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Theoretical study of skeletal structure evolution under topological constraints during sintering

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ABSTRACT

In this paper we will consider essential topological constraints and their influence on skeletal structure evolution during sintering. Skeleton structure will be represented by skeleton units and formation of large solid skeletons arranged in a long chain of connected solid-phase domains. Applying three-dimensional domain methodology, solid skeleton evolution will be simulated by an algorithm based on neck growth law and grain-level methodology.

1. Introduction

The theoretical analysis of densification during sintering must consider skeletal structure evolution (SSE) influenced by some topological constraints, because they can cause grain rearrangement and the development of the pore distribution during sintering. Thus the presence of topological constraints in the grain network (solid skeleton) triggers the time evolution of the microstructure, so that as the time progresses the grain boundary slowly migrates via topology changes.

Even though the sintering process has been intensely researched during the past fifty years, there is a lack of quantitative, predictive models for the microstructural evolution during sintering [1]. Various numerical models have been developed to study sintering, where most were idealized two- and three-grain models. The very early computer study that deals specifically with topological constraint effects in large-scale randomly packed grain arrays was due to Ross et al. [2].

Rearrangement of grains is also very important for a better understanding of topological constraints and their influence on sintering, especially during the initial stages [3]. Henrich et al. [4] used the discrete element method to model in the three-dimensional (3-D) case the initial and intermediate stages of solid-state sintering, with an emphasis on the role of grain rearrangement. Using the same method Martin et al. [5] introduced a simple coarsening model in which the compact is modeled as a 3-D random assembly of spherical particles interacting mechanically through their contacts, where contact forces are calculated depending on the physical mechanism activated (elasticity and/or plasticity or sintering). They showed that its use allows a favorable comparison of simulated densification rates with experimental data from the literature. Note that recent progress in X-ray synchrotron microtomography allowed rearrangement effects to be observed in metal powders directly on a grain scale [6].

As Smith [7] has pointed out, normal grain growth results from the interaction between the topological requirements of space-filling polyhedra and the need for surface tension equilibrium, which imposes further geometrical constraints. However, this detailed knowledge of the neighboring grain–grain interactions seems to be impossible to capture in an analytical framework in a self-consistent manner. Therefore, for a comprehensive understanding of this process, analytical

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methods coupled with computer simulations seem to be a possible way for theoretical study of the sintering process. Even more, computer simulation enables one to track the microstructural evolution including SSE of more general and complex systems and evaluate different quantities, e.g. the grain size distribution function, growth kinetics, etc. Such theoretical results can then be compared with experimental results.

In recent years, various types of numerical model have been developed for simulating the detailed microstructure evolution during grain growth (see e.g., a good review in [8]). These simulation models can be divided into two classes: stochastic and deterministic. Stochastic models are generally of the Monte Carlo type, where the most investigated is the Potts model [9–11]. In deterministic models, the motion of grain boundaries is followed by time integration of their positions assuming the normal velocity for the grain boundary to be proportional to the boundary curvature (e.g. the so-called "vertex models" [12–17]).

Generally speaking the sintering can be described as densification macroscopically, but as a complex microstructural evolution of many particles by atomic diffusion at sintering temperatures. In that sense, the elementary sintering processes can be described by analyzing the microscopic evolution of arranged particles. The most successful approaches to understand the mechanisms of neck growth and shrinkage have been the two-sphere model for the early sintering stages (particle bonding) [18,19], the cylindrical pore channel model [20] and the extrapolated two-particle model for the intermediate shrinkage state [21], and the spherical pore model for the late sintering stages [22]. More sophisticated models based on less simplified pore geometries and taking a superposition of the various sintering mechanisms into account have been developed too (see e.g., [23]).

As a matter of fact, the existing models for the microstructural changes can be divided into models based on two particle interactions and models based on topological structure evolution. Kadushnikov et al. [24] combined the two types of model, so that, for the initial sintering stages, they used the generally accepted model based on spherical particles, while for the final stages of sintering, a basic geometrical model has been a polyhedral structure. In their approach the shapes and metrical parameters of the polyhedra are governed by the positions of their neighbors and the internal structures of the approximating particles, while the evolution of the structure is described by the processes occurring in the system. In the intermediate stages, the particles are described in terms of spheres, with the contact zones determined as the intersections of the spheres with the corresponding polyhedra. If there is only a small intersection, it is possible for the spheres to be displaced and the contacts between them to be broken; if there are large intersections (at the later sintering stages), a rigid skeleton network is formed from the sintering particles, and closed pores and pore channels are also formed. Using a consistent model for the sintering of a system composed of a large number of spherical particles, they showed that the geometry of the particles deformed on sintering can be described by a mutually penetrating sphere model.

To improve the sintering models further, special attention should be paid to the consideration of effects such as anisotropic shrinkage [25]. In that sense, for investigation of how anisotropic shrinkage at microscopic length scales is affected by interaction with neighboring particles, specifically, by local rearrangement of particles [3], Wakai et al. [26] applied a three-dimensional computer simulation of idealized sintering. They analyzed the forces acting between particles and showed that rapid anisotropic shrinkage is induced by particle rearrangement during sintering.

As already mentioned, the evolution of grain structures including SSE in materials is a complex and multiscale process that determines the material's final properties. Thus theoretical and computational investigations can help to better understand the impact of SSE. In that sense, the primary task of this paper will be the development of a new computer simulation method to treat topological constraints and their influence on 3-D SSE during sintering. Similar work was reported by Leu et al. [27], where a physically-based two-dimensional simulation method for treating topological effects during the early stage of solid state sintering was considered. In our approach it will be assumed that the skeletal structure develops early in the process and hence it is important to define a model based on inter-grain behavior. Central to such a study will be unit problems for the interaction of grains within solid skeleton. The compact will be modeled as a 3-D random (irregular) assembly of grains interacting mechanically through their contacts (inter-grain level), whereas SSE could then be related directly to the neck size of contacting solid-grains and the reduction in the surface energy of the solid skeleton network subjected to topological constraints of space filling. Analysis of grain coarsening due to the neck growth poses other particular computational difficulties and will be published in a separate paper.

2. Modeling skeleton structure

For the simulation of solid skeleton structure during sintering we will define a numerical model based on domain methodology and on sub-models for constrained SSE. Skeletal structures will be computed by geometrical limitation for two grains in contact with an observable dihedral angle. Once formed, grain contacts grow to satisfy the dihedral angle, and beyond that point neck growth is paced by the rate of grain growth.

2.1. Domain topology

For simulation of sintering it is convenient to use multi-grain models of regular shape because they need to store only the position, orientation and size of each grain. The model does not need other geometric assumptions because the microstructural development can be simulated by a set of simple local rules and overall neck growth law. In that sense,