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Computer study of liquid phase sintering – three-dimensional time dependent rearrangement

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Abstract. The rearrangement process during liquid phase sintering has been generally accepted that driven by the capillary forces between solid grains embedded in liquid. This paper outlines a computer-based method for three-dimensional computer simulation of rearrangement during liquid phase sintering. The theoretical models dealing with the fundamental interaction forces that exist between grains attached by liquid bridges will be outlined and the development from these pair-wise interactions to multi-grain models will be described.

1. Introduction

During the initial stage of liquid phase sintering (LPS) rearrangement of the solid phase, in which surface tension forces act to bring about physical movement of the constituents of sintering body, takes place causing rapid densification. This process assumes that if there is good wetting between liquid and solid phases, solid grains can rearrange themselves under the action of surface tension forces, producing more stable packing. Therefore it is interesting to investigate how the solid grains rearrange [1-3].

An analytical solution for rearrangement during liquid phase sintering has never been attempted and may not be possible. Computer simulation has therefore played a key role in providing insight into rearrangement and its theoretical framework. When no external force is applied to the two spherical grains then the system rigid motion depends upon the driving force only. The driving force available to move two grains is the capillary force in the liquid bridge. However, topological constraints due to surrounding grains can also lead to grains rearrangement and the buildup moments on the grains. Three-dimensional (3-D) computer simulation method for treating this effect in large scale grain arrays will be discussed in the next.

2. Modelling the rearrangement

Let there is a mixture of two components: a major component that forms the particulate solid, and an additive phase as a liquid-producing component, in which the liquid phase will be formed when the mixed powders are heated to a certain temperature. In such system of grains connected by liquid bridges, several kinds of forces affect the grain motion: a capillary force within liquid bridge keeps the
grains approaching each other, while a viscous force resists their motion [4]. Other forces that can also affect considerably this motion, unfortunately, have not been still well described.

As the calculation cell, let us consider the two spherical grains of radius \( R_1 \) and \( R_2 \) (\( R_1 \geq R_2 \)) joined by a liquid bridge of constant volume providing their capillary interaction. The interparticle force between grains could be carried out by using the circle approximation in the form similar to the form given by Eremenko et al. [5], i.e.

\[
F = 2\pi \gamma R_1 \sin \phi_1 \sin(\phi_1 + \theta) + \pi \gamma (R_1 \sin \phi_1)^2 \left( \frac{1}{\rho_2} - \frac{1}{\rho_1} \right),
\]

where \( \gamma \) is the surface free energy of the liquid-vapor interface, \( \phi_1 \) and \( \phi_2 \) are inter-related angles [6] subtended by the contact at the center of the grains, \( \theta \) is the contact angle measured between the solid and liquid phases, and \( \rho_1 \) and \( \rho_2 \) are the radii of the meniscus [7].

**Figure 1.** Schematic description of interaction (solid arrows) between by bridges (gray colored) connected grains. (a) Capillary force field. (b) Displacement distance field.

Lee et al. [8] defined the total capillary force that acts on a grain as the sum of forces coming from its nearest neighbors. In this approach the total driving force in 3-D space for the model characterized by \( N \) solid grains and \( M \) liquid bridges will be computed by distributing all partial capillary forces between interfaces solid-liquid within each separate liquid bridge, i.e.

\[
F_i = \sum_{k=1}^{N} \sum_{j=1}^{n_k} F_{kj} = \sum_{k=1}^{N} \sum_{j=1}^{n_k} F_{kj},
\]

where \( F_k \) is the total driving force for \( k \)-th grain defined as a sum of partial capillary forces \( F_{kj} \) between \( k \)-th grain and \( j \)-th grain (equation (1)), and \( n_k \) is the ordinal numbers of nearest neighbors of \( k \)-th grain (figure 1a). Theoretical treatment of grain rearrangement must involve consideration of a network of capillary forces \( F_{kj} \).

Huppmann and Riegger [2] investigated the rearrangement process to show instantaneous shrinkage dependent on the mean capillary force per contact as \( \Delta D/D_o = (D_o - D)/D_o = \chi(F - F_o) \), where \( D \) is inter-grain distance, \( \chi \) is a constant at a given grain size and \( F_o \) represents an inherent resistance to grain rearrangement. This relation can be simply modified to serve as basic equation for 3-D simulation of grain motion induced by the total capillary force (2) during rearrangement.
Let $\Delta D_k^{n+1}$ is the change of the inter-grain distance of $kj$-th liquid bridge due to the capillary force $F_{kj}^{n+1}$ after time $t + \Delta t$ with direction of this force, and $D_k^n$ is the inter-grain distance at previous time step (figure 1b). The displacement of $k$-th grain in 3-D space (as vector distance) can be now defined as the total displacement of $k$-th grain that will be computed as the sum of corresponding vector distances, i.e.

$$\Delta D_k^{n+1} = \sum_{j=1}^{n_k} \Delta D_{kj}^{n+1} = \sum_{j=1}^{n_k} D_{kj}^n \chi (F_{kj}^{n+1} - F_o), \quad \|F_o\| = \min \left\{ \|F_{kj}^o\| \right\},$$

where the capillary force $F_{kj}^o$ will be computed according to initial inter-grain distances $\{D_{kj}^o\}$.

Although experimental and theoretical results of Hupmann and Riegger [2] strongly support above assumption that $F_o$ is caused by capillary forces, it could be extended toward liquid viscosity by incorporation the dynamic effects due to the viscosity of the liquid bridge. Now if $(x^k_c, y^k_c, z^k_c)$ is the center position of $k$-th grain before its displacement, then due to the action of total capillary force $F_k$ its position will be updated by sintering transformation

$$(x^k_c, y^k_c, z^k_c) \rightarrow (x^k_c + \Delta D_{k(x)}^{n+1}, y^k_c + \Delta D_{k(y)}^{n+1}, z^k_c + \Delta D_{k(z)}^{n+1}),$$

(3)

where the subscripts $(x)$, $(y)$ and $(z)$ indicate components of the vector $\Delta D_k^{n+1}$ along Cartesian coordinates. The rearrangement of selected $k$-th grain will be simulated by three-steps procedure: (a) computation of the displacement $\Delta D_k^{n+1}$, according to the capillary forces $\{F_{kj}\}$ within liquid bridges with its first neighbors; (b) relocation of $k$-th grain using transformation (3); (c) relocation of its first, second, etc. neighbors using the adjustment procedure: their relative relocations with no change of inter-grain distances, $\{\Delta D_{k(x)}^{n+1}, \Delta D_{k(y)}^{n+1}, \Delta D_{k(z)}^{n+1}\} = \{0,0,0\}$. Such rearrangement procedure will be applied to all grains starting from selected grain. The number of iterations is given in units of rearrangement step, RS. At 1 RS the displacement of all grains (chosen at random) induced by the mean capillary forces within liquid bridges, $\{F_k\}$, will be computed applying transformation (3). Thus the number of iterations will be incremented by $1/N$. In this approach RS will be considered a measure of time in arbitrary units, assuming that RS scales linearly with time by some constant.

### 3. Results and discussion

In order to avoid shrinkage by other processes than rearrangement, in our simulation the system W-Cu will be chosen since tungsten is practically insoluble in liquid copper [9]. The same system was investigated by Huppmann and Riegger [2]. They concluded that the rearrangement shrinkage is a function of the mean force per contact with $\chi \approx 100$ N$^{-1}$ for the average grain radius of $\sim 100$ µm. This value of $\chi$ we will take into account in our simulation. In all computer simulation the surface energy of copper $\gamma = 1.28$ N/m [10], the normalized liquid volume $V/V_o = 0.03$ ($V_o$ is the volume of the solid phase) and the contact angle of $10^\circ$ (good wetting) will be used.

In general, densification by rearrangement during LPS occurs very short time. In that sense, as an example for simulation of rearrangement, randomly generated (planar) multi-grain system with average radius of $\sim 55$ µm and average inter-grain distance of $\sim 10$ µm (figure 2a) will be used. The results of rearrangement after 130 RS and 250 RS are shown in figures 2b and 2c, where the average inter-grain distances were now $\sim 5.5$ µm and $\sim 3.5$ µm, respectively. It can be seen remarkable approaching of some grains combined with filling pores process, but also enlargement of some inter-grain distances followed by growing of inter-grain pores. The overall effect of separating the grains...
while keeping constant the liquid volume is therefore an elongation of the bridge, which becomes longer and thinner and less concave. It should be noted that, at greater separation distances, there is a maximum value of the separation which corresponds to liquid bridge rupture (the bridge is not stable any more and formation of liquid droplets on the surfaces of grains is preferable), which will be the subject of our future study. In practice, this enlargement combined with rupture of some liquid bridges can reinforce grain rearrangement, but also generate non-uniform distribution of small and large pores.

Figure 2. Snapshot of several interacting grains connected by liquid bridges ($N = 69, M = 120$).
(a) Initial structure. After 130 RS (b) and 250 RS (c). $\chi = 25 \text{ N}^{-1}$ and $V/V_0 = 0.03$. Light-gray colored are grains and dark-gray colored are liquid bridges.

Our theoretical investigation has shown that rearrangement is essentially governed by geometrical factors and nearest neighbor interaction (capillary forces between adjacent grains). For study the behavior of the multi-grain model, each grain and its neighborhood was treated as “elementary unit”. Grains were drawn together with increasing inter-grain forces as the separations between them were decreased. It can be concluded that the proposed simulation model correctly describe the rearrangement processes during LPS. The theoretical basis of this analysis is general and applicable to any multi-component metallic or ceramic system. The computed microstructures can be applied for investigation and prediction of microstructure evolution by rearrangement in LPS.

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References