

### Liquid-Liquid Dispersion on Mechanical Agitation.<sup>1)</sup> Effect of the Viscosity Ratio $\mu_d/\mu_c$ on the Energy Efficiency of Agitation

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This paper deals with the effect of the ratio of viscosities of dispersed and continuous phases,  $\mu_d/\mu_c$ , on the energy efficiency of agitation.

The viscosity ratios used were 0.836, 0.271, 0.0992, 0.0280 and 0.00133. The photographs of drops were taken by a microscopic method and the values of torque were measured with a rotary torque meter. Since, the values of mean diameter and particle size distribution were calculated, and further the energy efficiency of agitation was evaluated.

And then, the following results were obtained.

(1) When the viscosity ratio  $\mu_d/\mu_c$  became smaller than unity,  $N_{We}$  for each revolution number increased more larger values.

(2) The energy efficiency of agitation gradually decreased according as the values of  $d_{sv}$  became smaller and smaller.

If the relation between the interfacial energy and the mechanical or physical function can be synthetically formularized, it would be very useful for a scale-up of the emulsifying experimental equipment.<sup>3)</sup> There were many dynamic factors which affect emulsion formation. They are tank diameter, impeller diameter, revolution number, agitation time and so on. While the physical factors were viscosity ratio of dispersed and continuous phase,  $\mu_d/\mu_c$ , specific gravity, interfacial tension, emulsifying agent, temperature and so on.

On the studies of the chemical engineering, Rushton<sup>4)</sup> and co-workers<sup>5,6)</sup> had already found the approximate quantitative variation for propeller mixers in tanks leaving out the viscosity factor. Moreover, Sprow<sup>7)</sup> and Langlois<sup>8)</sup> have attempted to correlate the specific interfacial area with the Weber number  $D_i^3 N^2 \rho / \gamma$ , which is measure of the ratio of shear force to surface force in the system. These researches give a few reasonable description of the phenomena for turbine and propeller mixers.

We have investigated the liquid-liquid dispersion on mechanical agitation from the point of pharmaceutical engineering view. In our previous paper, we have already reported the efficiency of agitation energy,<sup>9)</sup> and the increasing rate of droplets number.<sup>10)</sup>

In this paper, it is purpose to investigate the effect of the viscosity ratio  $\mu_d/\mu_c$  for the energy efficiency of agitation. The following three points of view are discussed experimentally.

- (1) The relation between power number and Reynolds number.
- (2) The relation between Weber number and viscosity ratio  $\mu_d/\mu_c$ .
- (3) Effect of the viscosity ratio  $\mu_d/\mu_c$  on the energy efficiency of agitation.

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## Theoretical

### Reynolds Number and Weber Number

The value of Reynolds number shows the intensity of agitation flow in a tank. Reynolds number is represented in dimensionless terms, and it is defined in Eq. (1) as the ratio of the force of inertia to the viscosity force.

$$N_{Re} = \frac{\rho_c \cdot u^2}{\mu_c \cdot u / D_I} = \frac{D_I \cdot u \cdot \rho_c}{\mu_c} \quad (1)$$

Generally,  $N_{Re}$  below 2000 is called to a laminar region,  $N_{Re}$  range of 2000 to 4000 is a critical region, and  $N_{Re}$  above 4000 is a turbulent region.<sup>11)</sup>

Meanwhile, a behavior of droplets within the agitation flow is represented as the relative size of Weber number. That is, Weber number is a dimensionless term, and is defined by Eq. (2) as the ratio of the force of inertia to the surface force.<sup>12)</sup>

$$N_{We} = \frac{\rho_c \cdot u^2}{\gamma / d_{sv}} = \frac{d_{sv} \cdot \rho_c \cdot u^2}{\gamma} \quad (2)$$

Thus, the above equation shows that the drop is easily possible to be broken up even with a slight agitation force as the value of  $N_{We}$  is smaller. The value of  $N_{We}$  is also greatly suffered with the effect of viscosity.

If the value of average diameter or specific interfacial area can be related with  $N_{Re}$  or  $N_{We}$ , it would have many merits for discussion the formation mechanism of emulsion and a scale-up of the equipment.

### Agitation Energy

When a liquid-liquid dispersion is carried out in an agitation tank, the input power of agitation is generally given in Eq. (3).<sup>13)</sup>

$$P = N_p \frac{\rho_c \cdot N^3 \cdot D^5}{g_c} \quad (3)$$

where,  $N_p$  denotes a power number which is a function of an impeller type or Reynolds number and  $g_c$  is a gravitational conversion number. From Eq. (3), if physical properties such as a viscosity and a gravity of dispersed phase are constant, the input power of agitation is found to be proportional to the product of third power of revolution number and fifth power of impeller diameter. Furthermore, the relation between a rotary torque given by an impeller and an input power of agitation is written as:

$$P = \omega \cdot T \quad (4)$$

where  $\omega$  denotes as:

$$\omega = 2 \cdot \pi \cdot N \quad (5)$$

Accordingly, a rotary torque is rewritten as:

$$T = k'(\rho_c \cdot N^2 \cdot D^5) \quad (6)$$

Therefore, a rotary torque is proportional to the product of second power of revolution number and fifth power of impeller diameter.

Besides, an agitation energy given to a liquid-liquid dispersion is given as the product of input power and agitation time.

$$E_A = P \cdot \theta \quad (7)$$

Since a rotary torque is measured on this experiment, an agitation energy is obtained by substituting Eq. (4) and Eq. (5) to Eq. (7).<sup>14)</sup>

11) J.J. Martin, *Trans. A. I. Ch. E.*, **42**, 777 (1946).

12) C. Weber, *Z. Angew. Math. Mech.*, **11**, 136 (1931).

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$$E_A = 2\pi \cdot N \cdot T \quad (8)$$

### Energy Efficiency of Agitation

Mean surface-volume diameter of droplet is represented as:

$$d_{sv} = \frac{\sum d_i^3}{\sum d_i^2} = \frac{\sum d_i^3/n}{\sum d_i^2/n} = \frac{d_v^3}{d_s^2} \quad (9)$$

On the other hand, specific interfacial area per 1 cm<sup>3</sup> of dispersed phase is given by:

$$S = 6/d_{sv} \quad (10)$$

And a specific interfacial energy is written by:

$$E_s = \gamma \cdot S = 6\gamma/d_{sv} \quad (11)$$

So, in order to investigate how the effect of agitation energy contributes to the increasing of specific interfacial energy, an energy efficiency is defined as:

$$\eta = \frac{E_s}{E_A} = \frac{\gamma \cdot S}{P \cdot \theta} \quad (12)$$

If an agitation energy is calculated by using Eq. (12), it would be a valuable guide for the research of the physical and mechanical optimum conditions to make a good emulsion.

### Experimental

**Measurement of Agitation Torque**—Both an agitation tank and an impeller employed were the same ones described in our previous report.<sup>15)</sup> The rotary torque meter used was the Yamazaki SS-IR type (Yamazaki Seiki Laboratories). The stroboscope was Sugawara PS-240 type (Sugawara Laboratory).

The torque meter was tightly adapted, not to cause a precessional motion, to the middle of rotary shaft of agitator. After a liquid-liquid dispersion practically started, the values of torque were directly read with a beam of stroboscope to be exactly synchronized to the revolution number of impeller.

**Measurement of Particle Size**—Mixtures of sodium silicate (Kishida Chemical Co.) and distilled water in varying ratios were used as the continuous phase. The total concentration of sodium silicate in the mixtures varied from 17.0 to 88.0% (w/w). The dispersed phase was made up by mixing *n*-C<sub>7</sub>H<sub>16</sub> and CCl<sub>4</sub> in various volume ratios, the density of which was adjusted to be the same as that of the continuous phase (1.000–1.567 g/cm<sup>3</sup>).

In this experiment it was carried out under the following three conditions:

- (1) The viscosity ratios  $\mu_a/\mu_c$  used were at 0.836, 0.271, 0.0992, 0.0280, and 0.00133.
- (2) Revolution numbers were exchanged for 330, 400, and 660 rpm under the constant viscosity ratio.
- (3) Sampling times were selected to logarithmic five points from 5 min to 60 min under the constant revolution number. Every particle size was measured by a microscopic photograph method, and mean surface diameter and mean volume diameter were calculated. Physical properties of each continuous phase were given in Table I.

### Results

#### Relation between Power Number and Reynolds Number

Agitation torque was measured in each viscosity ratio and each revolution number.

Firstly, the relations between the viscosities of continuous phase and agitation torques were shown in Fig. 1. It showed a similar curve on a log-log plots for each revolution number. Although the value of agitation torque increased slowly in range of 1–100 centi poise, it increased remarkably as the viscosity became more higher than 100 centi poise.

Secondly, both values of  $N_p$  and  $N_{Re}$  were calculated from Eq. (3) and Eq. (1), respectively. And  $N_p$  against  $N_{We}$  were plotted on a log-log coordinate as shown in Fig. 2. The resultant plots showed three different correlations in each region. In a laminar region it has a linear correlation having a negative slope. In a critical region concave curve having a minimum value is observed. And in a turbulent region it has an almost constant value. These results

15) Presented at the 95th Annual Meeting of Pharmaceutical Society of Japan, Nishinomiya, April, 1975.

TABLE I. Physical Properties of Each Continuous Phase

Concentration of sodium silicate (% w/w)	Concentration of Tween-20 (% w/w)	Gravity of continuous phase $\rho_c$ (gr/cm <sup>3</sup> )	Specific gravity $\rho_d/\rho_c$ (-)	Viscosity of continuous phase $\mu_c$ (gr/cm <sup>2</sup> ·sec)	Viscosity ratio $\mu_d/\mu_c$ (-)	Interfacial tension $ \gamma_d - \gamma_c $ (dyn/cm)
0.0	0.100	1.000	1.000	1.005	$8.36 \times 10^{-1}$	22.85
17.5		1.290		4.000	$2.71 \times 10^{-1}$	15.50
58.0		1.400		12.20	$9.92 \times 10^{-2}$	15.82
70.5		1.482		47.00	$2.80 \times 10^{-2}$	18.03
88.0		1.567		1100	$1.33 \times 10^{-3}$	21.58

impeller diameter: 49.0 mm, tank diameter: 150 mm  
revolution number: 330, 400, and 660 rpm

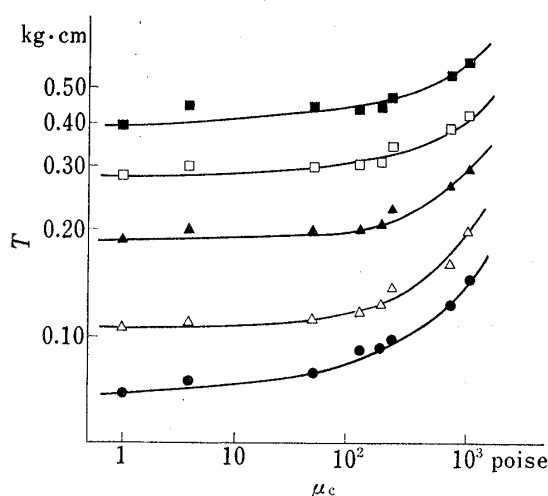


Fig. 1. Plots of Agitation Torque vs. Viscosity of Continuous Phase

$D_I$ : 49.0 mm,  $D_T$ : 150 mm, Tween-20: 0.1% (w/w)  
●: 330 rpm, △: 400 rpm, ▲: 530 rpm,  
□: 660 rpm, ■: 812 rpm

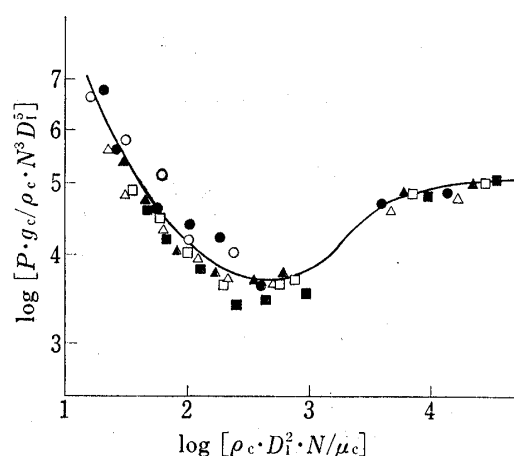


Fig. 2. Plots of Powder Number vs. Reynolds Number

$D_I$ : 49.0 mm,  $D_T$ : 150 mm, Tween-20: 0.1% (w/w)  
○: 200 rpm, ●: 330 rpm, △: 400 rpm,  
▲: 530 rpm, □: 660 rpm, ■: 812 rpm

were excellently agreement with the results which had already investigated by Rushton<sup>13)</sup> and Nagata.<sup>16)</sup> It was, therefore, apparent that the range of  $N_{Re}$  in this experiment expanded over not only a laminar zone but also a turbulent zone on the hydrodynamics.

#### Relation between Weber Number and the Viscosity Ratio $\mu_d/\mu_c$

The relations between specific interfacial area and agitation time for each of four viscosity ratios at 400 rpm were given in Table II. Furthermore, the relations between specific interfacial area and revolution number for each of four viscosity ratios at 60 min were summarized in Table III.

And then, the values of  $N_{we}$  were calculated from Eq. (2). The logarithmic plots of  $N_{we}$  against the ratio  $\mu_d/\mu_c$  for three revolution numbers were shown in Fig. 3. When the ratio  $\mu_d/\mu_c$  became smaller than unity,  $N_{we}$  for each revolution number increased more larger values. On the contrary, as the ratio  $\mu_d/\mu_c$  became near the unity, the change degree of  $N_{we}$  was remarkably increased.

#### Effect of the Viscosity Ratio $\mu_d/\mu_c$ on the Energy Efficiency of Agitation

The changes of the energy efficiency during the agitation from 5 to 60 min at 400 rpm were shown in Fig. 4. In each of four viscosity ratios the values of  $\eta$  decreased straightly as the

16) S. Nagata, K. Yamamoto, H. Yokoyama, and S. Shiga, *Kagaku Kōgaku*, **21**, 708 (1957).

agitation time increased. These negative slopes of straight lines show the same values. As a result described above, it is obviously concluded that the energy efficiency of agitation increases according as the value of viscosity ratio approaches unity.

Subsequently, the relations between  $\eta$  and the ratio  $\mu_a/\mu_c$  at 60 min of agitation time were shown in Fig. 5. The order of energy efficiency  $\eta$  showed in the range of  $10^{-6}$ — $10^{-7}$  orders,

TABLE II. Values of Specific Interfacial Area at Each Experimental Condition

Viscosity ratio $\mu_a/\mu_c$	Agitation time $\theta$			
	5 min	10 min	20 min	35 min
$8.36^{-1}$	919	1090	1282	1653
$2.71^{-1}$	1242	1463	1667	1875
$2.80^{-2}$	995	1200	1405	1579
$1.33^{-3}$	838	972	1143	1415

impeller diameter: 49.0 mm, tank diameter: 150 mm  
revolution number: 400 rpm

unit:  $\text{cm}^2/\text{gr}$

TABLE III. Values of Specific Interfacial Area at Each Experimental Condition

Viscosity ratio $\mu_a/\mu_c$	Revolution number		
	330 rpm	400 rpm	660 rpm
$8.36 \times 10^{-1}$	1333	1690	3010
$2.71 \times 10^{-1}$	1875	2222	3333
$9.92 \times 10^{-2}$	1729	2034	3025
$2.80 \times 10^{-2}$	1542	1852	2500
$1.33 \times 10^{-3}$	1282	1439	2076

impeller diameter: 49.0 mm, tank diameter: 150 mm  
agitation time: 60 min

unit:  $\text{cm}^2/\text{gr}$

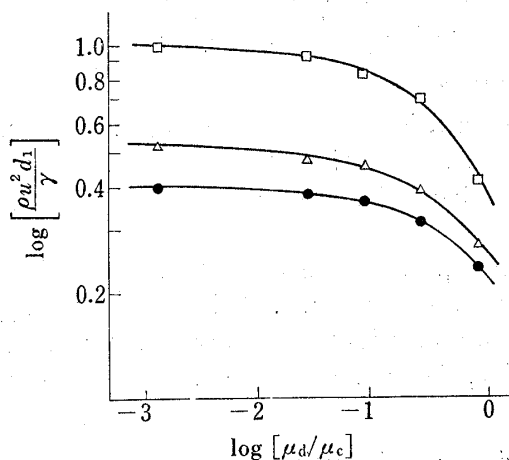


Fig. 3. Relations between Weber Number and Viscosity Ratio

$D_I$ : 49.0 mm,  $D_T$ : 150 mm, Tween-20: 0.1% (w/w)  
 $\theta$ : 60 min  
●: 330 rpm,  $\Delta$ : 400 rpm,  $\square$ : 660 rpm

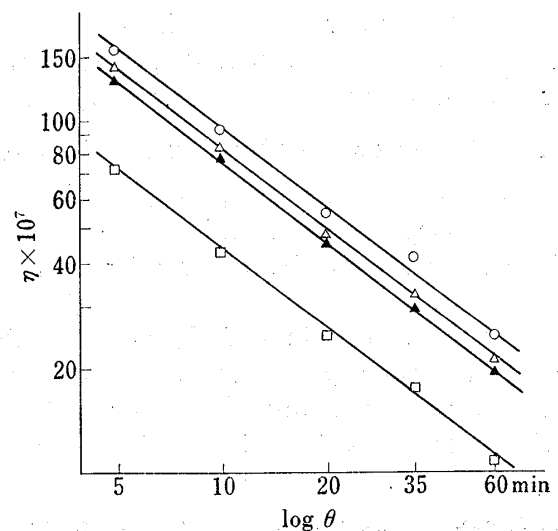


Fig. 4. Changes of Energy Efficiency during Agitation at Four Viscosity Ratios

$D_I$ : 49.0 mm,  $D_T$ : 150 mm, Tween-20: 0.1% (w/w)  
 $N'$ : 400 rpm  
 $\circ$ :  $\mu_a/\mu_c=0.836$ ,  $\Delta$ :  $\mu_a/\mu_c=0.271$ ,  
 $\blacktriangle$ :  $\mu_a/\mu_c=0.0280$ ,  $\square$ :  $\mu_a/\mu_c=0.00133$

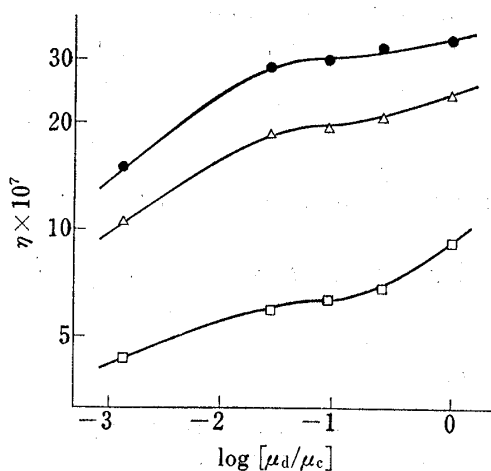


Fig. 5. Relations between Energy Efficiency and Viscosity Ratio

$D_1$ : 49.0 mm,  $D_T$ : 150 mm, Tween-20: 0.1% (w/w)  
 $\theta$ : 60 min  
 ●: 330 rpm,  $\Delta$ : 400 rpm,  $\square$ : 660 rpm

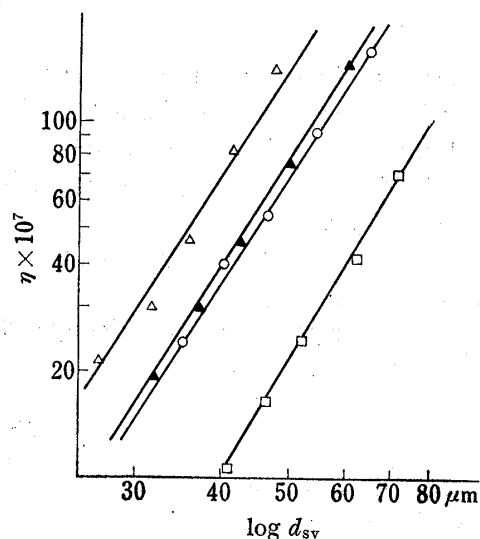


Fig. 6. Relations between Energy Efficiency and Mean Length Diameter

$D_1$ : 49.0 mm,  $D_T$ : 150 mm, Tween-20: 0.1% (w/w)  $N'$ : 400 rpm  
 $\circ$ :  $\mu_d/\mu_c=0.836$ ,  $\Delta$ :  $\mu_d/\mu_c=0.271$ ,  
 $\blacktriangle$ :  $\mu_d/\mu_c=0.0280$ ,  $\square$ :  $\mu_d/\mu_c=0.00133$

and  $\eta$  has a lower value as the ratio  $\mu_d/\mu_c$  decreased. Especially, the energy efficiency became lower distinctly for the ratio  $\mu_d/\mu_c$  of  $10^{-2}$ — $10^{-3}$  orders.

Finally, the relations between  $\eta$  and  $d_{sv}$  were shown in Fig. 6. The order of energy efficiency  $\eta$  had the range of  $10^{-5}$ — $10^{-6}$ . From the results of Fig. 6, it is apparently clear that the energy efficiency of agitation gradually decreased as the efficiency for the average diameter of droplets became smaller.

## Discussion

At first, the effects of the ratio  $\mu_d/\mu_c$  on the agitation power were discussed. When the viscosity of continuous phase was over 100 centi poise, the value of agitation torque had a large value (Fig. 1) and  $N_{Re}$  indicated a value of laminar region (Fig. 2). Therefore, the fluid circulation was worse in comparison with the great consumption of agitation of agitation power and it did not occurred easily to break a drop.

Secondly, on the effects of the ratio  $\mu_d/\mu_c$  for  $N_{we}$ , although  $N_{we}$  had considerably a small value at near the unity of the ratio  $\mu_d/\mu_c$ , it increased according as the ratio  $\mu_d/\mu_c$  became smaller. From these results, it would be apparent that a droplet was hardly broken up as the ratio  $\mu_d/\mu_c$  decreased.

On the other hand, the effect of the ratio  $\mu_d/\mu_c$  on energy efficiency  $\eta$  was observed as follows. Energy efficiency of agitation decreased abruptly as the agitation time increased.

It would seem that the effect of agitation was little even if the agitation was continuously done for a long intervals. The optimum condition concerned with the agitation time may be existed in compliance with the operation for the formation of emulsion.

Finally, on the relations between  $d_{sv}$  and energy efficiency  $\eta$ , it was found out that the value of  $\eta$  decreased as the average diameter became smaller. This trend would be considered as follows. In the case of large size drops, the breakup of them may be possible only by a weak shear force. While, in small size drops, the breakup of them may not be possible without a strong shear force. So, the probability for the burst of small size drops would be a little.

### Conclusions

The results of the experimental work can be summarized as follows.

- (1) In the range of low viscosity, the values of agitation torque increased slowly. But, it showed remarkable increase as the viscosity became higher than 100 centi poise.
- (2)  $N_{we}$  for each revolution number increased to a large value as the ratio  $\mu_d/\mu_c$  became smaller than unity.
- (3) The values of energy efficiency  $\eta$  against the agitation time decreased straightly on a log-log plots.
- (4) The energy efficiency  $\eta$  decreased gradually according as the values of  $d_{sv}$  became smaller and smaller.

### Nomenclature

$D_I$ : impeller diameter (mm)	$S$ : specific interfacial area (l/cm)
$d_s$ : mean surface diameter ( $\mu$ )	$T$ : agitation torque (kgw·cm)
$d_{sv}$ : mean surface-volume diameter ( $\mu$ )	$u$ : impeller velocity (cm/sec)
$d_v$ : mean volume diameter ( $\mu$ )	$\gamma$ : interfacial tension (dyn/cm)
$g_c$ : gravitational conversion factor (kg·m/kgw·sec <sup>2</sup> )	$\eta$ : energy efficiency of agitation (—)
$E_A$ : agitation energy (erg/cm <sup>3</sup> )	$\theta$ : agitation time (min)
$E_s$ : specific interfacial energy (erg/cm <sup>3</sup> )	$\mu_c$ : viscosity of continuous phase (poise)
$k'$ : constant, Eq. (6)	$\mu_d$ : viscosity of dispersed phase (poise)
$N$ : revolution number (rps)	$\mu_d/\mu_c$ : viscosity ratio (—)
$N_p$ : powder number (—)	$\rho_c$ : specific gravity of continuous phase (gr/cm <sup>3</sup> )
$N_{Re}$ : Reynolds number (—)	$\rho_d$ : specific gravity of dispersed phase (gr/cm <sup>3</sup> )
$N_{we}$ : Weber number (—)	$\rho_d/\rho_c$ : specific gravity ratio (—)
$P$ : agitation powder (watt)	$\omega$ : angular velocity (angle/sec)