanical Conditions in Three Emulsifying Methods

Equipment	Concentration of dispersed phase c_d (%, v/v)	Density ratio ρ_d/ρ_c (—)	Interfacial tension $\lvert\gamma_d-\gamma_c\rvert$ (dyn/cm)	Revolution number N (rpm)	Shearing power P (watt)
Agitator	3.55—27.50	1.000	48.50	660	230
Flow Jet Mixer	3.55—27.50	1.000	48.50	400—2400	530—550
Colloid-mill	3.55—27.50	1.000	48.50	13600	860

enlarged under the same conditions. The number of droplets measured was one thousand[15] in all cases. The mean length diameter, the mean surface diameter, the mean volume diameter, the standard deviation, and the particle size distribution were calculated by means of a Seiko-S-301 computer.

Results and Discussion

Effect of the Concentration of Dispersed Phase on the Particle Size Distribution (Flow Jet Mixer)

The mean length diameters and the particle size distributions were measured under the various experimental conditions. On the other hand, the concentration of dispersed phase were kept at 3.55, 13.76, and 27.50% (v/v), respectively. The ranges of revolution number of Flow Jet Mixer were from 400 to 2400 rpm. Figure 4 gives the relation between the mean length diameters and the concentration of dispersed phase. From Fig. 4, it is found that the mean length diameter has a larger values at a lower revolution number. Furthermore, the mean length diameter increases noticeably with increasing concentration of dispersed phase, indicating that it is greatly affected by the concentration of dispersed phase.

The particle size distributions calculated by using the length diameters measured at three different concentration of dispersed phase, at 888 rpm, are shown in Fig. 5. In these cases, the intervals of distribution was 10 μm and the concentration of dispersed phase were 3.55, 13.76, and 27.50% (v/v), respectively. At each ratio, the size distribution had only one peak, at a particle size of about 35 μm. Furthermore, the size distributions took a narrower and sharper shapes with decrease of the concentration of dispersed phase.

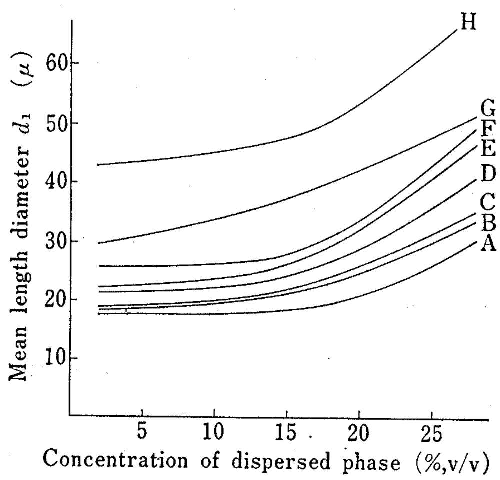

Fig. 4. Relations between Mean Length Diameter and Concentration of Dispersed Phase

A: 2400 rpm B: 2120 rpm C: 1900 rpm D: 1668 rpm E: 1460 rpm F: 1258 rpm G: 1072 rpm H: 888 rpm.

Figure 6 gives the relation between the cumulative frequency of particle number and the particle size in the three different volume ratios of dispersed phase at 888 rpm. The cumulative size distribution curves increased steeply and showed the almost linear relation, and thus the particle size distributions had a tendency of the logarithmic normal ones when the emulsion was formed by the Flow Jet Mixer in absence of the emulsifying agent.

Effect of the Revolution Number on the Particle Size Distribution (Flow Jet Mixer)

Figure 7 gives the relation between the mean length diameter and the revolution number at three different concentration of dispersed phase. It was obvious from Fig. 7 that the mean

15) H.H.G. Jellinek, *J. Soc. Chem. Ind.* (London), **69**, 225 (1950).

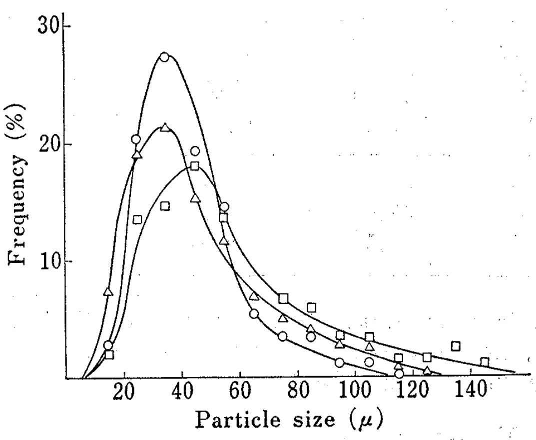

Fig. 5. Comparison of Particle Size Distributions in the Three Different Concentration of Dispersed Phase at Revolution Number of 888 rpm

○, 3.55%(v/v) △, 13.76%(v/v) □, 27.50%(v/v)

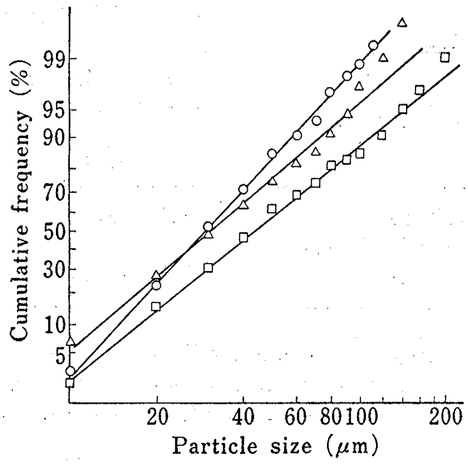

Fig. 6. Comparison of Particle Size Distribution in the Three Different Concentration of Dispersed Phase at Revolution Number of 888 rpm

○, 3.55%(v/v) △, 13.76%(v/v) □, 27.50%(v/v).

length diameter of oil droplets remarkably decreased at lower revolution number ranged in 400—800 rpm and then had a gentle slope as the revolution number increased more higher ones. Accordingly, it seemed that the emulsion formation was greatly affected when the revolution number of Flow Jet Mixer was low.

Figure 8 gives the particle size distribution at three different revolution numbers. In this case, the interval of distribution was 10 μm. And Fig. 9 shows the cumulative frequency of particle size at three different revolution numbers. The revolution numbers were 888, 1258, and 2120 rpm, respectively. In two figures, the concentration of dispersed phase was kept at 13.76% (v/v). From comparison of these data, it can clearly be understood that the revolution number strongly affected the particle size distribution of emulsion, too. That is to say, the liquids must be applied more vigorous shear force in order to get smaller droplets. To be more specific, the shear force must be strong so that the bigger droplets, which are formed initially, are torn into smaller droplets.

Comparison of the Mean Length Diameter and the Homogeneity at the Three Different Emulsifying Methods

The comparisons of particle size distributions of emulsion prepared by the three different emulsifying methods are shown in Fig. 10. The two size distribution curves at Colloid-Mill and Flow Jet Mixer have a sharper and narrower shape. On the contrary, the remarkably broader size distribution curve was observed at Agitator. From these results, it was thought that the emulsion formation was greatly affected by the kinds of emulsifying equipments. In particular, this influence was clearly appeared on the shape of particle size distribution. The mean length diameter was calculated by the measurements of the sizes of 500-particle number at each experimental condition. The comparisons of mean length diameter at three emulsifying methods are shown in Table II. The mean length diameter of particle at Colloid-Mill was smaller than that at Flow Jet Mixer. Owing to these above results, it was found that the particle size of droplets were strongly affected by the kind of machine of preparing emulsion.

Table II gives the standard deviation σ and the normalized ones σ/d_1 at three emulsifying methods. The value of σ increased in the order of Colloid-Mill, Flow Jet Mixer, and Agitator, in particular, this value was remarkably high at Agitator.

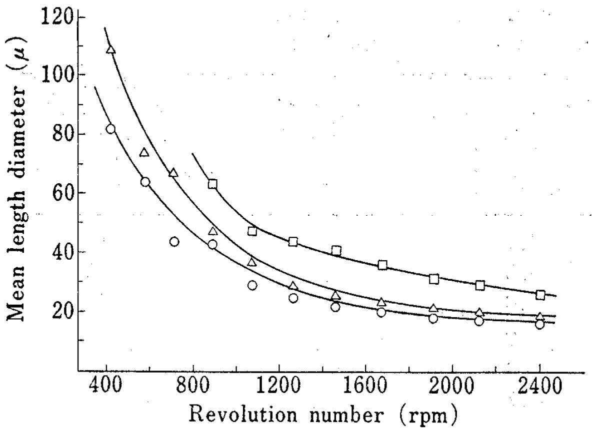

Fig. 7. Relations between Mean Length Diameter and Revolution Number at Three Different Concentration of Dispersed Phase

○, 3.55%(v/v) △, 13.76%(v/v) □, 27.50%(v/v).

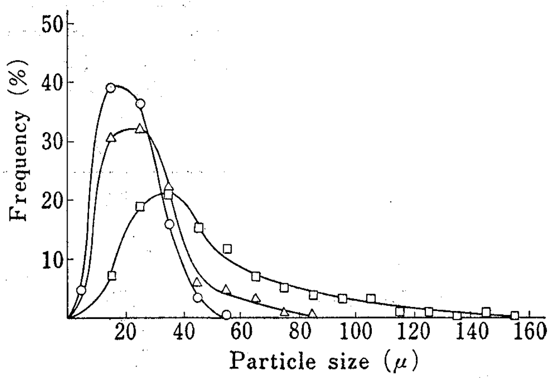

Fig. 8. Comparison of Particle Size Distributions at Three Different Revolution Number

○, 2120 rpm △, 1258 rpm □, 888 rpm.
Concentration of dispersed phase: 13.76%(v/v).

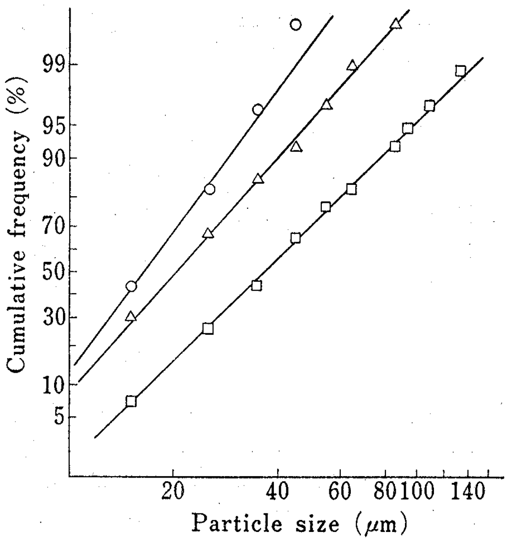

Fig. 9. Comparison of Particle Size Distributions at Three Different Revolution Numbers

○, 2120 rpm △, 1258 rpm □, 888 rpm
Concentration of dispersed phase: 13.76 %(v/v).

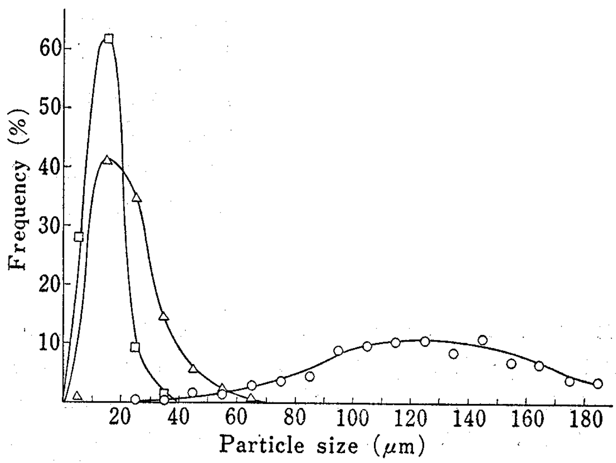

Fig. 10. Comparisons of Particle Size Distributions of Emulsion Prepared by Three Emulsifying Method

○, Agitator △, Flow Jet Mixer □, Colloid-Mill.
Concentration of dispersed phase: 13.76 %(v/v).

In order to discuss the homogeneity of emulsified droplets, it is proposed that the normalized standard deviation σ_s is defined as the ratio of the standard deviation σ and the mean length diameter d_l (cm). Then, it is given by,[16]

$$\sigma_s = \frac{\sigma}{d_l} = \frac{\sqrt{\sum(d_i - d_l)^2/n}}{d_l} \qquad (1)$$

The comparisons of normalized standard deviations at three emulsifying methods are shown in Table II, too. The values of σ_s at Flow Jet Mixer and Colloid-Mill are slightly larger ones compared with that at agitator. Reason for this was that the mixing time by Agitator was much longer than the other two methods. Thus, the formation of emulsion requires a sufficiently long mixing time.

16) S. Tsukiyama, A. Takamura, Y. Wakamatsu, and I. Takashima, *Yakugaku Zasshi*, **93**, 191 (1973).

TABLE II. Comparisons of Mean Length Diameter, Standard Deviation, and Normalized Deviation of Emulsion Prepared Three Emulsifying Methods

Emulsifying equipment	Concentration of dispersed phase %(v/v)	Mean length diameter d_l (μm)	Standard deviation σ (−)	Normalized deviation σ/d_l (−)
Agitator	3.55	66.1	23.0	0.348
	13.76	126.0	38.4	0.305
	27.60	182.0	68.3	0.375
Flow Jet Mixer	3.55	21.7	8.23	0.379
	13.76	23.8	11.0	0.462
	27.50	36.7	23.1	0.629
Colloid-Mill	3.55	7.4	2.82	0.381
	13.76	13.2	5.28	0.398
	27.50	18.4	9.43	0.512

Calculation of the Energy Efficiency and the Shear Velocity of Mixing

When the total interfacial area of the emulsion, S(cm^2/gr), increases from S_1 to S_2, the increase in the total interfacial energy, E_s (erg/gr), is calculation by the following equation:

$$E_s = \gamma(S_2 - S_1) = \frac{6\gamma}{\rho_d}\left(\frac{1}{d_{sv_2}} - \frac{1}{d_{sv_1}}\right) \tag{2}$$

where σ is the interfacial tension of two liquids (dyn/cm), ρ_d is the density of dispersed phase (gr/cm^3), and d_{sv} is the mean surface-volume diameter (cm), respectively. In order to investigate to what extent the mixing energy affects the increase in the total interfacial energy, the energy efficiency of mixing is defined as:[17]

$$\eta = \frac{E_s}{E_A} = \frac{\gamma(S_2 - S_1)}{P \cdot \theta} \tag{3}$$

where E_A is the agitation energy (erg/gr), P is the agitation power (erg/sec), and θ is the agitation time (sec), respectively. Equation (3) gives the ratio of the total interfacial energy to the mixing energy, and then this is named the energy efficiency of mixing in mechanical dispersion. If the energy efficiency can be calculated under an optional mixing condition, it will be very useful for the research which looks for the optimum physical and mechanical condition to obtain a small size distribution.

In this study, the parameters characterizing mixing are chosen to be the kinds of emulsifying equipments and the concentration of dispersed phase.

The calculated values of η are given in Table III. When the concentration of dispersed phase were 3.55% (v/v), the values of η were 0.869×10^{-5} at Agitator, 7.68×10^{-4} at Flow Jet Mixer, and 2.34×10^{-3} at Colloid-Mill, respectively. Furthermore, the value of η showed a larger ones over the three emulsifying equipments with decrease of the concentration of dispersed phase.

Nextly, the shear velocity in the emulsifying equipment was calculated and discussed. The deformation and breakup of droplets in the shear flow has already theorized by Taylor[18] and proved experimentally by Mason.[19] The dynamic balance at the transition state is represented as following,

$$\mu_c \cdot f(\mu) \frac{du}{dx} = \frac{\gamma}{d_{sv}} \tag{4}$$

17) S. Tsukiyama, A. Takamura, Y. Fukuda, and M. Koishi, *Chem. Pharm. Bull.* (Tokyo), **24**, 414 (1976).
18) G.I. Taylor, *Proc. R. Soc.*, **A138**, 41 (1932).
19) S.G. Mason and W. Bartok, *J. Colloid Sci.*, **16**, 210 (1961).

where, μ_c is the viscosity of continuous phase (gr/cm·sec), and $f(\mu)$ is the correction coefficient of viscosity (—), respectively. From Eq. (4), the shear force $\mu_c f(\mu) du/dx$ or the shear velocity du/dx can be experimentaly obtained at each emulsifying equipment. These results are also summarized in Table III. For example, the values of shear velocity were 6.37×10^5 at Agitator, 1.94×10^6 at Flow Jet Mixer, and 5.70×10^6 (1/sec) at Colloid-Mill, respectively, when the concentration of dispersed phase was 3.55% (v/v). The shear velocity yield at Colloid-Mill was most violently, in succession, in the order of Flow Jet Mixer and Agitator. To get smaller oil droplets, more vigorous mixing should be applied to the liquids. The breakup of smaller oil droplets would happen only when the larger velocity gradients existed in the surrounding of the droplets. The Colloid-Mill had more furious shear velocity than the other two emulsifying equipments.

TABLE III. Experimental and Calculated Values

Equipment	Concentration of dispersed phase (%,v/v)	Specific interfacial area S (cm²/gr)	Specific interfacial energy E_s (erg/gr)	Mixing energy E_A (erg/gr)	Energy efficiency of mixing η (—)	Shear velocity du/dx (1/sec)
Agitator	3.55	742	3.5987×10^4	4.14×10^9	0.869×10^{-5}	6.37×10^5
	13.76	401	1.9449	4.19	0.464	2.13
	27.50	266	1.2901	4.32	0.299	0.552
Flow Jet Mixer	3.55	2042	9.9038×10^4	1.29×10^8	7.68×10^{-4}	19.43×10^5
	13.76	1693	8.2111	1.18	6.96	11.38
	27.50	899	4.3602	1.05	4.15	2.68
Colloid-Mill	3.55	6227	30.200×10^4	1.29×10^8	2.34×10^{-3}	56.99×10^5
	13.76	3427	16.621	1.18	1.41	20.53
	27.50	2185	10.597	1.05	1.01	5.33

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