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183. Nobuyoshi Kaneniwa, Akiko Ikekawa, and Haruko Aoki[*1]: Influence of Particle Size on Physicochemical Properties of Pharmaceutical Powders. I. On Fluidity of Sodium Borate and Boric Acid Powders. (1).

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Angles of repose and sliding angles of sodium borate and boric acid powders are investigated by four methods.

These angles gradually increase with decrease of particle size by any method, and critical diameters are found to be 160 μ for sodium borate and 110 μ for boric acid powders.

The relation between angle (α) and particle diameter (D) is represented as follows.

$$D \leq D_c \qquad \tan\alpha = A_1 \exp(-B_1 D)$$
$$D \geq D_c \qquad \tan\alpha = A_1' \exp(-B_1' D)$$
$$A_1 > A_1' \qquad B_1 > B_1' > 0$$

It is concluded that the cohesive force between particles of powders is negligibly small in the region of particle diameter larger than the critical size, and the cohesive force remarkably influences the angle at smaller diameters.

The following relation is found between C/Mg and D, in the case of sodium borate powders.

$$C/Mg = 0.81 \exp(-147\,D)$$

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"Particle size" is an important parameter of pharmaceutical powders which must be considered in order to control the effect and its duration ingested medicines and injected antibiotics.[1] Particle size has further important effects on tablet compression. The present study has been initiated to elucidate some problems of particle size in pharmaceutics.

Uematsu, *et al.*[2] indicated that powders in a wide sense consisted of powders composed of cohesive particles and granules composed of particles whose cohesive force was negligibly small. Further the presence of critical size, which seemed to differentiate powders from granules, has been reported in the general studies on the relation between particle size and fluidity of powders. From studies on angles of repose of sulfathiazole

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[1] J.H. Fincher, J.G. Adams, H.M. Beal: J. Pharm. Sci., **54**, 704 (1965); R.M. Atkinson, C. Bedford, K.J. Child, E.G. Tomich: Nature, **193**, 588 (1962). K. Kakemi, T. Arita, S. Ohashi: Yakugaku Zasshi, **82**, 1468 (1962); M. Aoki, T. Hukuda: *Ibid.*, **75**, 878 (1955).

[2] T. Uematsu: Nippon Kikaigaku Kaishi, **56**, No. 408, 53 (1953); T. Uematsu, *et al.*: Kikai Gakukai Ronbunshu, **17**, No. 56, 72 (1951); T. Kojo: Kagaku Kogaku, **18**, 205 (1954).

granules, Nelson[3] found that the critical diameter was 500~600 μ, for a minimum angle of repose. Aoki and others[4] also indicated the same phenomena for static frictional coefficients of powders. Furthermore Smalley[5] studied the relation between particle size and flow rate of sifted sands and found the flow-stick transition diameter at 200 μ, where flow rate showed maximum values.

In this paper we report on the fluidity of sodium borate and boric acid powders.

Experimental

Sodium borate and boric acid powders (J.P. grade) were sieved with sieves standardized by JIS. Sodium borate powders were kept in a desiccator which maintained 75% relative humidity with a saturated solution of sodium chloride, and boric acid powders were dried in a desiccator containing anhydrous calcium chloride. The surface mean diameter of the powders passing through a 200 mesh sieve was determined by microscopic examination. The arithmetic mean values of the scales of the sieves were regarded as the particle diameters of the larger powders.

Angles of repose were measured by Nogami-Sugiwara's,[6] Nelson's[7] and Neumann's[8] methods and sliding angles by Awata-Nakajima's method.[9] In order to minimize wall effect, measurement of angles by Nogami-Sugiwara's method were made with a plate 3 cm. × 10 cm.

Results and Discussion

The experimental results are shown in Fig. 1 and 2. By all four methods of measurement, the angle of repose and sliding angle increase gradually with the decrease of particle diameter, and the rate of increase of these angles is much larger at the

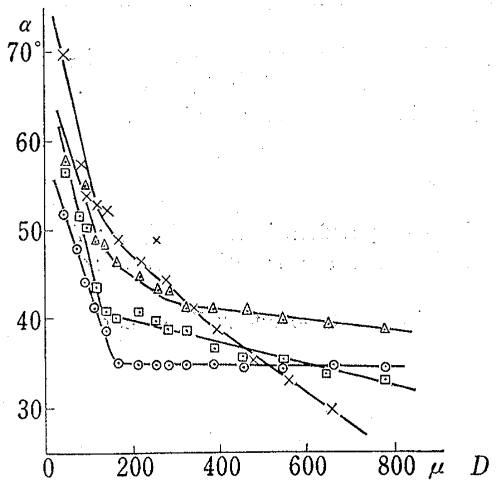

Fig. 1. Dependence of Particle Size on Angle of Repose or Sliding Angle of Sodium Borate Powders

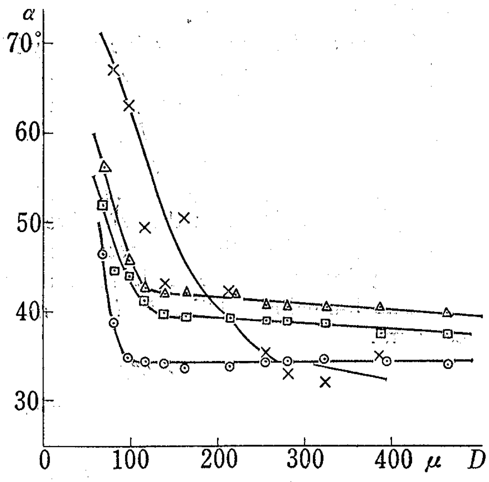

Fig. 2. Dependence of Particle Size on Angle of Repose or Sliding Angle of Boric Acid Powders

By four methods.

⊙ Nogami-Sugiwara

⊡ Neumann

△ Nelson

× Awata-Nakajima

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[3] E. Nelson, *et al.*: J. Am. Pharm. Assoc., **44**, 435 (1955).

[4] M. Aoki, T. Hukuda: Yakugaku Zasshi, **76**, 140 (1956). T. Tanaka, S. Kawai: Kagaku Kogaku, **20**, 114 (1956). N. Kaneniwa: Kanazawa Daigaku Yakugakubu Kenkyu Nempo, **13**, 75 (1963).

[5] I.J. Smalley: Nature, **201**, 173 (1964).

[6] H. Nogami, M. Sugiwara, S. Kimura: Yakuzaigaku, **25**, 260 (1965).

[7] E. Nelson: J. Am. Pharm. Assoc. Sci. Ed., **44**, 435 (1955).

[8] J.J. Hermans: "Flow Properties of Disperse Systems," 406 (1955). North Holland Publishing Co.

[9] E. Awata, E. Nakajima, T. Morioka, Y. Ikegami, M. Yoshizumi: Yakugaku Zasshi, **80**, 1657 (1961).

region of smaller diameter. The critical diameter was found to be 160 μ for the rate of increase of angles by Nogami-Sugiwara's method in sodium borate powders.

Since the tangent of the angle of repose or sliding angle ($\tan \alpha$) is directly related to the apparent frictional coefficient of powders, these tangents have important physical meanings.

In the present study, the influence of particle size on $\tan \alpha$ was investigated, and the existence of critical diameters was confirmed. The relation between $\log \tan \alpha$ and particle diameter D is represented by two straight lines, one for particles smaller than the critical diameter D_c, and another for larger particles, as shown in Fig. 3 and 4.

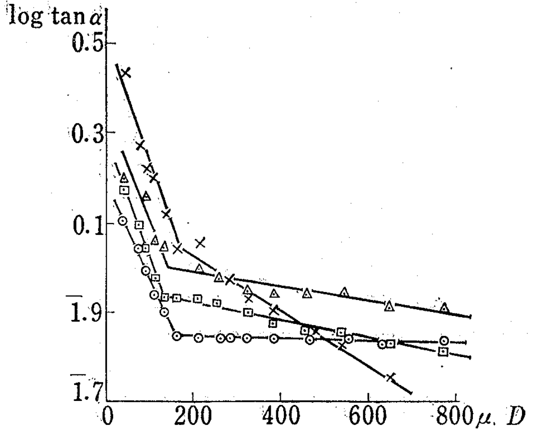

Fig. 3. Relation between Particle Size and Angle of Repose or Sliding Angle of Sodium Borate Powders

By four methods.

⊙ Nogami-Sugiwara

⊡ Neumann

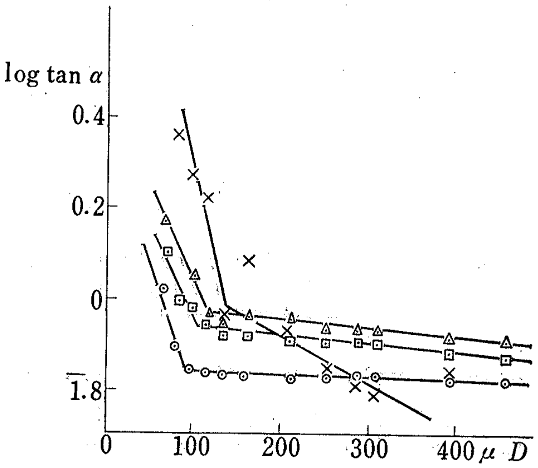

Fig. 4. Relation between Particle Size and Angle of Repose or Sliding Angle of Boric Acid Powders

△ Nelson

× Awata-Nakajima

The relation between D and $\tan \alpha$ is represented as follows.

$$D \leq D_c \qquad \tan \alpha = A_1 \exp(-B_1 D) \qquad (1)$$

$$D \geq D_c \qquad \tan \alpha = A_1' \exp(-B_1' D) \qquad (2)$$

$$A_1 > A_1', \qquad B_1 > B_1' > 0$$

When a solid begins to slide down a slope, Coulomb's equation applies.

$$Mg \sin \alpha = \mu Mg \cos \alpha + C \qquad (3)$$

Here α is the angle of slope at the time when a solid begins to slide, M is the mass of the solid, μ is the frictional coefficient between the solid and the slope, C is the adhesive force between them, and g is the gravitational constant.

We can apply this principle to powders, assuming that α is angle of repose or sliding angle, μ is the internal frictional coefficient of powders and C is the cohesive force between particles.[10] If F denotes $Mg \sin \alpha$, the following equations can be applied

$$\tan \alpha = \mu \cdot F / (F - C) \qquad (4)$$

$$\log \tan \alpha = \log \mu + \log F / (F - C) \qquad (5)$$

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[10] Z. Kuri: Oyo Butsuri, **20**, 74 (1950); H. Hayashi, S. Minami: Yakugaku Zasshi, **84**, 229 (1964).

Equation (5) shows that log tan α consists of the first term, the logarithm of internal frictional coefficient (log μ), and the second term related to cohesive force between particles of powders [$\log\{F/(F-C)\}$].

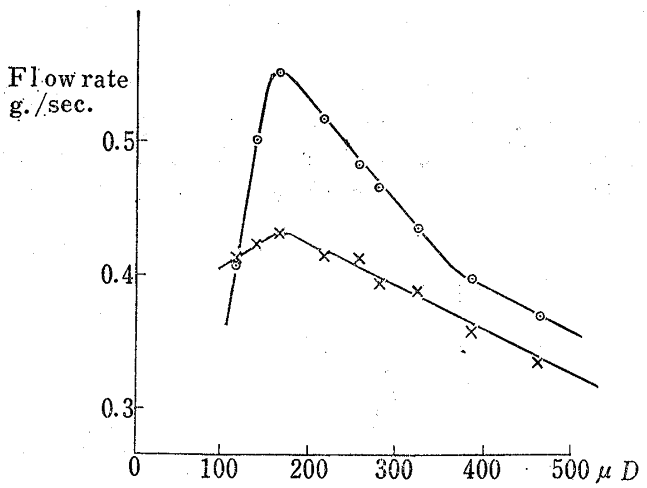

Fig. 5. Effect of Particle Size on Flowing Rate
⊙ Sodium borate powders
× Boric acid powders

Powders with large diameters flow freely, but the powders below 200 mesh are sticky. Fig. 5 shows the influence of particle size on the rate of powder flow through a brass funnel (cone angle : 60°, outlet diameter : 2.6 mm. and depth : 1 mm.). For both sodium borate and boric acid powders, the rate gradually increases with decrease of particle size for particles larger than 160 μ, but for smaller particles the rate rapidly decreases because of powder coherency, and for 100 μ particles the powders stop flowing.

From the above facts it seems to be most reasonable to assume that cohesive force between particles of powders is negligibly small for the region larger than the critical size but for the region smaller than that, cohesive force influence angles of repose and sliding angles. Accordingly equation (2) is identical with the internal frictional coefficient of powders (μ), and equation (1) with the product of the internal frictional coefficient of powders (μ) and the term related to cohesive force [$F/(F-C)$]. The results shown in Fig. 3 and 4 are all in good agreement with the above assumption, and on this basis the phenomena shown in Fig. 5 are understandable.

If the straight lines for the region larger than the critical size are extended to the smaller region in Fig. 3 and 4, the difference between the true line and the extended line corresponds to the difference between the logarithm of tan α and that of the internal frictional coefficients, and therefore, this difference corresponds to the second term of the equation (5). By combining equations (1), (2) and (4), we obtain

$$\tan\alpha / \tan\theta = F/(F-C) = A_1/A_1' \exp\{-(B_1-B_1')D\}, \qquad (6)$$

where θ is the value of angle on the extended part of the straight lines for the larger region in Fig. 3 and 4. Equation (6) illustrates that the ratio of tan α to the internal frictional coefficient of powders is equal to the ratio of the force of particles of sliding down, F, to the force caused only by the internal friction, $(F-C)$, and further this ratio increases exponentially with a decrease of particle size.

The coefficients of the equations (1) and (2), A_1, A_1', B_1, B_1' and the critical diameter D_c are tabulated in Table I.

The critical diameter D_c is almost constant irrespective of the method of analysis, but the coefficients A_1, A_1', B_1, B_1' and those of the equation (6), A_1/A_1' and (B_1-B_1') are dependent on the method. Therefore it appears that each method measures different coefficient of friction.

If the internal frictional coefficients (μ) is set equal to tan θ, equation (3) becomes

$$C/Mg = \sin(\alpha-\theta)/\cos\theta, \qquad (7)$$

where the value of θ is obtained from the extended part of the straight lines in Fig. 3

Table I. Experimental Constants on Fluidity of Powders

Sample	Method	D_c (μ)	A_1	A_1'	B_1 (cm^{-1})	B_1' (cm^{-1})	A_1/A_1'	(B_1-B_1') (cm^{-1})
Sodium borate	Ⅰ	160	1.61	0.71	51	4.3	2.3	47
	Ⅱ	180	1.92	0.98	40	2.5	2.0	37
	Ⅲ	140	2.00	0.91	56	4.2	2.2	52
	Ⅳ	150	3.55	1.42	74	14.0	2.5	60
Mean		158						
Boric acid	Ⅰ	95		0.68		0.2		
	Ⅱ	105		0.94		2.9		
	Ⅲ	100		0.86		2.8		
	Ⅳ	130						
Mean		108						

Ⅰ : Nogami-Sugiwara Ⅱ : Nelson Ⅲ : Neumann Ⅳ : Awata-Nakajima

and 4. By any method nearly equal values were obtained for C/Mg as shown in Fig. 6. The relation between particle size and C/Mg was investigated only for sodium borate powders, because the plotted numbers in the region smaller than the critical size are few in the case of boric acid.

The results in Fig. 6 show that log C/Mg is approximately linearly related to D in the region between 45 μ and 160 μ. Accordingly, the relation between C/Mg and partcle diameter D is represented as follows.

$$C/Mg = \mathrm{E}\exp(-FD). \qquad (7)$$

The numerical values of the coefficients, E and F are obtained from Fig. 6, and are 0.81 and 147, respectively, for sodium borate powders.

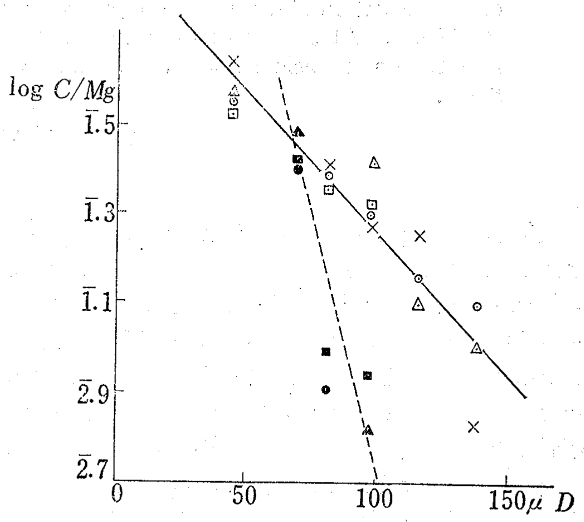

Fig. 6. Relation between Particle Size and Cohesive Force

Method	Sodium borate	Boric acid
Nogami-Sugiwara	⊙	●
Nelson	△	▲
Neumann	⊡	■
Awata-Nakajima	×	

If the equation (7) is applicable to boric acid powders, the coefficients E and F, obtained from Fig. 6, are larger than those of sodium borate powders. Therefore, the coefficients E and F seem to depend on the characteristics of the particles of powders.

We wish to acknowledge Prof. T. Kwan, University of Tokyo, for his kind advice. Thanks are also due to Prof. H. Nogami, University of Tokyo, for his guidance of experimental techniques.