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Comparison of dispersion behavior of agglomerated particles in liquid between ultrasonic irradiation and mechanical stirring



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ABSTRACT

The particle dispersion behavior was compared for ultrasonic irradiation and mechanical stirring. The experiment and calculation were carried out with polymethylmethacrylate (PMMA) particles. The dispersion rate of the agglomerated particles increased with the decreasing ultrasonic frequency and the increasing electric power, whereas it increased with the increasing rotation speed for the mechanical stirring. The temporal change in the particle dispersion proceeded stably after passage of a long time. The dispersion of the ultrasonic irradiation was suggested to occur by the erosion from the surface of the cluster one by one due to the bulk cavitation as well as the division into smaller particles because of the inner cavitation, and that of the mechanical stirring mainly by the division into smaller clusters due to the shear stress flow. Based on the experimental results, mathematical models for the ultrasonic irradiation and mechanical stirring were developed with the dispersion and agglomeration terms and the calculation of the temporal change in the total cluster number at the different operational factors agreed with the experiments. The dispersion efficiency of the ultrasonic irradiation was larger than that of the mechanical stirring at the lower input power, but it was reversed at the higher input power.

1. Introduction

Well-dispersed fine particles added in products [1] exhibit better characteristics and they are applied to the cutting-edge fields such as composite materials, cosmetic products, and pharmacy [2]. The suspended particles in the preparation of raw materials, crushing, mixing and forming of materials manufacturing processes often exist in the agglomerated state. Therefore, it is important to understand the particle dispersion methodologies.

There are several techniques to disperse the particles homogeneously. For example, the particle surface coating [3] and a high shear stress flow [4,5] in a mechanical mixing process were used for the homogeneous dispersion. Moreover, an ultrasonic irradiation is well known to disperse particles efficiently [6–8] due to a cavitation effect. The cavitation behavior during the ultrasonic irradiation is to repeat the formation, growth and collapse of bubbles, to lead to local hot spots [9] with high pressure and temperature and to result in a local cavitation force due to collapsing of the tiny bubbles. The cavitation-induced force gives rise to many chemical and physical effects [10,11] such as chemical reactions, erosion [12], dispersion [13,14] etc. There are reports on the effects of ultrasonic frequency and liquid height [15], irradiation mode [16], sound amplitude [17] and temperature [18] on the particle dispersion in fluid during the ultrasonic irradiation. On the other hand, a mathematical model [19] on the agglomerated particles dispersion by mechanical stirring was applied to the ultrasonic irradiation process [20]. However, it was not known about the dispersion behavior of each cluster of *i*-particles due to the overall first-order reaction model. No comparison of the particle dispersion behavior was carried out between the ultrasonic irradiation and mechanical stirring.

In this study, the experiments on the dispersion behavior of the agglomerated particles were made for the ultrasonic irradiation and mechanical stirring, and mathematical dispersion models were developed with a population balance model which gave us the information of the temporal change in each cluster of *i*-particles. As the temporal change in the clusters number proceeded stably after passage of a long time, an agglomeration term was introduced in addition to a dispersion one for the mathematical model. After the confirmation of a good agreement between the experimental and calculated results, the dispersion and agglomeration properties were discussed about the ultrasonic irradiation and the mechanical stirring.

2. Experimental setup

2.1. Method

The schematic diagram of experimental apparatuses for (a)

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Fig. 1. Experimental apparatuses of ultrasonic irradiation and mechanical stirring.

ultrasonic irradiation and (b) mechanical stirring is shown in Fig. 1. For the ultrasonic irradiation (Branson, Series 8500), a cuboid vessel was filled with tap water without degassing and the bath depth was fixed to 210 mm to obtain the best cavitation activity. Dissolved oxygen concentration of the tap water measured by DO meter (WTW, Multi3410) was 8.8 mg/L at 293 K. The frequency, *f*, was changed to 40, 80 and 120 kHz, and the input power, P_{set} was varied to 150, 300 and 500 W in the ultrasonic bath. On the other hand, for the mechanical stirring, the rotation speed, R_{s} , changed to 200, 300, 500 and 700 rpm with an impeller (diameter, *d*: 50 mm, thickness, *b*: 14 mm).

Polymethylmethacrylate (PMMA) particles of 2.8×10^{-6} m in mean diameter and 3.0×10^{3} mol/m³ KCl solution were used as solid and liquid phases, respectively. These physical properties are shown in Table 1. The KCl solution was used to obtain the rapid agglomeration condition of the particles by making thinner electric double layer around the particles and causing the negligible repulsion between particles. 0.05 g of PMMA particles were put into 100 mL (inner diameter, *D*; 60 mm, bath depth, *h*; 35 mm) of KCl solution for each experimental condition. The dissolved oxygen concentration of KCl solution became 9.0 mg/L at 294 K before the experiment.

The sample-contained beaker arrangement at the ultrasonic irradiation bath is shown in Fig. 2. After changing the immersion depth of the beaker as shown in Section 3.2, we fixed the position to 5 mm in depth, whereas the bath depth of KCl solution in the beaker was always kept to 35 mm.

To adjust the initial condition before the experiment, the agglomerated particles in the KCl solution were prepared as follows. Irradiated with ultrasonic waves at 400 kHz and 500 W for 10 min, particles were firstly suspended in 10 mL of ion exchanged water. Next, by adding it into 90 mL of KCl solution, 100 mL of $3.0 \times 10^3 \text{ mol/m}^3$ KCl solution was prepared, and the particles were agglomerated by the mechanical stirring at 300 rpm for 10 min. After the preparation, the agglomerated particles concentration was about $2.1 \times 10^{12} \text{ m}^{-3}$. During each experiment, samplings were made at a given interval and temporal changes in the cluster number of particles and the size distribution were measured by a particle size distribution analyzer (Beckman Coulter, Inc., Multisizer 3). The cluster size of the particles measured here was a volume-equivalent diameter.

Table 1

Physical properties of particle and liquid used for the experiment.

PMMA particle	Mean diameter [m] Density, ρ _P [kg/m ³] Hamaker constant, A ₁₃₁ [J]	$\begin{array}{c} 2.8 \times 10^{-6} \\ 1200 \\ 1.05 \times 10^{-20} \end{array}$
Liquid	KCl concentration, C [mol/m ³] Density, ρ _L [kg/m ³] Viscosity, μ [Pa·s]	3000 1130 9.27x10 ⁻⁴



Fig. 2. Sample-contained beaker arrangement at the ultrasonic irradiation bath.

2.2. Measurement of ultrasonic power by calorimetry method

The change in heat energy of the solution caused by the ultrasonic irradiation was measured by a calorimetry method [21]. The input power, $P_{\rm ult}$ [W], into the solution was calculated from Eq. (1).

$$P_{\rm ult} = \left(\frac{\mathrm{d}T}{\mathrm{d}t}\right) C_{\rm P} m \tag{1}$$

where *T* is the solution temperature [K], C_p is the heat capacity of water [=4.2 J/g] and *m* is the mass of the solution [g]. The $\frac{dT}{dt}$ value was obtained from 5 temperature plots for 20 min. The measured P_{ult} values for each set power, P_{set} [W], are shown in Table 2.

2.3. Calculation of energy input rate of mixing energy into liquid bath

The supplied mixing energy rate, $P_{mec}[W]$, into liquid bath was calculated by the following equations [22].

$$P_{\rm mec} = N_{\rm p} \rho_{\rm L} n_{\rm s}^3 d^5 \tag{2}$$

Table 2	
Electric power supplied into	solution.

f [kHz]	P_{set} [W]	$P_{\rm ult}$ [W]	Power [W/kg]
40	150	5.88	52.5
40	300	9.53	85.1
40	500	13.73	122.6
80	300	6.85	61.2
120	300	4.75	42.4

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