

Liquid-Solid Separation of Photo-Catalyst Suspension Induced by Ultrasound

Yasuyuki Suzuki[†], Warsito, Akinori Maezawa, and Shigeo Uchida*

Department of Materials Science and Chemical Engineering, Shizuoka University, Hamamatsu 432-8561

[†]Department of Environmental Science, Graduate School of Science and Engineering, Shizuoka University, Hamamatsu 432-8561

(Received October 6, 1999; CL-990853)

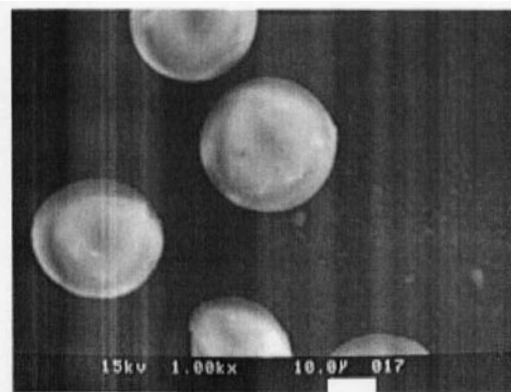
A method for the induction of the agglomeration of different kinds of particles by the ultrasonic irradiation in the water was studied. This method has been proven to be applicable to the separation of photo-catalyst particles from the drainage of photo-oxidation process. The catalyst particle was titanium oxide with the average diameter of 3 μm . The agglomeration was observed when the ultrasound with the frequency of 26 kHz and the power of 80 W was irradiated and a certain amount of glass beads with the diameter between 33-44 μm were added. The catalyst particles deposited on the glass beads, and then settled together with the glass beads leading to the liquid-solid separation. It was also found that the addition of the glass beads has also enhanced the agglomeration among the catalyst particles themselves.

There are two ways to make liquid-solid into contact in photo-oxidation systems, i.e. by suspension¹ and immobilization.² The major advantage of the former method is to give the large liquid-solid contact area. However, it is difficult to recover the micro size catalyst particles from the treated water. The separation of micro size particles in suspension has been a difficult problem for a long time. In this regards, the latter method has been considered to have a major advantage of the ease of the catalyst particles recovery. However, the main problem in the method is the relatively small liquid-solid contact area. In this paper, a separation method of micro size photo-catalyst particles in suspension induced by ultrasound is proposed.

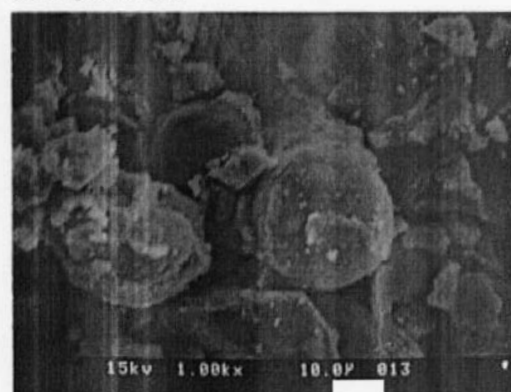
A sample of the suspension of titanium dioxide particles (P25, Nippon Aerosil Co., Japan) was prepared. Glass beads (hereinafter referred as GB, Toshiba Glass Corp., Japan) with the diameter of 33-44 μm were then added to the sample as collectors of the micro size catalyst particles. The details of the physical properties of the particles and the experimental conditions are given in Table 1. The experiment was conducted in a cylindrical vessel made of stainless steel (110 mm ID, 180 mm height). A beaker with the size of 500 mL filled with the sample was placed inside the vessel, which was filled with water. The ultrasonic transducer was equipped at the bottom of the vessel. The ultrasound propagated through the water jacket to

Table 1. Details of samples and experimental conditions.

Properties of particles		D_p (μm)	Surface area (m^2/g)	Apparent Gravity (g/mL)	True Gravity (g/mL)
	P25	3	500	0.25	3.6
	GB	33-44	57	2.6	2.6
Sample	0.05 g/500 mL(P25) + GB				
Ultrasound	Frequency : 26 kHz, Power : 80 W				



(A) GB particles only



(B) GB particles with P25

Figure 1. SEM images of the surface of GB after US irradiation. \longleftrightarrow : 10.0 μm

the sample in the beaker. After 60 min of irradiation, the sedimentation phenomenon was observed by naked eyes and SEM imaging, and the particle size of the sediment was measured by a Laser refraction-scattering method (LA-910, Horiba Corp., Japan).

The sedimentation of the catalyst particles was not observed without glass beads or without ultrasonic irradiation, i.e. no agglomeration occurred. However, when glass beads with the weight ratio of 4:1 to the catalyst particles were added and the ultrasound with the frequency of 26 kHz and the power of 80 W was irradiated, the sedimentation of the catalyst particles together with the glass beads was observed, indicating that agglomeration of the catalyst particles on the glass beads occurred. The agglomerated particles had a relatively fast settling velocity and settled completely in a few minutes. However, there were also agglomerates with the settling velocity which was relatively low but still much higher than that of catalyst particles without ultrasonic irradiation. It indicates that

the agglomeration among the catalyst particles themselves has also occurred. It required about 8 h for all the agglomerates to completely settle until a clear separation line between the liquid and sediment phases was observed.

The SEM photographs of the sediment of the samples after ultrasonic irradiation are shown in Figure 1. It is seen from Figure 1(A) (glass beads only) that the surface erosion of the glass beads was not observed even under the irradiation. From the SEM photograph of the sediment of the mixed particles after the irradiation (1(B)), the catalyst particle deposition on the glass beads was observed, i.e. the glass beads became collectors of the catalyst particles under the irradiation.

Figure 2 shows the particle size distribution of the agglomerates measured after the irradiation in comparison with the distributions for the glass beads and the catalyst particles before the irradiation. The broken line is the distribution for the catalyst particles, while the dashed one is that for the glass beads. The peaks A and B show the mean diameters of the catalyst and GB particles, respectively. After the irradiation, it is clearly seen that the size distribution of the agglomerate, it is shown with the solid line, shifted to the right (larger particle size region). The peak C is considered to be the distribution for the agglomerates among the catalyst particles themselves, while the peak D is for the agglomerates of the glass beads and the catalyst particles. The agglomeration phenomena are described below.

The agglomeration of micro particles in the suspension may be caused by the particles collision induced by the ultrasound. The collision occurs when particles jump from the streamline of the liquid, and collide with the others. The particle-jump may occur when the particle with the large inertia force can not follow the liquid streamline.^{3,4} Light and small particles will follow the liquid motion (vibration) caused by the sound pressure, and thus the particle-jump and the collision with the other particles may scarcely occur.

For sufficiently large and/or heavy particles, they are difficult to follow the curved streamline, and will jump from the streamline and may collide with the other particles. However, very large or very heavy particles may not be affected by the liquid motion, and remain stationary. Accordingly, the collision between the particles may not occur.

In the acoustic field, particles may also vibrate. Thus the particle-jump from the liquid streamline is determined by the difference between the vibrating amplitude of the particle and

that of the liquid. For light and small particles, the amplitude difference is small, and the particle vibration synchronizes with the liquid one. Accordingly, the particle-jump and thus the inter-particles collision may not occur. As for very large and/or very heavy particles, the particle vibration, and thus the collision may not occur. Hence, there should be an optimum condition for the collision determined by the particle size, the liquid properties and the sound frequency. The optimum condition can be obtained using the equation for the amplitude ratio proposed by Brandt and Hiedemann (1936) as follows.^{5,6}

$$\frac{\xi_p}{\xi_m} = \frac{1}{\sqrt{1 + \left(\frac{\pi \rho f d^2}{9 \mu}\right)^2}} = \cos \phi \quad (1)$$

where ξ_m and ξ_p are the amplitudes of the liquid and the solid particle, respectively. ρ is the density of the particle, f is the frequency of the ultrasound, d is the particle radius, and μ is the liquid viscosity. The amplitude ratio can also be expressed in a term of the phase difference between the liquid and solid particle, denoted by ϕ . From this equation, for large ϕ (large liquid viscosity, low density or small solid particle size), the solid particle will move together with the liquid motion, and thus the probability of collision is low. For small ϕ (small liquid viscosity, high density or large solid particle size), the effect of the sound pressure on the particles motion is small, and thus the probability of collision is also low. The optimum value of ϕ for the agglomeration is in the range of $0.5 \leq \cos \phi \leq 0.7$.

However, in our experiment, the value of $\cos \phi$ is nearly unity, which is beyond the above range for the agglomeration. This was also confirmed by our result that the agglomeration was not observed when no glass beads were added. By adding the glass beads, an amplitude difference of the vibration for the glass beads and that for the catalyst particles may exist as they have different inertia (different sizes and densities), and thus the collision between the two-particles is expected.

The agglomeration of particles is caused by the adhesive force. Since the adhesive force between P25 and GB is smaller than that of P25 particles themselves, some parts of P25 agglomerate is considered to be the agglomerate of P25 peeled off from the surface of the GB.

In order to confirm the photo-catalytic activity of the agglomerate obtained, an experiment was performed on the decomposition of the surfactant, polyoxyethylenealkyl(14)ether, $C_{14}H_{29}O(CH_2CH_2)_7H$, under the same conditions reported as elsewhere.⁷ The decomposition of the surfactant was completed in nearly the same time required for fresh P25 particles, showing that the activity was still remained.

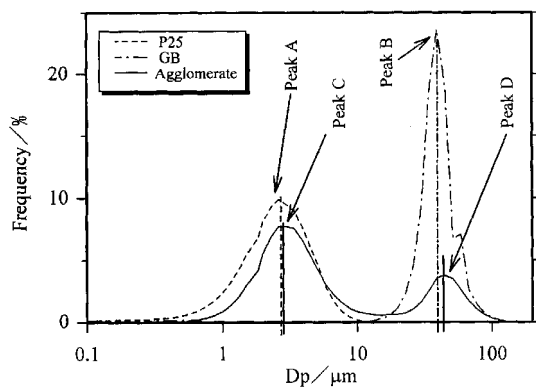


Figure 2. Particle size distributions of P25, GB and agglomerate.

References and Notes

- 1 E. Pramauro, A. B. Prevot, M. Vincent, and G. Brizzolesi, *Environ. Sci. Technol.*, **31**, 3126 (1997)
- 2 Y. Zhang, J. C. Crittenden, D. W. Hand, and D. L. Perram, *Environ. Sci. Technol.*, **28**, 425 (1994)
- 3 Warsito, S. Uchida, A. Maezawa, and S. Okamura, *J. Chem. Eng. of Jpn.*, **30**, 786 (1997)
- 4 Warsito, M. Ohkawa, A. Maezawa, and S. Uchida, *Chem. Eng. Sci.*, **52**, 3941 (1997)
- 5 O. Brandt and E. Hiedemann, *Trans. Faraday Soc.*, **32**, 1101 (1936)
- 6 O. Brandt and E. Hiedemann, *Zeitschrift für Physik*, **104**, 511 (1937)
- 7 Y. Suzuki, H. Arakawa, Warsito, A. Maezawa, and S. Uchida, *Int'l J. of PhotoEnergy*, (in press) (1999).