[Chem. Pharm. Bull. 33(9)4054—4056(1985)]

Measurements of the Adhesive Force between Particles of Powdered Organic Substances and a Glass Substrate by Means of the Impact Separation Method. II. Effect of Addition of Light Anhydrous Silicic Acid on the Adhesive Force of Potato Starch[1,2)]

AKINOBU OTSUKA,* KOTARO IIDA, KAZUMI DANJO, and HISAKAZU SUNADA

Faculty of Pharmacy, Meijo University, Yagoto-Urayama, Tempaku-ku, Nagoya 468, Japan

(Received January 30, 1985)

The effect of addition of light anhydrous silicic acid on the adhesive force between potato starch particles and a glass plate was investigated by the impact separation method. It was found that the adhesive force was reduced to approximately 1/1000 by adding light anhydrous silicic acid at concentrations of more than 0.05%. The theoretical attractive force was evaluated on the basis of the van der Waals force between a sphere and a half-space.

Keywords——adehesive force; impact separation method; pendulum-type shock testing machine; powdered organic substance; light anhydrous silicic acid; potato starch; glidant

Numerous studies have been reported on the effect of glidants on the adhesive and cohesive properties of powder layers or on the flowability of powders. However, few studies have been done on direct measurement of the adhesive force between a single particle and a substrate in the presence of a glidant. In the present study, the adhesive force between potato starch particles and a glass plate was determined with and without light anhydrous silicic acid (LASA) by the impact separation method.

Experimental

Materials——Sample powders were prepared by sieving potato starch (JP X) with a 200 mesh screen to obtain particles having a mean particle size of 42 μm (Feret's diameter). Two kinds of LASA of commercial origin, Adsolider 101 (Freund Ind. Co., Ltd.) and Aerosil 200 (Nippon Aerosil) were used as the additives. A microscope slide glass was used as the substrate after being rinsed with methanol and acetone and air-dried.

Method——A definite amount of LASA was added to potato starch and mixed (Vortex-Genie model K-550-G, Scientific Industries Inc.). The number of LASA particles adhering to 100 μm^2 of the surface of potato starch was counted by the use of a scanning electron microscope (JEOL JSM-T20, JEOL Ltd.).

The adhesive force between starch particles and a glass plate was determined by the impact separation method.[1)] Sample particles were placed on the glass plate in a measuring cell, which was fixed to the impact hammer of a pendulum shock testing machine (PST-300, Yoshida Seiki Co., Ltd.). The hammer was motor-driven to a desired height and then allowed to fall to impact a shock absorbing mat. The impact acceleration generated was measured with an accelerometer.

The percentage of particles adhering on the substrate was determined by counting the numbers of particles before and after separation. A plot of separation force *vs.* percentage of particles remaining on a logarithmic probability paper yields the average adhesive force f_{50} at which 50% of particles still remain on the substrate after separation. The experiments were carried out at room temperature and at a relative humidity of $60 \pm 10\%$.

Results

Concentration of LASA and Adhesiveness

Figure 1 shows the relationship between the percentage of LASA mixed and f_{50}. The

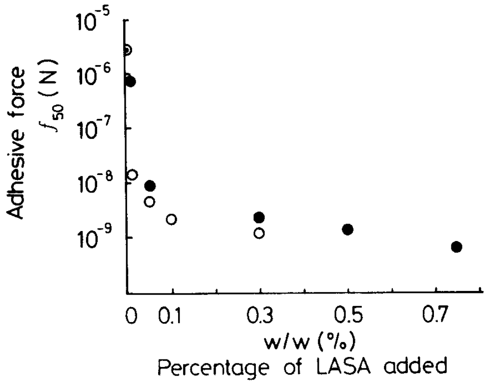

Fig. 1. Relationship between f_{50} and Percentage of LASA Added

○, Aerosil 200; ●, Adsolider 101; ⊙, without LASA.

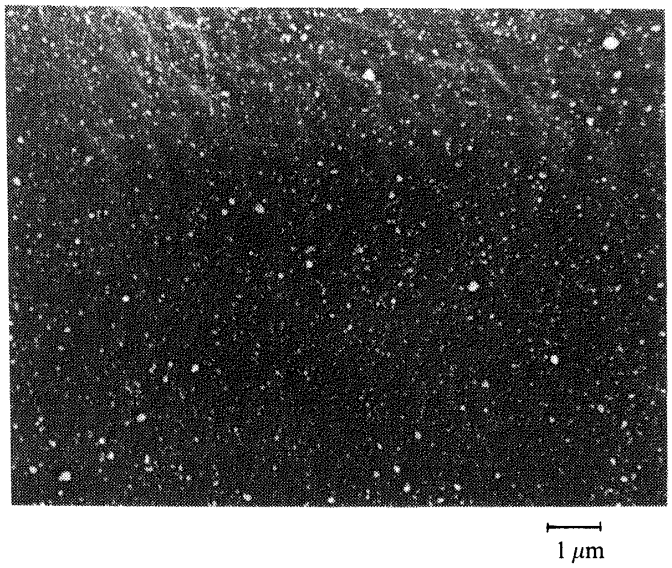

Fig. 2. Scanning Electron Micrograph of LASA on the Surface of a Starch Particle (Concentration of LASA; 0.05%)

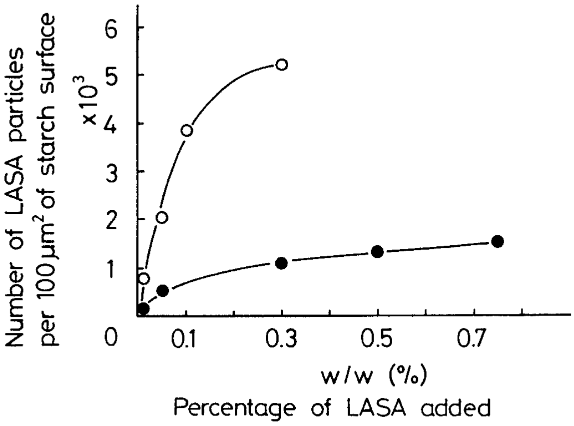

Fig. 3. Relationship between Number of LASA Particles per 100 μm² of Starch Surface and Percentage of LASA Added

○, Aerosil 200; ●, Adsolider 101.

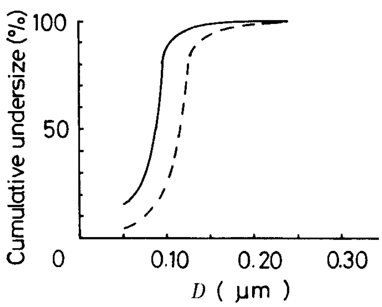

Fig. 4. Apparent Cumulative Particle Size Distribution Curves of LASA Aggregates on the Surface of Starch Particles

——, Aerosil 200; ----, Adsolider 101.

adhesive force rapidly decreased in the concentration range up to *ca.* 0.05% and fell gradually thereafter. Hayashi *et al.*[3,4] have reported that tensile strength or flowability of a potato starch powder layer changed remarkably with the addition of LASA in the same concentration range.

Number of Particles of LASA Adhering to the Starch Surface

Figure 2 shows a scanning electron microscopic photograph of a potato starch surface to which LASA particles are attached (concentration of LASA; 0.05%). Since it is known that the primary particle size of LASA used is *ca.* 10 nm,[3] the particles observed under SEM should be aggregates of the original particles. Figure 3 shows the relationship between concentration of LASA and the number of LASA aggregates adhering to 100 μm² of the surface of a starch particle. The adhesiveness of Aerosil 200 was found to be higher than that of Adsolider 101 on the basis of the SEM photographs. Size distributions of the aggregates on starch particles are shown in Fig. 4. The apparent mean diameters of Aerosil 200 and

Adsolider 101 aggregates obtained from these curves are 0.08 and 0.13 μm respectively.

Discussion

It is well known that addition of a small amount of LASA fine powder to highly adhesive powders may improve their flowability. A decrease in adhesiveness between particles or between particles and the wall of the device caused by the addition of LASA may play an important role in the improvement of flowability. In what follows, some theoretical evaluations of adhesive force, where only van der Waals force is considered, have been carried out. Van der Waals force F_{vdw} between a sphere having a radius of R and a half-space is represented by the following equation[5,6]:

$$F_{vdw}=\frac{\hbar\omega}{8\pi z_0^2}R \quad (1)$$

where $\hbar\omega$ represents the Lifshitz–van der Waals constant and z_0 is the adhesional distance between a particle and a half-space. In the absence of LASA, $\hbar\omega$ is estimated to be 8×10^{-19} J by substituting 21 μm for R, 0.5 nm for z_0[5] and 2.7×10^{-6} N for F_{vdw}, which is the measured value of the adhesive force. The value of $\hbar\omega$ is generally considered to be in the range of 1—15×10^{-19} J,[6] so that the obtained value may be reasonable. In the presence of LASA, the most probable contact situation is that between LASA aggregates and the substrate. If we assume that the actual separation takes place between a LASA aggregate and the glass plate, F_{vdw} can be estimated from Eq. 1 to be *ca.* 2.4×10^{-9} N, where the $\hbar\omega$ value of 3×10^{-19} J[5] and the R value of 0.05 μm (approximate value of radius of LASA aggregates) were used. On the other hand, when the separation occurs between a potato starch particle and a LASA aggregate, F_{vdw} can be obtained from Eq. 2, substituting the harmonic average radius for potato starch (21 μm) and LASA aggregate (0.05 μm) for R.

$$F_{vdw}=\frac{\hbar\omega}{16\pi z_0^2}R \quad (2)$$

If we use 8×10^{-19} J, which is the measured value for glass/potato starch, for $\hbar\omega$, we obtain the F_{vdw} value of 6.4×10^{-9} N. These calculated values are roughly in agreement with the adhesive force measured at concentrations of LASA of more than 0.05%.

References and Notes

1) A. Otsuka, K. Iida, K. Danjo, and H. Sunada, *Chem. Pharm. Bull.*, **31**, 4483 (1983).
2) This work was presented at the 104th Annual Meeting of the Phamaceutical Society of Japan, Sendai, March 1984.
3) H. Hayashi, I. Kawakage, and K. Takatori, *Yakuzaigaku*, **37**, 140 (1977).
4) H. Hayashi, T. Kasano, and K. Suhara, *Yakuzaigaku*, **39**, 187 (1979).
5) L. Massimilla and G. Donsi, *Powder Technol.*, **15**, 253 (1976).
6) H. Krupp, *Adv. Colloid Interface Sci.*, **1**, 111 (1967).