Computer Simulation of Milling Ball Motion in Mechanical Alloying (Overview)

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A model simulation of milling-ball motion during mechanical alloying done by the author's group was reviewed. The Kelvin's dashpot-spring model and the discrete element method were employed in the simulation. One, two and three dimensional milling-ball motions for three types of ball milling; vibratory, tumbling and planetary ball milling, were studied. The simulation formalism was first given, which takes into account the translational and rotational motion and three types of forces; visco-elastic, frictional and gravitational force. The whole motion of the milling balls was traced contact by contact. The simulation provided for each of the milling types the ball motion trajectories, impact velocities and frequencies of the balls, as well as the energy consumption during mechanical alloying. The results of the simulation were supported by the experimental observation.

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I. Introduction

Mechanical alloying, which was invented by Benjamin⁽¹⁾ as a solid-state alloying process by ball milling, has aroused a great deal of interest today because of its potentiality in developing new materials, such as dispersion strengthened materials, intermetallic compounds and amorphous alloys. Much has been done on the alloy and phase formation during mechanical alloying and on their material properties⁽²⁾⁽³⁾. However, the milling conditions have been selected only empirically for individual material systems chiefly because of poor knowledge of the dynamics of milling ball motion. From observations of morphological and structural evolutions of milled particles, the mechanical alloying process has been considered to be repeated microforging and kneading of powder particles by the collision impacts of milling balls. The lamella structure of the milled particles is a direct proof of this presumption. To analyze the mechanism of mechanical alloying, we must first know the ball motion and energy transmittance from milling balls to the powder particles.

Collisions of milling balls are considered to be a probability event which has a definite probability distribution. However, the ball motion is very complicated and the impact velocities of the balls at the collisions would have a very wide distribution. To elucidate the probability distribution for the collision events in ball milling the dynamics of the individual balls must be quantified. Another important aspect to be taken into account in the consideration of the milling ball dynamics in mechanical alloying is the adhesion of the fresh surfaces of the milled particles. Since the mechanical alloying occurs under dry milling and thus the contact area is in adhesive condition, milling balls are covered with stuck metallic layer. This

situation is quite different from conventional ball milling, in which the milling condition is dry and reactive and the adhesion is avoided as thoroughly as possible to assure an effective comminution. Hashimoto and Watanabe⁽⁴⁾ first reported the analysis of the milling ball motion during mechanical alloying by a numerical simulation based on the Cundall's discrete element method⁽⁵⁾. The effect of the adhered layer on milling balls has been expressed by a dashpot-spring model⁽⁶⁾⁽⁷⁾. In the simulation, one, two and three dimensional⁽⁸⁾ ball motions, and three types of ball milling; tumbler⁽⁶⁾⁽⁹⁾, vibratory⁽⁸⁾⁽¹⁰⁾ and planetary⁽¹¹⁾ ball milling, have been studied.

The present paper reviews the ball milling simulation for mechanical alloying done by the authors' group. The first part of this paper describes the dashpot-spring model employed and the simulation scheme by the discrete element method. Then, the application of the simulation to each of the ball milling methods is outlined. Finally the simulation results and the discussion are given.

II. Simulation Scheme

1. Basic model

Figure 1 shows an interaction model of two balls at collision during mechanical alloying. The model is consisted of a parallel coupling of a spring and dashpot, two no-tension joints and a slider. The normal and tangential forces are given at the contact. The spring simulates the elastic behavior of the balls, the dashpot a viscous response due to the adhered metal layer and the slider the frictional force. The tangential coupling⁽¹²⁾ was omitted for the ease of the calculation in the present simulation.

Fig. 1 Spring and dashpot model used in the two-dimensional discrete element method.

2. Discrete element method

The simulation uses an explicit numerical scheme in which the motion of the individual balls are traced and the interaction of the balls are monitored contact by contact. The time interval for each calculation is small enough that a single event disturbance propagates only to the nearest neighbours. The contact forces and the displacement of the individual balls are calculated assuming that the velocities and accelerations of the balls are constant over each time step⁽⁵⁾.

3. Simulation formalism

The elastic, damping, gravitational and frictional forces have been taken into account in the simulation. The elastic force acting in the normal direction is given by,

$$Fe_{i,j} = K_{i,j} \Delta_{i,j} \quad (1)$$

where $Fe_{i,j}$ [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 $\Delta_{i,j}$ [m] is an amount of approach of the two balls in the normal direction pertaining to the elastic deformation and is correspondent to the imaginary displacement of the spring component. Hertz contact theory⁽¹³⁾ was applied to deduce the force-displacement relationship. In this case, the spring shows non-linear behavior and the spring coefficient $K_{i,j}$ is given as a function of the displacement $\Delta_{i,j}$ as follows⁽¹³⁾:

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

where R_i [m], E_i [Pa] and v_i are radius, elastic coefficient and Poisson's ratio of ball i , respectively, and R_j [m], E_j [Pa] and v_j are those of ball j . The amount of approach was given as a difference between $(R_i + R_j)$ and the distance between two ball centers.

The dashpot represents the dissipation of energy at the collision of balls and is related to a coefficient of restitution of the elements⁽¹⁴⁾. In the present simulation, the dashpot was assumed to describe the energy consumption at the ball collisions during milling of metal powders. At the collision, the ball centers approach each other and the metal particles are compressed. The particles deform plastically by the compressive and shear forces. The plastic deformation by the normal compressive force takes place until a maximum approach of the ball centers and consumes the impact energy of balls during the plastic deformation. In such situation, the normal dashpot has been assumed to be active only before the maximum approach and no longer after that. It is 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 a non-linear behavior of the spring, the dashpot has been assumed to show also the non-linear behavior analogous to that of the spring. This means dashpot coefficient C_r is given as a function of the amount of approach $\Delta_{i,j}$, as given by eq. (3),

$$C_r = C_v \Delta_{i,j}^{1/2} \quad (3)$$

where C_r [$\text{kgm}^{-1} \text{s}^{-1}$] is a dashpot coefficient and C_v [$\text{kgm}^{-1/2} \text{s}^{-1}$] is a proportional constant. The damping force $Fv_{i,j}$ [N] is given by eq. (4).

$$Fv_{i,j} = C_r \frac{d\Delta_{i,j}}{dt} \quad \text{at} \quad \frac{d\Delta_{i,j}}{dt} \geq 0 \quad (4)$$

The constant C_v of the dashpot was determined by a simple restitution experiment in which a milling ball was vertically dropped by 150 mm on a stainless steel plate covered with a metal (copper) power layer with a thicknesses of from 0 to 0.8 mm. The energy consumption at the collision of the ball with the plate was defined as the difference in the potential energy of the ball before drop and after rebound, which was determined by a photographic method⁽⁶⁾. Figure 2 shows the energy consumption in terms of the restitution coefficient measured in the experiment as a function of the powder layer thickness. The restitution coefficient decreases with increase in the thickness and ranges from about 0.15 to almost zero. This indicates that the collision consumed the impact energy by 85 to 100%. The energy consumption under this condition is shown in Fig. 3⁽⁶⁾ as a function of C_v . The energy consumption between 85 and 100% of the impact energy gives C_v values of 1.13×10^5 to $2.0 \times 10^6 \text{ kgm}^{-1/2} \text{s}^{-1}$. In the present simulation, C_v values ranging from 1.13×10^5 to $3.5 \times 10^5 \text{ kgm}^{-1/2} \text{s}^{-1}$ were used, which give the energy consumption from 85 to 96%.

The gravitational and frictional forces are given by the following equations:

Fig. 2 Variation of restitution coefficient of 12.7 mm steel ball as a function of copper powder thickness. (Steel ball was dropped 150 mm on a stainless steel plate covered with the copper powder layer.)

Fig. 3 Energy consumption during impact of a 12.7 mm steel ball dropped 150 mm on a steel plate as a function of dashpot coefficient, C_v calculated by model simulation.

$$Fg_i = M_i g \quad (5)$$

$$Ff_{i,j} = \mu F n_{i,j} \quad (6)$$

where Fg_i [N] is the gravitational force acting on ball i , M_i [kg], the mass of ball i , g , the gravitational acceleration, $Ff_{i,j}$ [N], the frictional force acting on the contact surface between ball i and j , μ , the friction coefficient, and $F n_{i,j}$ [N], the normal force acting on the contact surface.

After finding all the forces acting on the individual balls, the equations of translation and rotation of ball i can be written as follows: (translational motion)

$$M_i \frac{d^2 x_i}{dt^2} = \sum_{j=0, j \neq i}^n F x_{i,j} \quad (7)$$

$$M_i \frac{d^2 z_i}{dt^2} = \sum_{j=0, j \neq i}^n F z_{i,j} - M_i g \quad (8)$$

(rotational motion)

$$I m_i \frac{d\eta_i}{dt} = \sum_{j=0, j \neq i}^n R_i F f_{i,j} \quad (9)$$

where $F x_{i,j}$ [N] and $F z_{i,j}$ [N] are x - and z -components of the forces acting on the contact surface between ball i and j , respectively, $I m_i$ [kgm²], the moment of inertia of ball i , and η_i [rads⁻¹] the rotational speed of ball i . In this coordinate system, x -axis is correspondent to a horizontal axis and z -axis to a vertical one.

The simulation was conducted through the following procedure with unit time step of 1 μ s (see Fig. 4): (1) initial condition (initial positions, velocities of the balls and the mill container) is given; (2) each ball is judged to be in or out of contact with other balls and with the container wall; (3) the amount of approach Δ and its time derivative $d\Delta/dt$ at each contact are calculated; (4) the normal elastic force and the normal damping force at each contact are calculated by using eqs. (1)–(4) (the damping force is calculated only in the case of $d\Delta/dt > 0$); (5) relative tangential velocity at each contact is calculated and also the frictional force is calculated by eq. (6), if the relative tangential velocity is not zero; (6) vector sum of these forces (including the gravitational force calculated by eq. (5)) and resultant moment acting on each ball are calculated; (7) translational and rotational accelerations of each ball are calculated by eqs. (7)–(9); (8) the accelerations

Fig. 4 Flow chart of model simulation program.

Fig. 5 Mathematical models of vibratory ball milling. (a) one-dimensional model (b) two-dimensional model (c) three-dimensional model.

are integrated to yield the translational and rotational velocities by assuming the accelerations to be constant over the time step; (9) the velocities are used to find new position and rotational angle of each ball in the next time step; (10) new position and velocity of the mill container are also calculated; (11) the process through (2)–(10) is repeated.

A series of the above-mentioned calculations was done to trace the movement of each ball through a series of time steps. The ball collision events were detected by comparing the amount of approach in a time step with that in the previous time step. The velocity of approach of colliding ball centers at the beginning of each collision event was calculated as the impact velocity.

III. Model of Ball Mills

1. Vibratory ball milling

To simulate the vibratory ball mill, three mathematical models, one-, two- and three-dimensional ones shown in Fig. 5⁽⁸⁾ were used. The one-dimensional model (Fig. 5(a)) consists of a cylindrical steel vial charged with steel balls whose diameter is the same as the inner diameter of the vial. No friction between the balls and the vial was assumed to assure smooth one-dimensional motion of the balls. A harmonic vibration was given to the vial in the longitudinal (vertical) direction. The two-dimensional model (Fig. 5(b)) consists of a cylindrical steel container charged with steel balls whose diameter is the same as the inner length of the container. To assure smooth two-dimensional motion of the balls, frictionless condition was adopted to the contact points between the balls and two parallel end walls of the container. The three-dimensional model (Fig. 5(c)) consists of a cylindrical steel pot and steel balls whose dimensions are the same as those used in our experiment (135 mm height $\times$ 70 mm diameter)⁽⁶⁾. However, the friction was not considered in the three-dimensional model for the ease of calculation. The same vibration as in our experiment (25 Hz) was given to the two- and three-dimensional models.

2. Tumbling ball milling

Figure 6⁽⁶⁾ shows a two-dimensional model of a tumbling ball mill used in the present simulation. The model

Fig. 6 Two-dimensional model of tumbling ball milling.

Fig. 7 Two-dimensional model of high-speed planetary ball milling.

consists of 12.7 mm steel balls and a steel mill container whose inner length is the same as the ball diameter. Frictionless condition at the contacts between the balls and the two end walls of the container was adopted to give smooth two-dimensional motion of the balls. The container was rotated at the same angular velocity as that in the experiment (11.55 rads^{-1}).

3. Planetary ball milling

A two-dimensional model of a high-speed planetary ball mill is shown in Fig. 7⁽¹¹⁾. The model is similar to the tumbling mill model. The container was revolved around the center axis of the turn table at an angular velocity 73.32 rads^{-1} with a radius of revolution of 180 mm and at the same time it was rotated at an angular velocity 36.66 rads^{-1} . The gravitational force was ignored because it is negligibly small compared with the centrifugal force induced by the revolution and rotation.

IV. Results and Discussion

1. Vibratory ball milling⁽⁸⁾

The average impact velocity of 12.7 mm balls as a function of volume fraction of ball charge (ball filling ratio) is shown in Fig. 8⁽⁸⁾, as a result of the one-dimensional model simulation. The average impact velocity has a tendency to decrease with increase in the ball filling ratio. The average impact velocity for the two- and the three-dimensional simulations are also plotted in Fig. 8. A similar tendencies to that of the one-dimensional simulation was seen, that is, the average impact velocity decreases with increase in the ball charge. Since these three model simulations showed the similar results on the relation between the average impact velocity and the ball filling ratio, the relation should be characteristic to the vibratory ball milling.

Figure 9⁽⁸⁾ shows the Vickers hardness of copper particles milled for 28.8 ks by our experimental vibratory ball

mill as a function of the ball filling ratio. The hardness tends to decrease with increasing ball filling ratio in a range of the ball filling ratios from 0.4 to 0.85. Since the hardness of the milled particles is supposed to reflect the degree of work-hardening, it is easily imagined that it should be related to the average impact velocity as will be

Fig. 8 Average impact velocity of 12.7 mm balls as a function of ball filling ratio. (1-, 2- and 3-dimensional model simulation of vibratory ball milling.)

Fig. 9 Vickers hardness of copper particles milled for 28.8 ks by vibratory ball milling as a function of ball filling ratio.

Fig. 10 Photographs of the ball movement in the tumbling ball mill with ball filling ratio 20%. (a) bare balls (b) balls coated with copper powder after 144 ks of milling.

seen by comparing this figure with the simulation results shown in Fig. 8. The deviation from this tendency at the ball filling ratio of 0.2 is considered that at this condition the hardness has not reached the saturated value because of the very low frequency of the collision.

2. Tumbling ball milling⁽⁶⁾⁽⁷⁾⁽¹⁵⁾

Figure 10⁽⁶⁾ shows the photographs of the milling balls moving in a laboratory tumbling mill, taken through 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 coated with stuck copper powder after 144 ks of milling. The bare balls in the mill pot seem to be sliding on the pot wall with small displacements. On the other hand, the balls coated with copper powder after long time milling show a cascading motion. The variation of the ball motion mentioned above should be attributed mainly to the difference in the frictional condition between the two cases. Figure 11⁽⁶⁾ shows trajectories of ball paths in the two-dimensional mill which have been simulated with the friction coefficients of (a) 0.2 and (b) 0.6. The simulated trajectories of the two cases are in relatively good agreement with the actual ones shown in Fig. 10. It is presumed that the friction coefficient is close to 0.2 for the bare balls in the mill pot and increases up to 0.6 with the formation of the coated layer on the balls during milling.

The photographs of the bare balls moving in the tumbling mill pot at ball filling ratios, (a) 20%, (b) 40% and

(c) 80% at a constant rotational speed of the mill container are shown in Fig. 12⁽⁷⁾. A remarkable variation in the ball trajectories can be seen with change in the ball filling ratio. The balls appear to be sliding at the ball filling ratio of 20%, cascading at 40% and cataracting at 80%. The calculated trajectories by the two-dimensional simulation with the friction coefficient $\mu=0.2$ are also shown in the lower part of Fig. 12. The present simulation almost completely reproduces the kinematics of the ball movements for a wide range of ball filling ratio.

In Fig. 13⁽⁶⁾ is shown the weight ratio of the copper powder cold-welded on ball surfaces and mill pot walls after 144 ks of milling to the initial powder charge as a function of ball filling ratio. The milling of copper powder

Fig. 11 Trajectories of ball paths in two-dimensional milling simulated with friction coefficients of (a) 0.2 and (b) 0.6.

Fig. 12 Photographs of bare balls movement and trajectories of ball paths simulated by two-dimensional model simulation in tumbling ball milling at ball filling ratios, (a) 20%, (b) 40% and (c) 80%.

was conducted at a constant weight ratio of charged powder to filled balls. This weight ratio increases with increasing ball filling in a range of 10 to 40% and it rapidly decreases between the ratios of 50 and 60%. The Micro Vickers hardness of the copper particles milled for 144 ks is also shown in Fig. 13 as a function of the ball filling ratio. The hardness shows a similar tendency, though it is not distinct compared to the coating weight ratio. It shows a maximum hardness in the range of the ball filling ratios from 40 to 50%. It should be noted that the weight ratio of the initially charged powder to the filled balls was kept constant. The retention of this ratio should in principle assure the constant milling energy. Therefore, the influence of the ball filling on the coating behavior and the hardness of the milled particles shown in Fig. 13 is supposed to be related to the ball motion pattern which is a only variable in this case. Then, the impact frequency for each ball motion was calculated in the simulation. The result is shown in Fig. 14⁽⁶⁾, where the impact frequency is plotted as a function of ball filling ratio. In the light of the discussion mentioned above, the simulated curves correspond well with those of the experimental curves shown in Fig. 13, particularly in the case of the simulation with $\mu=0.6$. This suggests that the impact fre-

Fig. 13 Changes in micro hardness and coating weight ratio of copper powder particles after 144 ks of milling with 12.7 mm balls as a function of ball filling ratio. (Powder filling ratio was kept 10%.)

Fig. 14 Impact frequency per ball calculated by two-dimensional model simulation with various friction coefficients, μ , as a function of ball filling ratio.

quency dominates a microforging to cause the hardness increase and the sticking.

Figure 15 shows the influence of the diameter of the tumbling mill pot on the impact action of milling balls⁽¹⁵⁾. As obtained by two-dimensional simulation for three different sizes of milling pot (70, 140 and 300 mm in inner diameter). The impact frequency of each ball increases with increase in the mill diameter, which means that the increase in the mill pot size accelerates the mechanical alloying process driven by the impact action of balls. Figure 16⁽¹⁵⁾ shows X-ray diffraction patterns of the powder premix of Ti and Al milled with two different-sized mill pots for the same milling time, where we can see that Ti_2Al was formed in the powder milled by the larger mill, while it was not the case in the powder milled by the smaller one. This supports the milling-pot-size effect depicted by the simulation.

Fig. 15 Impact frequency per ball as a function of mill diameter. (12.7 mm ball)

Fig. 16 X-ray diffraction patterns of powders milled for 360 ks by tumbling ball mills of different mill diameter sizes.

3. Planetary ball milling⁽¹¹⁾

Figure 17(a) and (b)⁽¹¹⁾ show the simulated trajectories of ball motion in a planetary ball mill for (a) normal rotation and (b) counter rotation of the mill pot, respectively. The normal or counter rotation means that the rotational direction is the same or reverse as the revolutional direction, respectively. The normal rotation gives the cataracting motion of the balls, while the counter rotation gives the cascading motion. Figure 18⁽¹¹⁾ shows the cumulative impact frequency distribution per ball as a function of the energy consumed at collision. The rightmost two points shows that the ball collision, at which the impact energy over 0.1 J is consumed, takes place about two times frequent for the normal rotation (open symbol)

Fig. 17 Trajectories of ball paths in planetary ball mill in the cases of (a) normal rotation and (b) counter rotation.

Fig. 18 Cumulative distribution of energy consumption during impact calculated by two-dimensional model simulation. ($\mu=0.6$, $J=35\%$)

compared with the counter rotation (solid symbol). The leftmost open and solid symbols indicate that total impact frequency per ball in the counter rotation is much higher than that in the normal rotation. The higher energy collisions take place more frequently in the normal rotation than in the counter rotation, while the counter rotation yields mild impact force. The simulated behavior accord well with the principle that the normal rotation multiplies the centrifugal force, while the counter rotation cancels it. This characteristic energy consumption behavior for planetary ball milling should be studied more in detail in the relation of phase formation during mechanical alloying of intermetallic systems.

V. Concluding Remarks

Computer simulation for the milling ball motion based on a simple Kelvin model, which takes into account a viscoelastic nature of the collision of balls during mechanical alloying, has proved to be useful to clarify the ball kinematics and dynamics which enable the evaluation of the energy consumption involved in this process. The average impact velocity and impact frequency of a ball are the two principal parameters obtained in the present simulation. These two parameters give the energy consumption at collisions of the milling balls during mechanical alloying. The ball kinematics is the another important depiction of the present simulation, which will show a overall energy consumption of the whole balls.

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