Simulation of Ball Motion for Analysis of Coating Phenomena during Tumbler-Ball Milling of Cu Powder

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A copper powder was milled in argon by a tumbler-ball mill to investigate the formation of copper coatings on ball surfaces and mill pot walls, which must be a significant process in mechanical alloying. Weight ratio of the coatings to an initial powder-filling was measured after milling at various ball- and powder-filling ratios. Experimental results on the coating weight ratio were analyzed by a two-dimensional computer simulation of ball motion.

It was found that the coating weight ratio increases with an increase in the ball-filling ratio in a range of the ratios from 10 to 40% and it decreases rapidly between 50 and 60%. It was also found that the coating mass ratio decreases with increase in the powder-filling ratio. The computer simulation revealed that the coating behavior is influenced by the ball motion which was simulated by taking into account the consumption of kinetic energy of milling balls. It was found that the coating mass ratio depends on the frequency of impacts per ball.

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I. Introduction

Typical mechanical alloying is a dry ball milling process of a premix of elemental metal powders in an inert atmosphere. In an early stage of milling, the metal powder particles are microformed into flakes and cold-welded on ball surfaces and walls of mill pot by ball collisions. Through repeated microforming and cold-welding thin composite metal layers or coatings are formed on the ball surfaces and pot walls. The coatings play a significant role in the mechanical alloying, since lamellar structure, which characterizes the mechanical alloying process, generally develops in the coatings. Helstern and Schultz[1] reported that the formation of an Fe-Zr alloy and its amorphization proceed in the lamellar coatings and an amorphous powder is formed by break-off of the amorphized coatings. Schwartz et al.[2] suggested that the coatings diminish the contamination from milling balls by preventing excess wear.

Recently, it was shown that the morphological change of powder particles in the mechanical alloying of ductile metal powders, which results in eventual formation of alloys or intermetallics, was described as follows; (1) formation of flaky particles of elemental powders, (2) formation of disk-shaped composite particles having layered structure, (3) formation of large equiaxed particles with randomly oriented fine lamellar structure, (4) formation of alloy or intermetallic fine particles on the equiaxed particle surfaces[3-5]. The morphological change from elemental particles to equiaxed ones is very important since the intermetallic fine particles are formed on the equiaxed particle surfaces. The authors[6] reported that the equiaxed particles were formed by agglomeration of the disk-shaped particles. The disk-shaped particles were formed by break-off of the coatings, which were formed on the ball surfaces and mill pot walls during milling, due to the impacting action of the balls[7]. Thus, the coating phenomenon has a special relation to the mechanical alloying, especially to the morphological change of particles. However, few studies have been reported on the coating phenomenon during milling of metal powders.

In this study, a copper powder was milled in argon by a tumbler mill to investigate the coating phenomenon. The reason why only the copper powder was milled in the present study is to investigate the simple mechanism of coating formation by eliminating influences of the alloy formation. Since the coatings are formed by the impacting action of the balls, the coating behavior is presumed to have a close relation to the ball motion. It is well known that the ball motion in tumbler mills strongly depends on milling conditions such as ball-filling ratio and powder-filling ratio. Therefore, weight ratio of the coatings to an initial powder-filling was chosen as a measure for various ball- and powder-filling ratios. Experimental results on the coating weight ratio were analyzed by a two-dimensional computer simulation of ball motion during milling.

II. Experimental Method

An electrolytic Cu powder (purity of 99.9%, average size of 48 μm) was milled in argon by a laboratory tumbler mill. Before milling, the powder was annealed at 676 K for 3.6 ks in a H2 atmosphere to remove oxide. The annealed powder was put in a stainless steel (SUS304)
mill pot (70 mm in inner diameter and 135 mm in inner length) with 12.7 mm hardened steel (SUJ2) balls in an argon glove box. In order to investigate the influence of ball-filling quantity on the coating behavior, the ball-filling ratio was varied from 10 to 70% of a full ball-filling in the pot with a constant powder/ball mass ratio, 1/63. The powder occupied 10% of void space in the ball-filling under this condition (powder/ball mass ratio 1/63). The effect of powder-filling amount was investigated by varying the volume ratio of powder to void space in the ball-filling from 10% to 100% at a constant ball-filling ratio, 50%. The milling pot was rotated at an angular speed of 11.4 rad/s. After milling for 144 ks (40 h), all the milled powder and the milling balls were taken out of the pot and were separated by a sieve. The free separated powder was weighed to determine the mass of coatings as a difference in mass between the initial powder-filling and the milled free powder. The weight ratio of the coatings to the initial powder-filling was used as the coating ratio in the following.

III. Model Simulation

The coating behavior is presumed to have a close relation to the ball motion as mentioned above. Since the motion of individual balls in tumbler mills is very complicated, it has never been studied in detail. However, recent developments in computer technology enable us to simulate the motion of a great number of individual discrete elements. In this study, the ball motion in the laboratory tumbler mill was simulated by a computer simulation based on a two-dimensional discrete element method (2D-DEM).

As shown in Fig. 1, 2D-DEM assumes that two parallel couplings of a spring and a dashpot act between two elements in contact, one acts in the normal direction to the contact surface and the other in the tangential direction. The spring represents the elastic behavior of the elements and the dashpot the viscous response in motion. The elastic force acting in the normal direction to the contact surface between two milling balls is given by,

$$ F_{e,i,j} = K_{i,j} A_{i,j} $$

where $F_{e,i,j}$ [N] is the normal elastic force acting on the contact surface between ball $i$ and $j$, $K_{i,j}$ [N/m] is Hook’s coefficient of the normal spring and $A_{i,j}$ [m] is an amount of approach of the balls in the normal direction owing to the elastic deformation and is correspondent to the displacement of the normal spring. In the present simulation, Hertz contact theory was applied to deduce the force-displacement relationship. In this case, the normal spring shows non-linear behavior and the spring coefficient is given as a function of $A_{i,j}^{1/2}$ as follows:

$$ K_{i,j} = \frac{1}{3} \left( \frac{R_i R_j}{R_i + R_j} \right)^{1/2} \left( \frac{E_i}{1-v_i^2} + \frac{E_j}{1-v_j^2} \right) A_{i,j}^{1/2} $$

where $R_i$ [m], $E_i$ [Pa] and $v_i$ are radius, elastic coefficient and Poisson’s ratio of ball $i$, respectively. $R_j$ [m], $E_j$ [Pa] and $v_j$ are those of ball $j$. In the case of contact of ball $i$ with a wall of the milling pot, radius of the wall was assumed to be infinite. The amount of approach was given as a difference between $(R_i + R_j)$ and distance between the two ball centers.

The dashpot is able to be related to a coefficient of restitution of the elements. It means that the dashpot is able to be related to the dissipation of energy at collision of the elements. In this study, the dashpot was assumed to describe the energy consumption at ball collisions during milling of the copper powder. During milling, the balls collide with each other and with the pot walls. These collisions can make compressive forces act on the copper particles and may deform them plastically. The plastic deformation makes new fresh surfaces of the particles which are so active that the particles are easily cold-welded on the ball surfaces and pot walls by the compressive forces. Therefore, the normal compressive forces are presumed to play an important role in the coating formation. On the other hand, the role of the tangential forces in the coating formation has not yet been clear. For this reason, the tangential coupling was omitted in the present simulation. Colliding balls deform themselves and simultaneously compress the copper particles between them. The ball centers approach each other due to the elastic deformation and go away from each other after the maximum approach due to the elastic repulsion force. The copper particles compressed between the balls are deformed plastically until the maximum approach of the ball centers and they are no longer
deformed plastically after the maximum approach. In other words, the plastic deformation of the particles consumes a part or full of the impact energy of balls until maximum compression. Therefore, the dashpot was assumed to be active only before the maximum approach in this study. It was expressed by a no-tension joint connected with the dashpot as shown in Fig. 1. As the Hertzian type force-displacement law given by eqs. (1) and (2) shows non-linear behavior of the spring, the dashpot was assumed to show also the non-linear behavior analogous to the spring (This means dashpot coefficient $C_r$ was given as a function of $A_{i,j}^{1/2}$, as shown in eq. (4).) and the damping force $F_{d,i,j}$ [N] was given by

$$F_{d,i,j} = C_r \frac{dA_{i,j}}{dt} \text{ at } \frac{dA_{i,j}}{dt} > 0$$

$$C_r = C_v A_{i,j}^{1/2}$$

where $C_r$ [kg m⁻¹ s⁻¹] is dashpot coefficient and $C_v$ [kg m⁻¹/² s⁻¹] is a constant.

The constant $C_r$ was determined by a simple restitution experiment in which one milling ball was dropped 150 mm on a stainless steel plate covered with the copper powder layer in a range of thicknesses from 0 to 0.8 mm. The energy consumption at a collision of the ball with the plate was calculated as a difference in the potential energy of the ball before drop and after rebound, which was determined by a photographic method. Figure 2 shows the energy consumption in terms of the restitution coefficient measured in the experiment as a function of the powder thickness. The restitution coefficient decreases with an increase of the thickness and ranges from about 0.15 to almost zero. This shows that the collision consumed from about 85 to 100% of the impact energy. The energy consumption under this condition was simulated through a simulation process mentioned later by using the above equations and is shown in Fig. 3 as a function of $C_r$ value. $C_r$ value giving the energy consumption between 85 and 100% range from 112500 to 2000000 kg m⁻¹/² s⁻¹.

In the present simulation, we used $C_r$ values ranging from 112500 to 3500000 kg m⁻¹/² s⁻¹. These $C_r$'s give the energy consumption from 85 to 96% under this condition.

Figure 4 shows a model of the tumbler mill used in the simulation. The model consists of 12.7 mm steel balls and a steel mill pot having the same inner diameter as the experimental mill pot. The pot was assumed to have an inner length equal to the diameter of the balls and smooth side walls to assure the two-dimensional motion of the balls. The gravitational and frictional forces given by the following equations were considered in addition to the elastic and damping ones,

$$F_g = M_i g$$

$$F_{f,i,j} = \mu F_{n,i,j}$$

where $F_g$ [N] is the gravitational force acting on ball $i$, $M_i$ [kg] the mass of ball $i$, $g$ the gravitational acceleration, $F_{f,i,j}$ [N] the frictional force acting on the contact surface between ball $i$ and $j$, $\mu$ the friction coefficient, and $F_{n,i,j}$ [N] the normal forces acting on the contact surface.
including \( F_{d,j} \) and \( F_{e,j} \).

Translational and rotational accelerations of individual balls were calculated by using vector sum of the forces and resultant moment acting on the balls. The accelerations were assumed to be constant over 1 \( \mu s \) and were integrated to yield the translational and rotational velocities of the individual balls. The velocities were used to find new positions and rotational angles of the balls. These new velocities and positions were updated by time integration to find new force values by using eqs. (1) through (5). This cycle was repeated every 1 \( \mu s \), which resulted in successive determination of the motion of individual balls every 1 \( \mu s \). From the simulation, we got useful information on collision events, for example, impact velocity, impact frequency, energy consumption, etc.

IV. Result and Discussion

Figure 5 shows photographs of the balls which were moving in the laboratory tumbler mill, taken through a transparent cover plate attached to an end of the mill pot. The photograph (a) shows the motion of bare balls and (b) shows that of the balls after coating formation. The bare balls in the mill pot seem to be sliding on the pot wall with a little change of their positions. On the other hand, the balls after coating formation show the cascading motion. The variation of ball motion should be attributed mainly to the difference in the frictional condition between the two cases. Figure 6 shows trajectories of ball paths in the two-dimensional mill which were simulated with friction coefficients of (a) 0.2 and (b) 0.6. The simulated trajectories of the two cases are in relatively good agreement as shown in Fig. 5. It is presumed that \( \mu \) is close to 0.2 in an early milling stage and increases up to 0.6 with increasing milling time.

Figure 7 shows the coating ratio as a function of the ball-filling ratio after milling for 144 ks at the constant powder/ball mass ratio, 1/63. The coating ratio increases with an increase in the ball-filling ratio in a range from 10 to 40\% and it rapidly decreases between 50 and 60\%. It should be noted that the powder/ball mass ratio was kept constant. Therefore, the influence of the ball-filling on the coating behavior shown in Fig. 7 should be ascribed to the ball motion which is dependent on the ball-filling ratio. The simulated trajectories of ball paths

Fig. 5  Photographs of the ball movement for various condition in the tumbler-mill with the 20\% ball-filling. (a) bare balls in the mill pot (b) balls in the mill pot after coating formation.

Fig. 6  Trajectories of ball paths in the two-dimensional mill at 22\% ball-filling which were simulated with friction coefficients of (a) 0.2 and (b) 0.6.

Fig. 7  Coating ratio measured after milling for 144 ks with 12.7 mm balls as a function of ball-filling ratio.
in a range of two-dimensional ball-filling ratios from 17 to 78% are shown in Fig. 8 ($\mu = 0.4$). The ball motion varies from the sliding (17%) to a mixture of the cascading and cataracting (39%) and finally to the cataracting (78%). In order to investigate the influence of ball motion on the impacting action, impact frequency for each ball was calculated in the simulation and is shown in Fig. 9 as a function of the ball-filling ratio. Paying attention to the frequency in the case of $\mu = 0.6$, it increases with an increase in the ball-filling ratio in a range from 17 to 52% and decrease between 52 to 78%. This corresponds well with the variation of coating ratio shown in Fig. 7. It suggests that the coating ratio depends on the impact frequency per ball.

The variation of coating ratio with the powder-filling ratio at a constant ball-filling (50%) is shown in Fig. 10. The increase in the powder-filling diminishes the coating ratio. Since the powder takes up a certain volume of gaps between the balls, the increase in the powder-filling ratio may thicken powder layers compressed between the balls.
As already shown in Fig. 2, the energy is consumed more as the powder layer becomes thicker. Therefore, the increase in the powder-filling ratio may increase the energy consumption at collisions. Figure 11 shows the impact frequency per ball calculated with $C_n$ values ranging from 112500 to 350000 kg m$^{-1/2}$ s$^{-1}$ which correspond to the energy consumption from 85 to 96%. The impact frequency decreases with an increase in $C_n$. This again corresponds with the variation of coating ratio shown in Fig. 10.

V. Conclusion

The copper powder was milled by the tumbler mill to investigate the coating phenomenon. The experimental results on the coating ratio were analyzed by the two-dimensional computer simulation of ball motion.

As a result, it was found that the coating ratio increases with the increase in the ball-filling ratio in a range of the ratios from 10 to 40% and it decreases rapidly between 50 and 60%. It was also found that the coating ratio decreases with the increase in the powder-filling ratio. The computer simulation revealed that the coating behavior is closely related to the ball motion which was simulated by taking into account the consumption of ball energy during milling. It was found that the coating ratio depends on the impact frequency per ball calculated in the simulation. From these results, we concluded that the coating is formed by repeated impacts of the milling balls.

REFERENCES