

3-DIMENSIONAL STRUCTURE-IMITATION MODEL OF EVOLUTION OF MICROSTRUCTURE OF POWDER BODY DURING SINTERING

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This paper describes a 3D structure-imitation computer model of evolution of the powder compact during sintering and recrystallization without nucleation. At the initial stages of the evolution processes (sintering until a mosaic structure of boundaries is formed) the model particles are spheres, and two-particle interaction laws control their evolution. During sintering the degree of mutual penetration of the particles increases, regions where spherical particles are wholly faceted by contacts with neighboring particles are formed and grow. These particles are described using the formalism of Voronoi radical polyhedra, and grain growth laws govern their evolution. The model predicts the time dependencies of the following structure parameters of the polyhedra: average polyhedron size and dispersion, total surface of the facets of the polyhedra and total length of the edges of the polyhedra.

Keywords: Structure-imitation model; Sintering; Recrystallization;
Voronoi radical polyhedra

INTRODUCTION

Analyzing the physical phenomena that occur during sintering in the powder body, it turns out that the evolution of the microstructure plays the main role (Geguzin, 1984; Skorokhod and Solonin, 1984; Kadushnikov and Beketov, 1989; Ross *et al.*, 1982). The existing models of microstructural changes can be reduced to two main types, which

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are based on pair-interactions and on the evolution of the topological structure. For the first case an advanced apparatus to describe the kinetics of the process through a phenomenological equation of the sintering theory has been developed (Geguzin, 1984). Within the framework of this approach the elementary sintering models of the system consisting of equisized spheres are investigated. In the second case model approximations of the microstructure of the material by sets of polyhedra are constructed. In agreement with some main principles (maximum entropy, equilibrium of dihedral angles, etc.) imitation models of grain structure evolution are investigated. In all cases, simulations are performed for separate stages of the entire sintering process.

The authors have undertaken the approach to construct a uniform concept of simulation of the microstructure evolution during all stages of the sintering process (Kadushnikov and Beketov, 1989; Kadushnikov and Alievsky, 1989; Kadushnikov *et al.*, 1991a,b,c; 1998). The sintering process is described by 4 main stages. Each stage is characterized by a different geometrical model of the microstructure (Table I).

The first 2 stages correspond to the initial stages of the sintering process. A geometrical model of the microstructure is a set of spheres.

The 3rd stage corresponds to the late stage of the evolution of the sintered bodies, when the degree of mutual penetration of particles is high enough and the quantity of completely closed particles becomes significant. The set of spheres is still used for mathematical simulation of the sintering process. However, the skeletal polyhedral and triangulation models (corresponding to the configurations of spheres) are being

TABLE I Stages of sintering and geometrical models of the microstructure

<i>Stage</i>	<i>Description of the model</i>	<i>Geometry</i>	<i>Comments</i>
1	Building the close packing of spheres	Spheres	Generates an initial configuration of spheres for Stage 2
2	Evolution of the packing of spheres	Spheres	Initial stage of the sintering: mutual penetration of the spheres is low
3	Simulation using "skeletal" models	Spheres and polyhedra	Mutual penetration of the spheres is high
4	Simulation of recrystallization and normal grain growth	Polyhedra	The sintered bodies occupy the whole space and are described by polyhedra

built during the sintering process. Here *skeletal polyhedral model* is based on the 3D Voronoi diagram built on the given set of spheres, *skeletal triangulation model* is based on the 3D Delaunay triangulation corresponding to this Voronoi diagram (Preparata and Shamos, 1985). These models describe the real physical processes better than the set of spheres at the stage 3. Terms corresponding to the development of grain structures in the phase of normal grain growth are added to equations of the dynamics of the sintering spheres.

The 4th stage corresponds to the final phase of the sintering process. The set of spheres is not used now. Instead, the skeletal models built on the last step of the stage 3 become the base geometrical models. Following evolution of the system is determined by the dynamics of the skeletal models. This stage corresponds to recrystallization processes.

In this paper, we give a brief description of the algorithms used to simulate all stages of the sintering process.

DESCRIPTION OF THE MODEL

Stage 1: Building the Close Packing of Spheres

To build a close packing of spheres with disperse sizes within a bunker with flat walls we chose an algorithm named “rolling algorithm”. The physical idea of this algorithm is formulated as follows. The spheres are generated according to the chosen law of size distribution and are dropped to the bunker either from one point, or from randomly chosen positions. As soon as the dropped sphere encounters an obstacle – the bunker wall or an already packed sphere – it sticks to it (without impact) and begins to slide on its surface in the direction of the minimum of the potential energy to the following obstacle. (This direction is a projection of the free fall direction on the surface of the obstacle.) The movement of the sphere stops, obviously, at a point of intersection of 3 surfaces (3 spheres, 2 spheres and 1 plane and etc.) or on a surface situated perpendicular to the direction of the free fall of the sphere (for example, on the bottom of the bunker).

It should be noticed that the movement of the current packed sphere is completely described by the trajectory of its center. In this case, the sliding of the sphere with center O and radius R on the surface of another sphere with center O_1 and radius R_1 corresponds to sliding of

the center O on the surface of a sphere with center O_1 and increased radius $R_1 + R$. Similarly, the sliding of the sphere with center O and radius R on the plane

$$\mathbf{ax} = b, \quad (1)$$

$\mathbf{a} = (a_x, a_y, a_z)$, $a > 0$, corresponds to sliding of the center O on a shifted plane

$$\mathbf{ax} = b + |\mathbf{a}|R. \quad (2)$$

Thus, it is possible to formulate the following algorithm of movement of the next packed sphere:

- (1) Choose radius R of the packed sphere and an initial position of its center O . Initialize the set of restrictions: no restrictions.
- (2) Analyze the current set of restrictions: the system of 0, 1, 2 or 3 inequalities describing positions in the space which are disabled for the center of the packed sphere. Choose the new direction of movement of the sphere center. The movement of the center along this direction must satisfy the restrictions, this direction must form the minimal allowable angle with the direction of the free fall of the sphere (vector $(0, 0, -1)$), and this angle must be less than $\pi/2$. In absence of allowable directions, i.e. when all angles are more than $\pi/2$, stop and choose the next sphere for package.
- (3) Determine the next segment of the movement trajectory of the center. It is a direct line or an arc segment. Perhaps, remove one of the restrictions.
- (4) Calculate points of intersection of the current segment of the trajectory with all packed spheres (with radii increased by R) or bunker walls (shifted by a normal vector with length R). Select the nearest to the beginning of the segment. Add one new restriction: all points inside the sphere or behind the wall (which intersects the current segment at the nearest position) are disabled for the center of the packed sphere. Return to step 2.

It is easy to see that the described algorithm depends quadratically on the number of packed spheres. To convert it to a nearly-linear algorithm, the special technique of grid hashing was used. This technique will be outlined in the following.

Let us consider a cubic grid with the cell size comparable with sizes of spheres. For each cell, let us build the list of packed spheres the centers of which lie in this cell. Then analysis of spheres disposed in several grid cells around the trajectory allows to find all intersections of the trajectory segment and the packed spheres. So, when the parameters of the grid are chosen well, the algorithm becomes almost linear with a multiplier depending on the parameters of the whole packing of spheres (such as dispersion of radii of the spheres, etc.).

Stage 2: Evolution of the Packing of Spheres

The constructed close packing of spheres represents the initial data for the second stage – simulation of the evolution of the spheres during sintering. For deriving the equations of movement and dynamics of the sizes of the sintered body, the main incentive was to satisfy the equations of kinetics of pair sintering for each pair of contacted spheres in the system.

Let us consider sintering of a system consisting of N spheres: $S_i(R_i, \mathbf{r}_i, \mathbf{v}_i)$, $i = 1, 2, \dots, N$; R_i – radii of the spheres, \mathbf{r}_i – coordinates of their centers, \mathbf{v}_i – speed of movement.

To describe the free movement of the spheres, the well-known equations of Newton are used:

$$\begin{cases} \dot{\mathbf{r}}_i = \mathbf{v}_i, \\ m_i \dot{\mathbf{v}}_i = \mathbf{F}_i. \end{cases} \quad (3)$$

Here $m_i = \frac{4}{3}\pi\rho R_i^3$, ρ – density of the material of the spheres, \mathbf{F}_i – sum of external forces, acting on each sphere, such as the force of gravity ($m_i\mathbf{g}$) or force of liquid friction ($-\nu\mathbf{v}_i|S|_i$, $|S|_i$ – area of free surface of i th sphere).

At existence or occurrence of contact:

$$|\mathbf{r}_{ij}| = |\mathbf{r}_i - \mathbf{r}_j| < R_i + R_j, \quad (4)$$

it is necessary to take into account the interaction of the contacted spheres, resulting in sintering. Let us record the equations of the kinetics of pair sintering for all pairs of incident (contacting) spheres in the

following form:

$$\mathbf{v}_{ij} = \dot{\mathbf{r}}_{ij} = f(|\mathbf{r}_{ij}|)\mathbf{r}_{ij}, \quad i = 1, 2, \dots, N; \quad j \rightarrow i; \quad (5)$$

here and below $j \rightarrow i$ means that the spheres i and j are contacted – see (4).

And let us construct the deviation functional:

$$Q = \sum_{i,j:j \rightarrow i} |\mathbf{v}_{ij} - f(|\mathbf{r}_{ij}|)\mathbf{r}_{ij}|^2. \quad (6)$$

Let us modify the trajectory of movement of the corresponding point for an ensemble of spheres in the phase space so that it comes nearer to a minimum of the deviation functional Q . To do it, let us add the anti-gradient Q with a weight multiplier α in the right parts of Eq. (3), where the multiplier α determines the degree of influence of sintering on movement of the system of spheres (or vice versa):

$$\begin{cases} \dot{\mathbf{r}}_i = \mathbf{v}_i - \alpha \sum_{j:j \rightarrow i} (\mathbf{v}_{ij} - f(|\mathbf{r}_{ij}|)\mathbf{r}_{ij})(f(|\mathbf{r}_{ij}|) + f'(|\mathbf{r}_{ij}|)|\mathbf{r}_{ij}|), \\ \dot{\mathbf{v}}_i = \frac{\mathbf{F}_i}{m_i} - \alpha \sum_{j:j \rightarrow i} (\mathbf{v}_{ij} - f(|\mathbf{r}_{ij}|)\mathbf{r}_{ij}). \end{cases} \quad (7)$$

The conservation of weight means that during movement and sintering the volume V of the geometrical union of spheres remains constant:

$$\dot{V} = \sum_i \left(\frac{\partial V}{\partial \mathbf{r}_i} \dot{\mathbf{r}}_i + \frac{\partial V}{\partial R_i} \dot{R}_i \right) = 0. \quad (8)$$

Let us balance the change in volume V of the union of spheres connected with the movements of some sphere S_i with the change in the radius R_i of this sphere:

$$\frac{\partial V}{\partial \mathbf{r}_i} \dot{\mathbf{r}}_i + \frac{\partial V}{\partial R_i} \dot{R}_i = 0 \quad (9)$$

for each i .

Thus, we obtain the following equations of dynamics of the radii of the spheres:

$$\dot{R}_i = |\dot{\mathbf{r}}_i| \Xi_i / |S_i|. \quad (10)$$

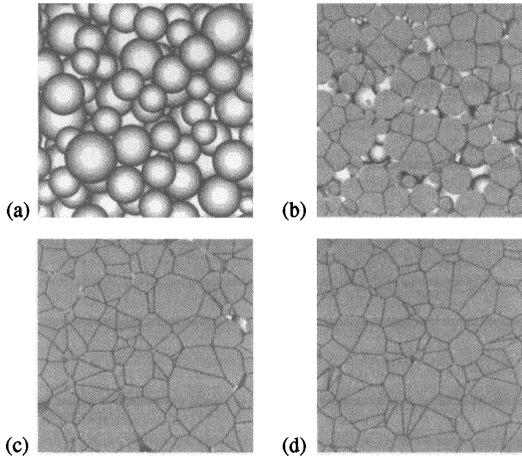


FIGURE 1 View of initial spheres package (a), and sections of model during evolution (b) $t = 50$, (c) $t = 80$, and (d) $t = 110$ time steps.

Here $|S_i| = \iint_{\Phi_i} ds = (\partial V)/(\partial R_i)$ – area of the free surface Φ_i of the sphere S_i , $\Xi_i = \iint_{\Psi_i} (\dot{\mathbf{r}}_i/|\dot{\mathbf{r}}_i|) \mathbf{n} ds$ can be interpreted as an oriented area of the projection of occupied surface Ψ_i of the sphere S_i on the plane perpendicular to its velocity (\mathbf{n} is the unit normal vector to the surface).

The integration of the systems of Eqs. (7), (10) is carried out by the Euler method with fixed time step (Hockney and Eastwood, 1981). Areas $|S_i|$, and Ξ_i , are calculated numerically. The packing of 1000 spheres with distributions of radii $N(1.0, 0.3)$ in a cubic bunker and sections of the model during the evolution process are shown in Fig. 1.

About Skeletal Models

To understand the algorithm used to simulate the sintering process at the stages 3 and 4, it is necessary to describe the concept of *skeletal models* and some connected terms.

We consider 2 kinds of skeletal models based on *Voronoi polyhedra* and *3D Delaunay triangulation*. These geometrical concepts are some generalization of the classic concepts described, for example, in Preparata and Shamos, 1985. These models allow to transit naturally from the set of spheres to polyhedral description of the system.

The basic concepts of our skeletal models will be outlined in the following.

Let there are the set of N spheres: $S_i(R_i, \mathbf{r}_i)$, $i = 1, 2, \dots, N$; R_i – radii of the spheres, \mathbf{r}_i – radius-vectors of their centers. We define the *Voronoi polyhedron* P_i as a set of points

$$\{\mathbf{r} \in \mathbb{R}^3: |\mathbf{r} - \mathbf{r}_i|^2 - R_i^2 \leq |\mathbf{r} - \mathbf{r}_j|^2 - R_j^2 \text{ for all } j \neq i\} \quad (11)$$

(it is equivalent to the classic definition when all $R_i = 0$). Voronoi polyhedra occupy the whole space \mathbb{R}^3 . Adjacent pairs of polyhedra P_i, P_j are separated from each other by *radical planes* of pairs of the corresponding spheres S_i, S_j ;

$$\{\mathbf{r} \in \mathbb{R}^3: |\mathbf{r} - \mathbf{r}_i|^2 - R_i^2 = |\mathbf{r} - \mathbf{r}_j|^2 - R_j^2\} \quad (12)$$

(when the spheres S_i and S_j are intersected, their radical plane contains the intersection circumference). So, we also call the Voronoi polyhedra “*radical polyhedra*”. We also will consider *radical axis* of 3 spheres S_i, S_j, S_k :

$$\{\mathbf{r} \in \mathbb{R}^3: |\mathbf{r} - \mathbf{r}_i|^2 - R_i^2 = |\mathbf{r} - \mathbf{r}_j|^2 - R_j^2 = |\mathbf{r} - \mathbf{r}_k|^2 - R_k^2\} \quad (13)$$

and *radical center* of 4 spheres S_i, S_j, S_k, S_l : the point \mathbf{r} determined by the condition

$$|\mathbf{r} - \mathbf{r}_i|^2 - R_i^2 = |\mathbf{r} - \mathbf{r}_j|^2 - R_j^2 = |\mathbf{r} - \mathbf{r}_k|^2 - R_k^2 = |\mathbf{r} - \mathbf{r}_l|^2 - R_l^2. \quad (14)$$

Delaunay triangulation is defined as the partition of the convex hull of the set of centers of all spheres S_i into tetrahedra, such as:

- vertices of the tetrahedra are all centers \mathbf{r}_i ;
- there is an edge of some tetrahedra connecting the vertices \mathbf{r}_i and \mathbf{r}_j if (and only if) the radical polyhedra P_i, P_j are adjacent (have a common facet);
- there is a facet of some tetrahedra with the vertices $\mathbf{r}_i, \mathbf{r}_j$ and \mathbf{r}_k if (and only if) the radical polyhedra P_i, P_j, P_k are adjacent (have a common edge);
- there is a tetrahedron with the vertices $\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k$ and \mathbf{r}_l if (and only if) the radical polyhedra P_i, P_j, P_k, P_l are adjacent (have a common vertex).

Our *skeletal triangulation model* is obtained from the Delaunay triangulation by removal of edges corresponding to not contacted spheres

$(|\mathbf{r}_i - \mathbf{r}_j|^2 > (R_i + R_j)^2)$ and removal of all facets and tetrahedra containing these edges. Similarly, our *skeletal polyhedral model* is obtained from the Voronoi polyhedra by removal of the corresponding facets, edges and vertices.

The duality of the triangulation and polyhedral skeletal models is convenient for description of grains by the radical polyhedra and pores by tetrahedra of the triangulation.

It is assumed that the radical plane of each pair of spheres separates their centers. This property is kept during the evolution of the sintering of the spheres.

Both skeletal triangulation model and skeletal polyhedral model can be trivially built if we have already built the Delaunay triangulation. So, we give a brief description of an algorithm of building Delaunay triangulation in the following.

This algorithm is iterative. Let some tetrahedra of the triangulation be already built. The algorithm chooses any external facet Φ of some tetrahedron T_m ("external" means that it is not a facet of any other tetrahedron) so that Φ does not belong to the convex hull, and builds a new tetrahedron T of the triangulation on the facet Φ (the second tetrahedron incident with this facet). Vertices of the facet Φ are centers of 3 spheres S_i, S_j and S_k . Building the tetrahedron T means selecting the fourth sphere S_l the center of which will be the fourth vertex of T . The algorithm selects the sphere S_l from all spheres lying "above" the plane of the facet Φ (where "above" means "from the other side than T_m ") so that the position of a radical center of 4 spheres S_i, S_j, S_k, S_l is the most "lower". Formally,

$$l = \operatorname{argmin}_{p, (\mathbf{r}_p - \mathbf{c}) \cdot \mathbf{n}_c > 0} \left(\frac{w_p - w_c}{2(\mathbf{r}_p - \mathbf{c}) \cdot \mathbf{n}_c} \right), \quad (15)$$

where \mathbf{c} is an intersection point of the plane of the facet Φ and a radical axis of spheres S_i, S_j, S_k ("radical center of the facet Φ "), \mathbf{n}_c the unit normal vector to the facet Φ directed "upward", $w_p = |\mathbf{c} - \mathbf{r}_p|^2 - R_p^2$, $w_c = |\mathbf{c} - \mathbf{r}_i|^2 - R_i^2$ ($= |\mathbf{c} - \mathbf{r}_j|^2 - R_j^2 = |\mathbf{c} - \mathbf{r}_k|^2 - R_k^2$).

After choosing the fourth vertex of the new tetrahedron T , new edges and facets are added to the triangulation. The process of building is finalized when all external facets of tetrahedra belong to the convex hull.

This algorithm, as well as the “rolling algorithm” from stage 1, can be made almost linear by means of the grid.

Stage 3: Simulation Using “Skeletal” Models

In the third stage of the simulation, the dynamics of the skeletal models is practically completely determined by movement and sintering of the spheres. Thus, it is necessary to continue the integration of the system of equations of dynamics of the sintering spheres, building the skeletal models repeatedly and calculating the target characteristics of the model. The influence of effects of normal grain growth is reflected only in the equations of sphere size dynamics – for spheres with completely closed (by neighbors) surface (not influencing on conservation of mass) the equation of dynamics of normal grain growth is used instead of Eq. (10):

$$\dot{R}_i = \lambda \sum_{j:j \rightarrow i} \left(\frac{1}{R_j} - \frac{1}{R_i} \right). \quad (16)$$

The plots in Fig. 2 show the time dependencies of the following characteristics of skeletal models:

- average grain size i.e. the diameter of a sphere, the volume of which equals the volume of the polyhedron;
- dispersion of polyhedron sizes;
- total area of the sides of the polyhedra;
- total length of the edges of the polyhedra.

Stage 4: Simulation of Recrystallization and Normal Grain Growth

The fourth stage is characterized by a change of the basic geometrical model – particles become polyhedra without spherical surface. The evolution of the model is determined by minimization of the redundant free energy of system:

$$A = \sigma S + \varepsilon L + \gamma V, \quad (17)$$

where σ , ε , γ denote the specific redundant free energies of facets, edges and vertices of the polyhedra and S , L , V denote the total surface of

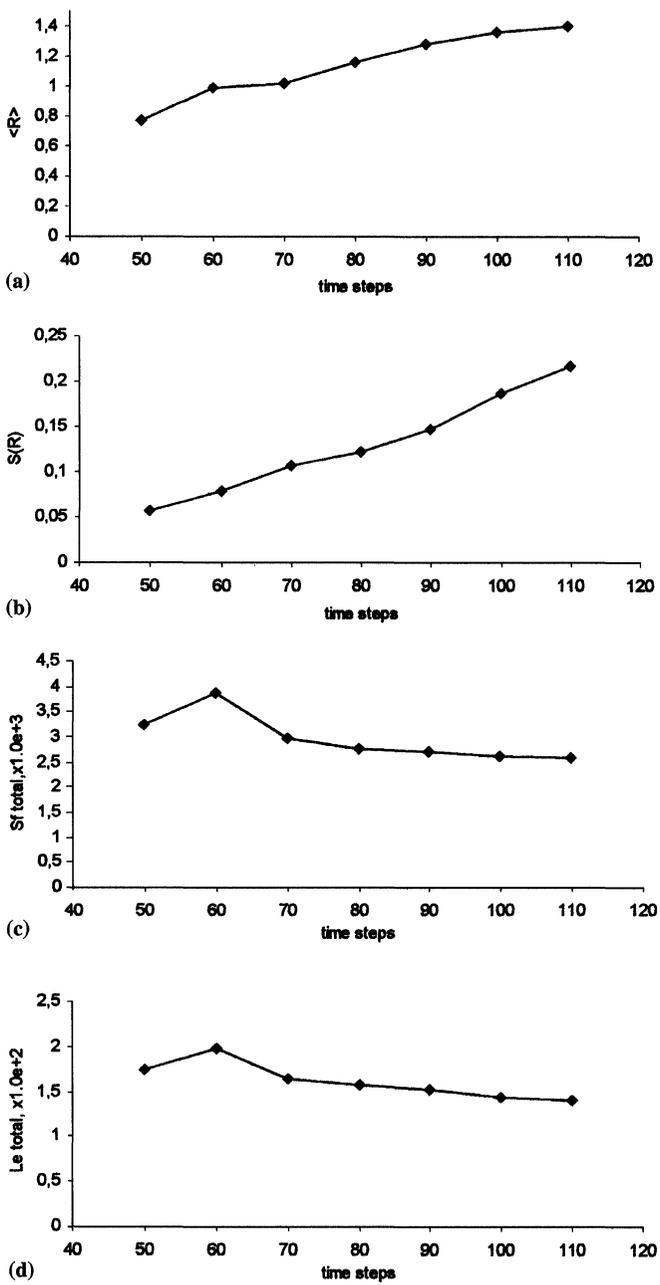


FIGURE 2 Time dependencies of average polyhedron size (a), polyhedrons' sizes dispersion (b), total surface of polyhedrons facets (c) and total length of polyhedrons' edges (d).

facets, edge lengths and vertices number, respectively. The model of Voronoi polyhedra was chosen as a model of particles. As independent variables used in the simulation equations we choose the coordinates and radii of the spheres determining the Voronoi diagram, i.e. $A = A(\mathbf{r}_i, R_i)$. Unlike the stages 2 and 3, here the set of spheres is used only in mathematical formalism and is not essential. The equations of movement of the spheres can now be written as follows:

$$\begin{cases} \dot{\mathbf{r}}_i = -\xi \frac{\partial A}{\partial \mathbf{r}_i}, \\ \dot{R}_i = -\psi \frac{\partial A}{\partial R_i}, \end{cases} \quad (18)$$

(ξ, ψ – factors of mobility). While calculating the right parts we consider all the edges of triangulation, that contain vertices \mathbf{r}_i , and then for each edge all the facets, that contain this edge. Each facet belongs to the pair of tetrahedra, whose radical centers are vertices of the radical polyhedra, forming the edge of polyhedron. Let us designate them by \mathbf{c}_+ and \mathbf{c}_- , let \mathbf{c}_j denote the center of the facet of the polyhedron, that corresponds to j th edge. The gradient of A is written as follows:

$$\begin{aligned} \frac{\partial A}{\partial \mathbf{r}_i} &= \sigma \sum_{e \supset \mathbf{r}_i} \sum_{f \supset e} \frac{\partial S_f}{\partial \mathbf{r}_i} + \varepsilon \sum_{e \supset \mathbf{r}_i} \sum_{f \supset e} \frac{\partial L_f}{\partial \mathbf{r}_i}, \\ \frac{\partial A}{\partial R_i} &= \sigma \sum_{e \supset \mathbf{r}_i} \sum_{f \supset e} \frac{\partial S_f}{\partial R_i} + \varepsilon \sum_{e \supset \mathbf{r}_i} \sum_{f \supset e} \frac{\partial L_f}{\partial R_i}, \end{aligned} \quad (19)$$

where

$$L_f = |\mathbf{c}_+ - \mathbf{c}_-|, \quad S_f = \frac{1}{2} (|\mathbf{c}_+ - \mathbf{c}_i|^2 |\mathbf{c}_- - \mathbf{c}_i|^2 - (\mathbf{c}_+ - \mathbf{c}_i, \mathbf{c}_- - \mathbf{c}_i)^2)^{1/2}. \quad (20)$$

The summation in (19) is performed over all the edges e containing the vertex \mathbf{r}_i and over all the facets f containing the edges e .

The equations of movement for spheres are integrated by Euler's method, with recalculation of skeletal models during each time step.

CONCLUSION

This paper describes the complete concept of simulation of the evolution of the microstructure of the powder compact during all stages of the sintering process. The four stages of the process are considered, the appropriate geometrical model is suggested for each stage. It was shown that the evolution of the system can be described by the equations of movement of the centers and radii of spheres at all the stages of the process. The original algorithms for building the packing of spheres and the skeletal triangulation are presented. The described algorithms for the first three stages are implemented in a computer program. This program is tested with models containing up to 1,000,000 spheres for packing and up to 30,000 spheres for triangulation. We used the personal computer with the Pentium 200 CPU and 64Mb RAM.

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