Basic and Mixed Models for Computer Simulation of Liquid Phase Sintering of a Porous Structure

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Abstract:
A two-dimensional method based on basic and mixed models for simulation of liquid phase sintering of a porous structure will be developed. These models will be tested in order to conduct a study of diffusion phenomena and gravitational effects on microstructural evolution during liquid phase sintering of a W-Ni system.

Keywords: Liquid phase sintering, Porous structure, Computer simulation.

Introduction
Controlling microstructural evolution during liquid phase sintering (LPS) is sometimes a difficult problem because of the large number of parameters that must be understood and controlled. In recent years, a range of computer simulation models has been developed with the aim of simulating the detailed evolution of microstructure during grain growth. Coarsening models are the most numerous and most mature of the microstructural evolution models. Many investigators have used a number of different numerical techniques to simulate coarsening by processes such as grain growth and Ostwald ripening. Recently the results of a computer simulation of boundary migration during LPS have been reported [1]. This approach was further applied for a computer study of LPS under gravity and microgravity conditions [2,3]. A similar method was developed by introducing basic, extended and combined models [4]. However, all these studies ignored the effects of pores on grain growth and on grain coarsening assuming zero porosity. In this paper we will develop a two-dimensional (2-D) method based on basic and mixed models for simulation of gravity induced microstructural evolution of a porous structure during LPS.

Model topology
For 2-D computer simulation of LPS we will use a model of $N$ domains, instead of the very often used model of $N$ particles of a regular-circular shape. Such model system domains can be a good 2-D representation of a real system, since even during sintering of initially spherical particles most of them will no longer be circular because of the highly asymmetric diffusion field around and between them. A 2-D domain (Fig. 1) can be defined by its $n$ boundary points - sites located on the boundary contour or on the phase interface, i.e.,

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During LPS most domains have a rigid body motion as a result of a nonuniform concentration gradient over their surfaces. Even small changes in the domain location relative to one another can have a large effect on the resultant morphological model evolution. An assumption is made that domains during movement will not overlap. Thus the domain position after its motion, $D_{after}$, must be restricted to those parts of the experimental region not already occupied by other domains, i.e.

\[ D = \{(x_k, y_k) \mid k = 1, 2, ..., n\}. \]

Even complex domain motion can be represented by discrete domain translation defined by the next algorithm.

\[
\text{For } i = 1 \text{ To } N \\
\quad \text{If } D_{after}^i \cap D^j = \emptyset \text{ Then } D^i \mapsto D_{after}^i \\
\quad \text{Next } i
\]

Brownian motion model

Unbalanced random molecular forces induce Brownian motion. Using Einstein's treatment [5] for the mean displacement of a grain, the average time between domain contacts can be calculated as
\[
\tau_{\text{Brown}} = \frac{3\pi \langle r \rangle \eta \langle \lambda \rangle^2}{kT}
\]

where \( \langle r \rangle \) is the average domain radius, \( \eta \) is the liquid viscosity, \( \langle \lambda \rangle \) is the average separation distance between domains [4], \( k \) is the Boltzmann constant, and \( T \) is the sintering temperature.

Unbalanced random motion can be modeled by domain translation along positive or negative \( Ox \) axis or \( Oy \) axis. Let us assume that the experimental region is partitioned into subregions by a mesh with respective grid spacings \( \Delta x \) and \( \Delta y \) along two distance coordinates \( x \) and \( y \), respectively. Then random motion can be modeled by a domain translation algorithm along one of four possible random directions

\[
\begin{align*}
\text{For } i = 1 & \text{ To } N \\
D^i & = \{(x_k, y_k) \ (k = 1,2,\ldots,n_i)\} \\
D^i_{\text{after}} & = \{(x_k \pm \Delta x, y_k \pm \Delta y) \ (k = 1,2,\ldots,n_i)\} \\
\text{If } D^i_{\text{after}} \cap D^j & = \emptyset \text{ Then } D^i \mapsto D^i_{\text{after}} \\
& \quad j=1,2,\ldots,N; i \neq j \\
\text{Next } i
\end{align*}
\]

in which the sign \( \pm \) for each separate domain will be determined at random.

**Stokes’ law settling**

Systems with a large density difference between the liquid and solid (\( \rho_s - \rho_L \)) are characterized by macro- and micro-structural effects associated with gravity. This is a particular problem for tungsten heavy alloys.

Under Earth-based experimental conditions Stokes’s law settling usually dominates microstructure formation [6]. The settling time for solid domains to travel the average separation distance \( \langle \lambda \rangle \) in a liquid matrix can be calculated as

\[
\tau_{\text{sett}} = \frac{\langle \lambda \rangle}{v_{\text{sett}}} = \frac{2g \langle r \rangle^2 (\rho_s - \rho_L)}{9\eta},
\]

where \( g \) is the gravitational acceleration. The gravity force effect will be simulated by the settling procedure in which domains are subjected to a simulated gravity field [1]: they fall under gravity and slide down over the already settled domains. This procedure can be modeled by domain translation along the gravitational direction, i.e. by the algorithm

\[
\begin{align*}
\text{For } i = 1 & \text{ To } N \\
D^i & = \{(x_k, y_k) \ (k = 1,2,\ldots,n_i)\} \\
D^i_{\text{after}} & = \{(x_k, y_k - \Delta y) \ (k = 1,2,\ldots,n_i)\} \\
\text{If } D^i_{\text{after}} \cap D^j & = \emptyset \text{ Then } D^i \mapsto D^i_{\text{after}} \\
& \quad j=1,2,\ldots,N; i \neq j \\
\text{Next } i
\end{align*}
\]
Densification model

Densification via LPS generally requires transportation of dissolved solid through the liquid. The characteristic process is solution-reprecipitation in which smaller solid domains dissolve at the solid/liquid interface, diffuse through the liquid and precipitate on larger domains. As a result larger domains grow at the expense of the surrounding smaller ones. This process will be followed by a center-to-center approach. Rearrangement only involves the repositioning of domains, leading to limited densification.

Process modeling

The finite-difference technique will be used for computation of the liquid phase concentration (numerical solution of the diffusion equation) and solution-reprecipitation effect [1].

The pore filling process will be considered similarly as Park et al. [7] who developed models for liquid flow into isolated pores. It was assumed that the initially spherical pores exist within the solid-liquid system in which the grain shape is in equilibrium with the liquid menisci. In the initial stage the pores are stable and individual liquid menisci are maintained between grains around the pores. During sintering when grain growth reaches a critical point the meniscus radius becomes equal to the pore radius. As the grains grow beyond the critical point pores start to decrease. The balance of the liquid pressure between menisci at the pore and those at the surface is now broken and liquid can flow rapidly into the pore. Our model assumes that for each pore there is a critical grain size required for filling. Pore motion will be simulated by a Brownian motion model.

Fig. 2 Three typical snapshots (60 min) of the computed microstructure evolving under various models (White colored regions are solid phase, black colored regions are pores, and the gray colored region is a liquid phase). (a) Initial model; (b) Settling model; (c) Model (A); (d) Model (B).
Results and discussion

For simulation of LPS under gravity condition we will use one basic (Settling Model) and two mixed models: (A) Settling - Densification Model, and (B) Settling - Brownian Motion Model. These models will be tested in order to conduct a study of diffusion and gravitational effects on microstructural development of the W-Ni system. The following data will be used: the equilibrium concentration of liquid in contact with the solid: 35 at.% W [8]; the diffusion coefficient in the liquid W-Ni alloy $10^{-9}$ m$^2$s$^{-1}$ [9]; the sintering temperature: 1750 K; liquid viscosity: $5 \times 10^{-3}$ Pas (for liquid nickel); the acceleration: 9.81 m$s^{-2}$; the liquid-solid density difference: 9 gcm$^{-3}$, and the interfacial energy 0.8 Jm$^{-2}$.

The porous structure will be represented by $N$ domains: $S$ domains as a solid phase and $P$ domains as a pore phase, so that $S + P = N$. An initial model ($N = S + P = 58 + 38$, Fig. 2a) was obtained by applying a random generating methodology [1] and assuming the radius range $10 \text{–} 60 \mu m$ with the initial average domain radius of $34 \mu m$ for solid W, and the pore radius range $5 \text{–} 25 \mu m$ with the initial average pore radius of $9.7 \mu m$.

Under gravity influence the domains fall step by step and slide down over the already settled domains (Fig. 2b). Such a motion stops when each domain reaches a position of local equilibrium, which can be either a position at the experimental region border or the lowest position along the sliding surface of domains. Applying model A (gravity directed motion and center-to-center approach), the domains will be more closely packed inside domain clusters (Fig. 2c). Figure 2d shows the computed microstructure obtained by model B (gravity directed motion combined with random directed translation), which implies that the domains have moved more dynamically, including linear displacement along four possible directions. It can also be seen that most of the sintering time is spent in eliminating the large pores which are now embedded in a much more dense model system.

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References

течне фазе. Ови модели ће бити примењени за проучавање дифузионих феномена и утицаја гравитације на еволуцију микроструктуре током течно-фазног синтезовања система W-Ni.

Кључне речи: Синтезовање у присуству течної фазе, порозна структура, рачунарска симулација.