Modeling of Mechanical Alloying: Part II. Development of Computational Modeling Programs

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Computational modeling programs incorporating the physics of powder deformation, fragmentation, and coalescence occurring during mechanical alloying (MA) are developed. The programs utilize the equations developed in part I of this series; equations predicting the extent of powder deformation during an effective impact in MA and those specifying criteria for powder particle fragmentation and coalescence. Two programs have been developed for these purposes. One, MAP1, considers the behavior of a single species with the option of adding dispersoids. The other, MAP2, considers two ductile species being welded to form a third, composite species. Applications of the programs to previous experimental data, and for the purpose of identifying the effect of material and process variables on alloying behavior, are provided in the article following this one.

I. INTRODUCTION

IN the first article of this series,[1] we described the basic physics of deformation, coalescence, and fragmentation taking place during a ball-powder-ball collision of the type that occurs in mechanical alloying (MA) or mechanical milling (MM). This led to specification of conditions requisite for each of these events and to equations predicting how these events would affect the dimensions, morphology, and properties of the powder.

The conditions given are for a single impact in the sense that they specify physical requirements to be met during the course of a given impact. But with repetitive impacts, powder particles change their shape, hardness, and size. For each impact, then, it is necessary to determine new powder particle dimensions and powder properties and to test whether or not conditions have been met for a fragmentation* or coalescence event.

*Equation [16] of Part I, because it is written explicitly in terms of a critical deformation, obscures that the critical deformation is a cumulative one; *i.e.*, the left side of said equation is in reality a product of successive collisions. To illustrate this, we note that Eq. [1] (of this article and of part I) specifies the deformation (α) in a specific collision. In contrast, the α of Eq. [16] in part I is different. For example, suppose that $\alpha/h_o = 0.2$ for each and every collision according to Eq. [1] of part I. Thus, for the first collision, α/h_o (Eq. [16]) also is equal to 0.2. However, for the second collision, the left-hand side of Eq. [16] is equal to 0.2 + 0.2 (1 − 0.2) = 0.36 and the left-hand side of Eq. [16] after the third collision is equal to 0.36 + 0.2 (1 − .36) = 0.488, *etc.* We regret this confusion in the original article and apologize for any consequent misunderstandings.

In addition to conditions changing with successive impacts, they vary with position within the contact area during a single impact. The most basic "event" during milling is plastic deformation. The deformation of the powder trapped between two balls is given by[1]

$$\alpha(r) = Rv\left(\frac{\rho_B}{H_v}\right)^{1/2} - \frac{r^2}{R} \qquad [1]$$

where v is the relative collision velocity, r the distance from the center of contact, R the radius of the balls, ρ_B the density of the balls, and H_v the powder hardness. It is apparent that any computational scheme must monitor the deformation, and hence any events dependent on deformation, as a function of particle position during the impact.

In this article, we expand our "single impact" treatments of part I and give an overview of how they have been incorporated into a computational scheme simulating the evolution of powder characteristics with repetitive impacts. In part III of this series, some program applications are given.

II. DISCRETIZATION OF THE MODEL

Mechanical alloying and MM may be viewed as repetitive deformation processes taking place on a very small scale. Implicit in the relationships developed in part I is the dependence of powder particle coalescence and fragmentation on the deformation the powder experiences. As the critical strain requisite for these occurrences varies with material combinations and over a series of impacts, and as the strain imparted to the particles is a function of their radial position within the contact volume (the powder volume affected during a collision), the frequencies at which the specifications for coalescence/fragmentation events are met also depend upon this radial position. We proceed to determine the "number" of each of these events during an impact. The analysis is applied to a system of two starting powders, denoted as A and B, which can weld to form a third composite or alloy, species C.

The number of particles trapped between colliding balls can be approximated by

$$\psi = \frac{V_c}{\Omega} \qquad [2]$$

where V_c is the total volume of powder deformed plastically during a collision and Ω the average particle volume given by

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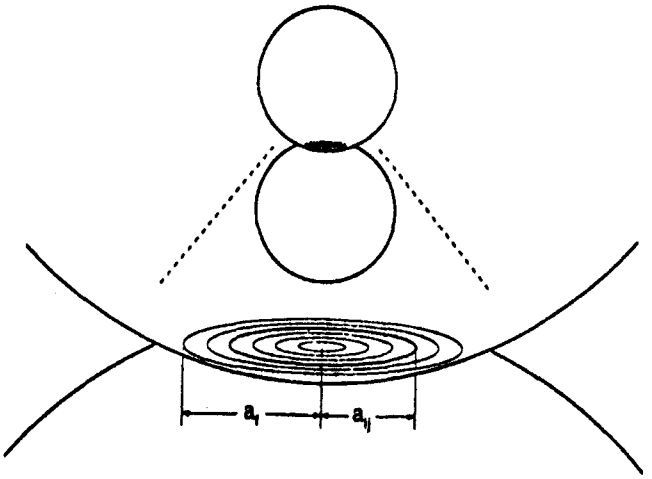

Fig. 1—Computational modeling of the heterogeneous deformation taking place in the contact area is accomplished by dividing this area into 100 concentric annuli. Within a given annulus (*e.g.* the annulus having the outer radius a_{ij}), testing for welding and fracture events is done. The outermost annulus has a radius equal to the plastic contact zone radius.

$$\Omega = \frac{4\pi}{3}\left(f_A R_{PA}^3 + f_B R_{PB}^3 + f_C R_{PC}^3\right) \qquad [3]$$

with f_i being the fraction of species i and R_{Pi} their volume equivalent radius. V_c is given by

$$V_c = \pi f_p h_o a_f^2 \qquad [4]$$

where f_p is the volumetric packing factor (typically = 0.6 to 0.7), h_o the powder thickness coating the balls, and a_f the radius of the plastic contact zone (the shape of the affected powder volume is that of a cylinder). Thus, the total number of trapped particles is approximated by

$$\psi \cong 0.5\left(\frac{h_o a_f^2}{f_A R_{PA}^3 + f_B R_{PB}^3 + f_C R_{PC}^3}\right) \qquad [5]$$

A specific combination of particles may be welded only over some fraction of the contact area between balls (*cf.* part I). We are therefore only interested in the volume per collision over which a specified type of weld event can take place. This is determined in a computational analysis by dividing the total area over which plastic deformation occurs into 100 concentric annuli of equal thickness (Figure 1). We then test within each annulus for welding of an i particle (*i.e.*, A, B, or C) to a j particle (also A, B, or C) and replace a_f with $W_{ij}a_f/100$, where W is the number of the outermost annulus in which welding can take place and ij designates the combination considered. We then have

$$\psi_T = \psi\left(\frac{W_{ij}}{100}\right)^2 \qquad [6]$$

Equation [6] represents the total number of particles found in the volume over which welding of the specified type may occur. Although each particle may be in contact with several others, we have assumed a particle may participate in only one weld event per impact. While it may be possible for a particle to weld to more than one other particle during a collision, any counting error introduced will be of order unity, an "error" that—given the complexity of the process and some of the assumptions we have used in modeling it—we consider acceptable. This simplification results in the number of weldable contacts being half the number of particles given by Eq. [6]. The number of contacts which are of the proper species combination is

$$\psi_{ij} = 0.5\psi_T P_{ij} \qquad [7]$$

where $P_{ij} = f_i^2$ for $i = j$ and $P_{ij} = 2f_i f_j$ for $i \neq j$.

Fracture events are handled similarly. We assume each particle may experience only one fracture event per collision; as in our treatment of weld events, this may introduce some counting error. The treatment of fracture is algebraically simpler, since the fracture of a particle is not influenced by the species with which it may be in contact. The number of fracture events of a species during impact is given by

$$\Phi_i = \psi\left(\frac{F_i}{100}\right)^2 f_i \qquad [8]$$

where F is the number of the outermost annulus in which fracture can occur and the subscript i denotes the species (A, B, or C).

We are now in a position to "count" the number of particles of each species present after a collision and to determine their number fraction. The following convention is used. When two particles of the same species weld, the new, larger particle is of the same species; when two particles of different species weld, they form a composite particle (species C). (Obviously, species C particles may have a range of compositions based on what combination of particles weld to form the C particle; this does not affect the count.) And when any particle fractures, it forms two smaller particles of the same species. (This also holds for composite particles, which are assumed not to fracture into "constituent" particles.) Using this convention, we can determine the following:

number of A particles reduced by welding $= \psi_{AA} + \psi_{AB} + \psi_{AC}$ [9a]
number of A particles gained by fracture $= \Phi_A$ [9b]
number of B particles reduced by welding $= \psi_{BB} + \psi_{AB} + \psi_{BC}$ [9c]
number of B particles gained by fracture $= \Phi_B$ [9d]
number of C particles reduced by welding $= \psi_{CC}$ [9e]
number of C particles gained by welding $= \psi_{AB}$ [9f]
number of C particles gained by fracture $= \Phi_C$ [9g]

Note that while we assume a particle may only fracture or weld once in a specific impact, we allow the possibility of a particle undergoing both processes in a single collision.

It is now simple to determine the number and number fractions of each species following a collision. Prior to coalescence/fragmentation during a specific collision, the number of particles of species i is given by

$$N_o^i = \psi f_i \qquad [10]$$

where the subscript o denotes the pre-event condition. Following the collision, the final counts for the respective species are

$$N_f^A = N_o^A - \psi_{AA} - \psi_{AB} - \psi_{AC} + \Phi_A \qquad [11a]$$

$$N_f^B = N_o^B - \psi_{BB} - \psi_{AB} - \psi_{BC} + \Phi_B \qquad [11b]$$

$$N_f^C = N_o^C - \psi_{CC} + \psi_{AB} + \Phi_C \qquad [11c]$$

Calculation of the respective species number fractions follows directly.

III. CHARACTERISTIC IMPACT FREQUENCY

Having explored the consequences of a single impact, it is now appropriate to determine an impact frequency, as this factor clearly influences the time required for processing. In particular, we seek an impact frequency that can be used to characterize a specific device. In other articles,[2,3,4] we explore how the velocity and frequency of impacts may vary between mills (and even within a given mill). Such "global modeling" is devoted to better understanding the action of the milling media and to demonstrating a heterogeneity of milling action too complex for incorporation into our "local" program. Thus, while powder particles are subjected to impacts over a spectrum of velocities and frequencies, for simplification in modeling, it is desirable to replace this spectrum with an equivalent "characteristic" impact. The impact frequency we determine in this section, then, is that of an average or characteristic impact to which a powder particle might be repeatedly subjected. The impacts referred to here are these "imaginary," characteristic impacts.

The number of impacts a particle experiences is distinctly different from the number the grinding media experience, although the two are clearly related. The powder volume affected by a collision (V_c, Eq. [4]) is only a small fraction of the powder "associated" with a ball, so it will take several impacts by a given ball before all particles associated with it have been struck once. The number of impacts required for each particle to be struck once (on the average) is given by V_p/V_c, where V_p is the volume of powder associated with each ball:

$$V_p = \frac{4\pi}{3}\left(\frac{R_b^3 \rho_B}{\rho_p CR}\right) \qquad [12]$$

where R_b is the ball radius, ρ_B the ball density, CR the charge ratio (mass of balls/mass of powder), and ρ_p the powder density. Using Eq [3] (with the expression for the radius of the contact zone from part I), we have

$$\frac{V_p}{V_c} = \frac{4}{\sqrt{3}}\left(\frac{R_b \rho_B^{1/2} H_v^{1/2}}{\rho_p CR h_o v}\right) \qquad [13]$$

where H_v is the powder hardness and v the relative velocity of the balls at impact. Using typical values for the variables in Eq. [13] (steel balls of 6.35×10^{-3} m diameter and density of 7800 kg/m^3, Al powder of density 2700 kg/m^3 and hardness of 400 MPa, CR = 10, coating thickness (h_o) = 10^{-4} m, and impact velocity = 4 m/s) V_p/V_c is approximately 2000 in a SPEX (SPEX Industries, Edison, NJ) mill. That is, a ball must participate in 2000 such characteristic impacts before we would anticipate that each powder particle associated with it is plastically deformed once. This number is even higher in an attritor with its lower characteristic collision velocity.[3]

To put these numbers in the perspective of an experiment, Davis *et al.*[5] determined that in a SPEX mill, containing 15 balls of 7.9-mm diameter, there were roughly 2000 (ball-ball and ball-vial) collisions per second. If we assume all collisions are of the ball-ball type, then each ball has roughly 250 impacts per second (two balls are involved in any impact). However, only 0.4 pct of these impacts are within 10 deg of a direct (head-on) collision. These are the impacts associated with the vast majority of fragmentation/coalescence events; other than normal impacts are relatively ineffective for such purposes.[2,4] Thus, a powder particle might experience a significant impact (*i.e.*, one associated with powder welding, fragmentation, or appreciable plastic deformation) once every 250 seconds. In program applications, we have used reasoning like this to estimate an effective (*i.e.*, characteristic) collision frequency.

The time (in minutes) between impacts involving an "average" powder particle is

$$\tau = \left(\frac{1}{60\ \Gamma}\right)\frac{V_p}{V_c} \qquad [14]$$

where Γ is the impact frequency (in s^{-1}). Note that as the powder hardens, the quantity of powder deformed in each subsequent collision is reduced, and more impacts by the ball are required to ensure that all particles affiliated with that ball are affected by a collision. The total processing time required for each particle to be subjected to, say, N impacts is given by the sum of Eq. [14]; *i.e.*, $\tau(1) + \tau(2) + \ldots \tau(N)$. In the program applications described in part III, processing time is calculated as described here.

IV. HOW THE PROGRAMS WORK

The programs use a representative volume element (RVE) of powder that is subjected to repeated collisions. This RVE is comprised of a set of particles impacted as a set only, though geometrical positioning of the particles within the set is random. There is no migration of powder in or out of the RVE.* The particle set is assumed to be a representative sample of powder in the mill; there are approximately V_p/V_c such sets associated with each ball. Although the RVE is a small fraction of the powder in a mill, we suggest it would correspond well to a sample which might be taken for actual measurements.**

*This assumption is clearly unrealistic. However, after some period of processing, it is reasonable to assume that for every powder particle migrating out of the element, an equivalent particle migrates into it. Mirroring the migration that must take place would require a substantial increase in programming complexity, *e.g.*, Monte Carlo simulation, but would not contribute proportionally to our understanding.

**The RVE has an approximate volume V_c ($=\pi h_o a_f^2$, Eq. [4]). Thus, the number of particles in the RVE is on the order of V_c divided by the average powder particle volume. In numerical applications described in part III, h_o is taken as 100 μm. The contact radius, a_f, depends upon several factors; *e.g.*, it scales with R_b and v and decreases with H_v. However, some idea of the number of particles in the RVE can be obtained. As discussed in Reference 1, for a collision between balls of radius 0.24 cm impacting at a velocity of 3.9 m/s, a_f = 177 μm. With h_o = 100 μm and f_p = 0.7, $V_c = 6.9 \times 10^{-12}$ m^3. For a particle size of 50 μm, the number of particles in V_c is 13. With a particle size of 10 μm, the number of particles in V_c is 1650. We note, therefore, that the number of particles in V_c is strongly dependent on initial particle size as well as process characteristics and material hardness.[1]

Prior to the first impact, the RVE consists of powders of uniform size and hardness (for a given species). The pro-

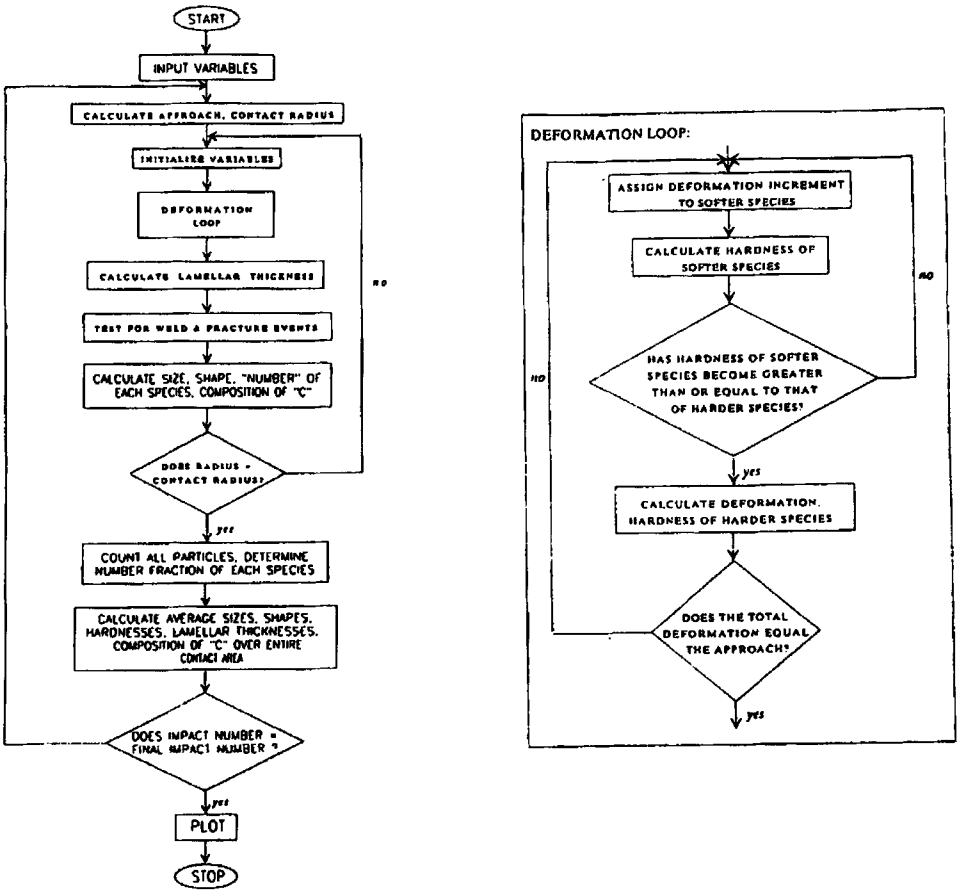

Fig. 2—The computational logic employed in MAP2. The diagram on the left represents the main program. The logic associated with the deformation loop of the main program is given on the right.

grams simulate the development of deformation as a function of radial position in the effective contact area of the colliding balls. If one species is softer than the other, the softer species deforms and work hardens to the hardness of the harder species prior to the latter deforming. The program then incrementally deforms the softer species, calculates a new hardness, and deforms the harder species to match the new hardness of the previously softer species. At the conclusion of each collision, the program tests for weld and fracture events and these are counted. The positions in which they occur are noted and the powder sizes and shapes redetermined. For example, when a welding or fracture event takes place, the number of each species is adjusted accordingly. If the "new" particle shape factor* exceeds one, it is assumed particle orientation changes for the next impact. In this case, particles are assigned a shape factor the inverse of that attained through events happening during the previous impact. After the program has completed this process for all radial positions, it determines average properties and dimensions for each species over the entire contact area. Each species is assigned a new average size,** shape, and hardness; the thickness of the lamellae (a measure of the degree of deformation) and the number of each species are calculated. These new properties and dimensions characterize the RVE for the subsequent collision. The process is repeated for the stipulated number of impacts, resulting in predictions of powder properties, shapes, and dimensions as a function of impact number or processing time.

*Particles are assumed to have shapes of ellipsoids of revolution (see Part I); f_s is the ratio of the ellipsoid minor to major axes.

**The particle size computed is a spherical volume equivalent radius; see Eq. [3] and Part I.

Two programs (MAP1 and MAP2) have been developed. MAP2 is more sophisticated. It considers two ductile species being welded to their own species and to each other (to produce a third, composite species) and welding of the composite species to itself and to the elemental species. Fracture of the three species is also considered. MAP1 is pertinent primarily to milling of single elemental species. However, it has the option of adding dispersoids to the powder charge. A flow chart, outlining program operation for MAP2, is shown in Figure 2. Appendices A and B provide a summary and further details of the program function.

V. SUMMARY

We have developed a schema that allows the physical descriptions derived in part I of this series to be incorporated into a computational algorithm which simulates the temporal evolution of powder in MA and MM. Such a program allows for comparison of predictions to observations, or for "thought experiments," in which the projected outcomes of altering process variables and material properties are explored. In part III, we apply the programs to previous MA studies for the purposes of discerning their predictive

capabilities and for exploration of the effect of material properties on the outcome of milling. Suggestions for program improvement and for empirical adjustments to the model are also provided there. In another article,[4] we use MAP2 in a different setting, one pertaining to global modeling wherein the effects of milling parameters on powder evolution are investigated.

APPENDIX A

This appendix provides brief summaries of how the programs function.

Size: When a particle fractures, its volume is halved. When two particles weld, their volumes are summed. A volume equivalent diameter is calculated for fractured or welded particles.

Shape: Particles remain oblate spheroids during processing, although their size and shape may change. Deformation reduces the shape factor; fracture doubles it. When two particles weld, their minor axes are summed; the larger of their major axes becomes the new major axis. Weld and fracture effects are incorporated after deformation is accounted for during an impact. If, after all the shape changes of transpired events have been accounted for, f_s for a particle exceeds one, the inverse is taken as f_s for the following impact.

Hardness: Strain is determined and hardness subsequently calculated through a (power-law) constitutive equation.

Lamellar thickness: The starting lamellar thickness is taken as the particle minor axis; it is adjusted by the cumulative strain the powder experiences.

Number fraction: The number of starting A and B particles is determined from initial weight fractions and sizes and material densities. Counting of particles and determination of number fractions is done as described in Section II.

The preceding procedures are followed for each annulus of contact. Results are totaled and averaged over the contact area. Values of the various parameters following a collision are used as input for the next collision.

Time: The number of impacts per ball needed to involve every particle associated with that ball in an effective collision is determined (Eq. [13]). Based on impact frequency, the interval needed for this number of ball collisions is calculated. The sum of all such intervals equals the elapsed time.

APPENDIX B
Data needed for programs

We summarize here the input data required to use the programs. (A protocol for obtaining these is provided in the Appendix of part III.) The input data for the programs are in commonly used units; the program automatically converts to SI units for all calculations. For example, inputting ball diameter in millimeters and coating thickness in micrometers is called for; the programs use these lengths in meters. The information required and the input units are listed below.

Media properties: Ball diameter (millimeters) and density (g/cm^3).

Process characteristics: Impact velocity (m/s), angle (in degrees, usually 0), number, frequency (s^{-1}), charge ratio, and powder coating thickness (μm).

Powder properties: Elastic modulus (GPa), tensile strength (MPa), fracture toughness (MPa$\sqrt{m}$), density (g/cm^3), fracture strain, values of strength coefficient (MPa), work-hardening coefficient in plastic constitutive equations, initial powder diameter (μm), shape factor, and starting hardness (Vickers, kg/mm^2).

Dispersoid properties: Dispersoid diameter (μm), density (g/cm^3), hardness (Vickers, kg/mm^2), and the ratio of dispersoid mass to powder mass.

Notes: Dispersoids can be considered in MAP1. Values of powder properties for both species must, of course, be specified in MAP2. In addition, the weight fraction for particle A is required as an input parameter.

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