Simulation of Composite Particle Growth in the Dispersion and Compounding Processes of Alloy Particles and Ceramic Powder[†]

Tomohiro Iwasaki, Takeshi Yanagida
and Munetake Satoh

Dept. of Chem. Eng., Osaka Pref. Univ.*

Abstract

The authors investigated a compounding process in which fine WC powder is uniformly dispersed into particulate Ag-Ni alloy with a high-speed elliptical-rotor-type powder mixer. The growth of composite particles is observed as processing time elapses. A model for the growth phenomenon is constructed on the basis of the theory of powder grinding with a ball mill, and of the dynamics of the plastic deformation of metal particles. In the model, a function of the probability that composite particles grow is expressed in terms of three factors that include the compounding conditions as parameters: (1) the probability that particles are caught between a pair of medium balls (zirconia beads), (2) that they have undergone plastic deformation, and (3) the frequency with which the mixture of composite particles and beads is compressed by the elliptical rotor at the minimum clearance per unit time. Temporal change in the size distribution of composite particles has been calculated by using model equations. Comparison of calculated values with the experimental data for particle median diameter shows that they agree closely. It was demonstrated that the model is valid and that it is possible to accurately estimate and control particle size.

1. Introduction

In the powder processing in which the particle size changes, such as granulation, comminution, and others, formulating the size distribution of products in time series makes it possible to control the products' physical properties and estimate the processing time in which products of a required state are obtained. Recently in the field of powder metallurgy there has been active study of mechanical alloying (MA), mechanical grinding (MG), and other processes in which materials are compounded mechanically. However, there has been little research analyzing temporal changes in the size of composite particles on the basis of kinetics.

The authors have evaluated processing in which fine ceramic powder (tungsten carbide) is dispersed uniformly into a particulate alloy of silver and nickel with a high-speed elliptical-rotor-type powder mixer. Effects of compounding conditions on the state of the

composite particles obtained were investigated by measuring the temporal change in the lightness and size of composite particles.^{1,2} A simplified model of chemical reaction between two species was applied to the growth phenomenon of composite particles as time passed during the process. It was found that experimental conditions, such as the volume ratio of the powder mixture to the medium (zirconia beads) and the amount of ceramic powder added, had a great qualitative influence on the rate of particle growth and the degree of compounding.²

In this paper, a kinetics model of particle growth has been constructed on the basis of the theory of powder grinding with a ball mill³ and of the dynamics of the plastic deformation of metal particles. This model takes into account the operating conditions of the powder mixer, the physical properties of powders, and the effects of ceramic powder in composite particles on plastic deformation. Temporal changes in the size distribution of composite particles have been simulated in terms of the model equations.

2. Model of particle growth

2.1 Kinetics model

The frequency distribution function $f(D_p, t)$ of the composite particles with diameter D_p (area basis) at

* 1-1 Gakuen-cho, Sakai, Osaka 599-8531, Japan
TEL. 0722-54-9307 FAX. 0722-54-9911

[†] This report was originally printed in J. of Japan Soc. of Powder and Powder Metallurgy, **45**, No. 4 362-367 (1998) in Japanese, before being translated into English by KONA Editorial Committee with the permission of the editorial committee of the Japan Society of Powder and Powder Metallurgy.

processing time t is derived from the population balance as follows:

$$\frac{d}{dt} [S(t) \cdot f(D_p, t) \cdot dD_p] = S_{in} - S_{out} \quad (1)$$

where $S(t)$ indicates the total projected area of composite particles. S_{in} and S_{out} are the projected areas of the composite particles, the sizes of which are within and without the size range between D_p and $D_p + dD_p$ per unit time, respectively.

Observations of composite particles with a scanning electron microscope confirmed that composite particles were discoidal and that their thickness was uniform in the compounding process.² We proposed a simplified model for the particle growth phenomenon of the process. As illustrated in **Figure 1**, the model indicates that composite particles grow in the following manner: (1) a pair of colliding beads holds two disk-shaped particles (diameter $D_p/\sqrt{2}$ and thickness h) and sticks the particles together, and (2) a particle of diameter D_p (particle density and thickness are same as those of the particles before growth) is newly formed by the fused particles, causing plastic deformation. S_{in} and S_{out} are given by Eqs. (2) and (3):

$$S_{in} = P_z(D_p/\sqrt{2}) \cdot S(t) \cdot f(D_p/\sqrt{2}, t) \cdot dD_p \quad (2)$$

$$S_{out} = P_z(D_p) \cdot S(t) \cdot f(D_p, t) \cdot dD_p \quad (3)$$

where $P_z(D_p)$ is a function of the probability that a particle of diameter D_p will grow per unit time.

Fig. 1 Compounding model for disk-shaped particles

2.2 Theoretical analysis of the probability function $P_z(D_p)$ of particle growth

The probability function $P_z(D_p)$ of particle growth is defined as the product of a function $P_n(D_p)$ of the probability that two particles are held between a pair of beads, $P_{pl}(D_p)$ that plastic deformation occurs, and the frequency Z with which the mixture of composite particles and beads is compressed by the rotor at the

minimum clearance per unit time.

$$P_z(D_p) = P_n(D_p) \cdot P_{pl}(D_p) \cdot Z \quad (4)$$

Z is given by

$$Z = 2(N_r + N_v) \quad (5)$$

where N_r and N_v are the rotational speeds of the rotor and the vessel, respectively.

Probability of holding composite particles

The apparent volume V_a of the mixture of metal particles and ceramic powder in the vessel is expressed on the basis of the charging condition of the powder mixture and the beads as follows:

$$V_a = V_m f \frac{V_p/V_b}{1 + V_p/V_b} \quad (6)$$

V_m is the effective volume of the vessel, and f is the charge volume ratio. V_p and V_b are the apparent charge volumes of the powder mixture and the beads, respectively.

By assuming that two composite particles held between a pair of beads adhere rapidly to each other and then form one particle in appearance, the apparent volume v_a of the composite particles existing in voids among the beads is expressed geometrically (see **Figure 2**).

$$v_a = \pi b^2 D_p \quad (7)$$

$$b^2 \cong \frac{1}{2} D_p D_b \quad (8)$$

where D_p and D_b are the diameters of the composite particle and the bead, respectively.

As illustrated in **Figure 2**, the forces $\mu F \cos^2 \alpha$ and $F \cos \alpha \sin \alpha$ act simultaneously on the composite particles held between a pair of colliding beads: the former is the force by which composite particles are pulled in between the beads, and the latter repels the particles. F is the impact force of the beads and μ is coefficient of dynamic friction. The composite particles are

Fig. 2 Schematic diagram of a composite particle held between a pair of beads

caught by the beads when the following condition is satisfied:

$$\mu F \cos^2 \alpha \geq F \cos \alpha \sin \alpha \quad (9)$$

$$\therefore \tan \alpha \leq \mu \quad (10)$$

The area ratio γ of the region where the composite particles are held and that where the forces act upon them is expressed by Eq. (11) in terms of $\xi = \tan^{-1} \mu$.

$$\gamma = \frac{[(D_b/2) \tan \xi]^2}{b^2} = \frac{\mu^2}{2} \left(\frac{D_b}{D_p} \right) \quad (11)$$

Therefore, $P_n(D_p)$ is expressed by Eq. (12).

$$P_n(D_p) = \frac{v_a}{V_a} \times \gamma = \frac{\pi \mu^2 D_p D_b^2}{4 V_m f} \left(\frac{V_p/V_b}{1 + V_p/V_b} \right)^{-1} \quad (12)$$

In consideration of the shape of composite particles in the compounding process, Eq. (12) is corrected with the shape factor ϕ .

$$P_n(D_p) = \frac{\pi \mu^2 D_p D_b^2}{4 V_m f} \left(\frac{V_p/V_b}{1 + V_p/V_b} \right)^{-\phi} \quad (13)$$

ϕ is defined by the following equation:

$$\phi = A/A_s \quad (14)$$

where A and A_s indicate the projected areas of composite particles and spherical particles, respectively, having volumes equal to that of the composite particle.

Probability of plastic deformation occurring

From the equilibrium of stress in the radial direction in the disk-shaped particle as shown in **Figure 3**, the following equation is obtained in terms of the approximation $\sin(d\theta/2) \approx d\theta/2$.

$$(\sigma_r h - \sigma_\theta h + 2\tau r) dr + r h d\sigma_r = 0 \quad (15)$$

If we assume that the composite particle is an ideal plastic solid and that the frictional condition between the composite particle and the bead is described by

Fig. 3 Stresses acting on disk-shaped particles

using a constant coefficient of Coulomb friction without sliding, the relation $\sigma_\theta = \sigma_r$ holds from the Levy-Mises equation.⁴ The shearing stress τ is expressed by Eq. (16) from von Mises' yield criterion.⁴

$$\tau = -\sigma_0/\sqrt{3} \quad (16)$$

σ_0 represents the yield stress of a composite particle. The following equation is obtained by substituting Eq. (16) into Eq. (15).

$$\frac{d\sigma_r}{dr} - \frac{2\sigma_0}{\sqrt{3}h} = 0 \quad (17)$$

By disregarding the effects of shearing stress at the edge of a composite particle on plastic deformation, σ_r is expressed by

$$\sigma_r = \sigma_0 - p \quad (18)$$

where p indicates the stress required to cause plastic deformation. The following equation is obtained by solving Eq. (17) (after substitution of Eq. (18)) with the boundary condition $p = \sigma_0$ at $r = D_p/2$.

$$p = \sigma_0 \left[1 + \frac{2}{\sqrt{3}h} \left(\frac{D_p}{2} - r \right) \right] \quad (19)$$

The average $\bar{p}$ of the stress p is given by Eq. (20).

$$\bar{p} = \frac{\int_0^{D_p/2} 2\pi p r dr}{\pi D_p^2/4} = \sigma_0 \left(1 + \frac{D_p}{3\sqrt{3}h} \right) \quad (20)$$

$P_{pl}(D_p)$ is defined as a function whose value decreases as diameter D_p of a composite particle increases because of a linear relation between $\bar{p}$ and D_p as indicated by Eq. (20).

$$P_{pl}(D_p) = k D_p^{-g(C_{WC})} \quad (21)$$

k is a constant that is dependent on physical properties of materials. $g(C_{WC})$ represents the magnitude of the effect of ceramic powder concentration C_{WC} in the composite particle on plastic deformation. The following equation is obtained by substituting Eq. (20) into Eq. (21).

$$P_{pl}(D_p) = k \left[3\sqrt{3}h \left(\frac{\bar{p}}{\sigma_0} - 1 \right) \right]^{-g(C_{WC})} \quad (22)$$

$P_{pl}(D_p)$ is calculated with Eq. (22) by assuming that the impact force between a pair of beads is equivalent to the centrifugal force generated by the rotational motion of the rotor.

Both the elastic modulus and strength of fine particle-dispersed metal material are proportional to the total volume of fine particles in the material. The movement of the dislocation line in the material,

which is necessary for causing plastic deformation, becomes more difficult as the amount of fine particles increases.⁵ Accordingly, a simple formula is used to express $g(C_{WC})$.

$$g(C_{WC}) = c_1 + c_2 \cdot C_{WC} \quad (23)$$

c_1 and c_2 are constants that depend on the physical properties of materials.

3. Experimental and calculation methods

The appropriate amount of the mixture of gas-atomized Ag-Ni alloy particles (Ni content 3.2 wt%, median diameter $7 \mu\text{m}$, true density $1.05 \times 10^4 \text{ kg/m}^3$) and WC fine powder (median diameter $0.7 \mu\text{m}$, true density $1.56 \times 10^4 \text{ kg/m}^3$) was charged together with a medium (zirconia beads) into a high-speed elliptical-rotor-type powder mixer.⁶ The charge volume ratio f (i.e., apparent volume of the powder-bead mixture divided by the effective volume of the vessel) was 0.2. The rotational speed of the vessel was maintained at 1.67 s^{-1} , and rotor speed N_r was set at 25.0 – 41.7 s^{-1} . The volume ratio V_p/V_b of the powders to the beads was varied within the range of 0.3–1.5, the concentration C_{WC} of WC in the composite particles within the range of 1.0–10 wt%, and the size D_b of the beads in the range of 0.3–1.0 mm. An image analyzer mounted on a metallurgical microscope was used to measure more than 5,000 particles to determine the size distributions (area basis) of composite particles.

The constants k , c_1 and c_2 , which were necessary to calculate size distribution, were determined ($k=5.00 \times 10^4$, $c_1=14.2$ and $c_2=3.54 \times 10^{-2}$) from the particle compounding experiments under the several conditions². Calculations were based on the assumptions that WC powder was dispersed uniformly in the powder mixture at the processing time $t=0$ and that the yield stress of composite particles was dependent only on the WC concentration.

4. Results and discussion

4.1 Size distribution of composite particles

Figure 4 shows a typical example of calculation and experimental results for temporal change in the size distribution of composite particles. In the early stage of processing, the particle growth phenomenon caused by the plastic deformation of particles was highly evident. Particle growth reached the equilibrium state as processing progressed owing to the balance between the forces necessary for causing plastic deformation and acting on the composite particles.

Fig. 4 Change in size distribution curves with the passage of processing time t ($C_{WC}=10 \text{ wt\%}$, $D_b=1.0 \text{ mm}$, $N_r=41.7 \text{ s}^{-1}$, $V_p/V_b=0.3$)

Fig. 5 Change in D_{50} with the passage of processing time t

Figure 5 shows the temporal change in median diameter D_{50} obtained from the data in Figure 4. Calculation results coincided closely with experimental results. In the following section, the validity of the proposed model is verified by comparing calculated values with the experimental data for D_{50} .

4.2 Effects of the compounding conditions

Figure 6 shows the relation between the median diameter D_{50} of composite particles and the processing time t when varying the powder-bead volume ratio V_p/V_b . In calculations and experimental results the particle growth rate decreased as V_p/V_b increased

Fig. 6 Effect of volume ratio V_p/V_b on temporal change in D_{50}

Fig. 8 Effect of rotor speed N_r on temporal change in D_{50}

Fig. 7 Effect of WC concentration C_{WC} on temporal change in D_{50}

Fig. 9 Effect of bead diameter D_b on temporal change in D_{50}

because of the decreasing probability that composite particles were held by beads.

Figure 7 shows the effect of WC powder weight concentration C_{WC} on the temporal change in D_{50} . The size of composite particles in the late stage of processing declined with the increase in C_{WC} . This means that the presence of WC powder in composite particles prevented the composite particles from causing the plastic deformation that is necessary for growth.

Figure 8 shows the effect of rotor speed N_r on particle growth. At higher N_r , the particle growth rate increased due to the increase in the frequency per unit time with which the mixture of composite particles and beads was compressed by the rotor at the minimum clearance. However, comparing the results in Figure 8 with those in Figure 6 shows that the effect of N_r on particle growth was smaller than that of V_p/V_b . At higher rotor speeds, particle growth occurs only with difficulty even though the compres-

sion frequency increases more if the amount of beads is insufficient to cause plastic deformation.

Figure 9 shows the calculation and experimental results obtained when bead diameter D_b was varied. Composite particle size decreased when using smaller-sized beads because of change in the stress acting on composite particles and in the probability of holding a composite particle.⁷

Close agreement between calculated values and experimental data for particle size in Figures 6 through 9 supports the validity of the proposed model.

5. Conclusion

We performed a kinetic analysis, and investigated the mechanism, of a composite particle growth phenomenon brought about by plastic deformation of composite particles occurring in a compounding process involving the uniform dispersion of fine

ceramic powder into alloy particles. The following conclusions can be drawn.

1) Particle growth in the compounding process has been conceived as a probability phenomenon in which composite particles are caught between a pair of media and subjected to plastic deformation. A simplified model of the phenomenon has been proposed in terms of the probability function of composite particle growth. The validity of the model has been confirmed by agreement between calculated values and experimental data.

2) Both the calculations and the experiments clarified the effects of compounding conditions, i.e., the powder-media volume ratio, the amount of ceramic powder added, rotor speed, and medium diameter, on particle growth.

Nomenclature

A	: projected area of composite particle	(m ²)
A_s	: projected area of spherical particle having volume equal to that of composite particle	(m ²)
b	: constant expressed by Eq. (8)	(m ²)
C_{WC}	: weight concentration of WC in composite particles	(%)
c_1, c_2	: constants in Eq. (23)	(-)
D_{50}	: area median diameter of composite particle	(m)
D_b	: bead diameter	(m)
D_p	: disk-shaped particle diameter	(m)
F	: impact force between a pair of beads	(N)
f	: charge volume ratio	(-)
$f(D_p, t)$	: frequency distribution function	(m ⁻¹)
$g(C_{WC})$	: function of C_{WC} defined by Eq. (23)	(-)
h	: height of disk-shaped particle	(m)
k	: constant in Eq. (21)	($\mu\text{m}^{g(C_{WC})}$)
N_r, N_v	: rotational speeds of rotor and vessel	(s ⁻¹)
$P_n(D_p)$	: probability function expressed by Eq. (13)	(-)
$P_{pl}(D_p)$	: probability function defined by Eq. (21)	(-)
$P_z(D_p)$	: probability function defined by Eq. (4)	(-)
p	: stress required to cause plastic deformation	(Pa)
S_{in}, S_{out}	: projected areas of composite particles expressed by Eqs. (2) and (3)	(m ²)

$S(t)$	: total projected area of composite particles	(m ²)
t	: processing time	(s)
V_a	: apparent volume of powder mixture	(m ³)
V_b	: apparent charge volume of beads	(m ³)
V_m	: effective volume of mixer vessel	(m ³)
V_p	: apparent charge volume of powder mixture	(m ³)
v_a	: apparent volume of composite particles present in voids among beads	(m ³)
Z	: frequency expressed by Eq. (5)	(-)
α	: angle representing contact point between composite particle and bead	(rad)
γ	: area ratio defined by Eq. (11)	(-)
μ	: coefficient of dynamic friction between composite particle and bead	(-)
σ_0	: yield stress of composite particle	(Pa)
σ_r, σ_θ	: stresses applied in directions of r and θ	(Pa)
τ	: shearing stress	(Pa)
ϕ	: shape factor defined by Eq. (14)	(-)

References

- 1) Satoh, M., T. Yanagida and T. Iwasaki, "Dispersion and Compounding Process of Particulate Ag-Ni Alloy and Fine WC Powder Using a High-speed Elliptical-rotor-type Powder Mill," *J. Jpn Soc. Powder and Powder Met.*, **44**, 618-621 (1997).
- 2) Iwasaki, T., T. Yanagida and M. Satoh, "Effect of Operating Conditions in Powder Metal Compounding Process on Particle Size Change," *J. Jpn Soc. Powder and Powder Met.*, **45**, 271-275 (1998).
- 3) Kuwahara, Y., F. Saito and S. Yashima, "An Analysis of Selection Function of Vibration Mill," *J. Res. Ass. Powder Technol., Japan*, **14**, 319-325 (1977).
- 4) Dieter, G. E., *Mechanical Metallurgy*, 3rd ed., McGraw-Hill, Inc. (1986).
- 5) Fukuda, H., R. Yokota and I. Shiota, *Fukugou Zairyou Kisokougaku*, Nikkan Kougyo (1994).
- 6) Satoh, M., T. Yoshida, K. Miyanami and Y. Okudaira, "Evaluation of the Performance of a High-speed Elliptical-rotor-type Powder Mixer," *J. Soc. Powder Technol., Japan*, **31**, 789-794 (1994).
- 7) Yamauchi, K., Y. H. Park, H. Hashimoto and R. Watanabe, "Change in Morphology and Some Properties of Ti-Al Powder Particles during Mechanical Alloying by Vibratory Ball Milling," *J. Jpn Soc. Powder and Powder Met.*, **38**, 42-46 (1991).

Author's short biography **Tomohiro Iwasaki**

Tomohiro Iwasaki received the master degree in chemical engineering from the Osaka Prefecture University in 1996. He joined the Department of Chemical Engineering, the Osaka Prefecture University, as an Assistant Professor in 1997. His major research interests are design of functional composite particle, evaluation of manufacturing process of composite particles, simulation of behavior of particle in compounding process, measurement and evaluation of dynamic physical properties of powder, and analysis of stress in powder bed.