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Multi-scale modeling of shape distortions during sintering of bi-layers

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ABSTRACT

Models for deformational behaviors of porous bodies during sintering often rely on limited number of internal variables as they are formulated based on simplified or ideal microstructures. Considering realistic microstructures can improve the predictive capabilities of the already established theories like the continuum theory of sintering. A new multi-scale numerical approach for modeling of shape distortions during sintering of macroscopically inhomogeneous structures combined with a microstructure model is developed. The microstructures of the porous body are described by unit cells based on kinetic Monte Carlo (*kMC*) model of sintering. During the sintering process the shrinkage rate is calculated from the *kMC* model. With the help of computational homogenization, the effective viscosity of the powder compact is also estimated from a boundary value problem defined on the microstructures of unit cells simulated by the *kMC* model. Examples of simulation of sintering of bi-layers based on different material systems are presented to illustrate the multi-scale model. The approach can be considered as an extension to the continuum theory of sintering combined with the meso-scale kinetic Monte Carlo model.

1. Introduction

Shape instabilities during sintering of functionally graded porous multi-layers is one of the problems often observed in the development of various technologies like the solid oxide fuel cells (SOFC), gas purification membranes, etc. [1–8]. Frequently, this problem is related to the differential shrinkage that arises from the macroscopic inhomogenities in the porous body [9–12]. The effect of the sample own weight has recently been shown to be another factor generating stress and distortions during sintering of multi-layer systems [4,6,7]. Generally there is a growing interest towards explicit understanding of the effect of inherent material properties on the extent of shape distortion during sintering of multi-layered porous structures [13]. Significant achievements in modeling of densifications and shape distortions in multi-layer systems have been made using the continuum theory of sintering [2,11,13,14]. The continuum theory of sintering predicts the macroscopic displacement rate in the porous bodies assuming that the powder particles behave in an incompressible linear viscous way [14]. The driving force for sintering (sintering stress) is treated as an additional hydrostatic pressure that arises due to the minimization of surface energy in the entire volume of the sintering specimen. Therefore, to implement the continuum theory of sintering, it is necessary to experimentally measure the sintering behavior i.e. sintering stress or shrinkage rate and the effective viscosity of the porous body. Usually separate set of experiments is used to study those two properties which are then implemented in a model of multi-layer systems [5,10,15,16].

In general, the magnitude of shrinkage rate and the effective viscosity are functions of the internal parameters of the porous structure [14]. These internal parameters are for example the amount and size of pores, grain size of particles, average neck radius, pore diameter to grain size ratio, powder packing, dihedral angle, etc. [17]. Traditionally porosity and grain sizes are considered as the internal parameters defining the evolution of sintering stress and viscosity in the continuum theory of sintering [11,18,19]. This practice however needs to be extended further to account for other internal parameters and hence refine the predicting capabilities of the models. The challenge here is not only the lack of explicit knowledge about which internal parameters are necessary for accurate modeling of the material behaviors, but also that these parameters are very cumbersome to obtain by direct experimental investigations.

The natural extension of the continuum model is to directly consider the internal geometric features of the porous structure to estimate the shrinkage as well as the viscous behaviors. This can be done through a so called multi-scale modeling procedures [17,20,21]. If a representative unit cell of the microstructures evolution is defined during the sintering cycle, it is possible to extract

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the viscous behaviors of the porous body using techniques in multi-scale modeling. The first challenge here is to be able to predict the proper microstructural geometry of the powder compact and to model its evolution in time. Kuzmov et al. have considered simplified unit cells with circular and elliptical pores and they have also demonstrated the impact of shape of pores on the densification as well as distortion of bi-layered structures [21]. Kuzmov et al., however, assumed the sintering stress to be a curvature dependent traction applied on the surface of the pore, which still limits the effect of other internal parameters on the shrinkage rate of the porous body [21].

Recently there have been a number of reports showing the capabilities of a numerical model based on a kinetic Monte Caro (*kMC*) method [22,23]. The *kMC* model is able to simulate both densification and microstructural evolution during sintering of a powder compact at the meso-scale level. This model is based on solid state sintering having the ability to simulate the different underlying phenomenon during sintering of a powder compact including grain growth, pore migration and vacancy annihilation. It has demonstrated its robust capabilities in predicting the microstructural evolution during sintering of porous bodies [22]. The model has been compared with the sintering of copper spheres as observed using X-ray tomography [22], as well as been used to study the sintering of close packed spheres and powders with a particle size distribution [23,24].

In an attempt to obtain more accurate expressions for sintering stress and bulk viscosity, Olevsky et al. used the 2D microstructures of porous body simulated by the *kMC* model [25]. Olevsky et al. were able to determine the interfacial-free energy for a series of microstructures in time so as to calculate the sintering stress. The sintering stress and normalized bulk viscosity obtained from the *kMC* model is also compared with the result found using the model suggested by Skorohod [41] where a good agreement is shown in the case of normalized bulk viscosity [25].

In this work, a unit cell based on the *kMC* model is established for multi-scale modeling of sintering. This approach is advantageous as the *kMC* model can be extended to provide the important parameters to the continuum model i.e. the densification/shrinkage and viscous behaviors of the powder compact, without having to conduct large amount of experiments. Furthermore the microstructural simulation based on the *kMC* model has the potential to incorporate any of the internal parameters for the given powder compact without limitations.

Therefore the objective of this study is to create a unified model, which couple the local kinetic Monte Carlo (*kMC*) model with a global finite element model in order to predict the shape distortions, e.g. during sintering of bi-layered porous structures. The *kMC* model provides the microstructure evolution as a function of time and temperatures for the representative volume elements (*RVEs*), which are then used to extract the shrinkage as well as viscous parameters of the powder compact that often require tedious experimental work.

2. Multi-scale modeling

The multi-scale modeling approach used in this study consists of two models; one at meso- and another at macro-scale. The meso-scale model is the previously discussed *kMC* model for solid state sintering, where as the macro-scale model is based on finite element method. The meso-scale model is considered to be the *RVE* of the macro-scale geometry, where the *RVEs* are assumed to be distributed throughout the finite element nodal points of the macrostructure.

Two main mechanisms are considered in the deformation of the microstructure, i.e. (1) sintering contraction through diffusion mechanisms (2) viscous deformation of the microstructure from stresses. (1) Is the shrinkage rate in the powder compact $\dot{\varepsilon}^s_{kk}$, which is simulated by the *kMC* model. Hereby introduction of concepts like sintering stress is avoided. In the current work the influence of stress on the annihilation processes and pore migration in the *kMC* are disregarded. This approach is valid for systems where the microstructural evolution is not strongly affected by the stresses throughout the system. While the change in microstructural evolution of course depends on the magnitude of the internal or external stresses, it has been shown that while pores orient depending on the external stress, properties such as the mean grain size can differ little between constrained and freely sintered samples [26,27]. The onset of microstructural anisotropy also depends on the sintering mechanisms; Alumina, sintering by solid state sintering, develops anisotropy during the final stage of sintering [28], while glass, sintering by viscous flow, is anisotropic during sintering but becomes isotropic at the end of the process [29]. Furthermore, the influence of the stress also varies within the sample, allowing parts of a constrained sample to behave as though it was freely sintering [30]. (2) The viscous deformation of the microstructure is handled by a microstructural finite element model with which a homogenization procedure is performed.

The extractions of the viscous properties of the powder compact D_{ijkl}, are based on a special set of boundary value problems (*BVPs*) established on the *RVEs*. The theory of computational homogenization is implemented on each *RVE*, which in this case is based on the microstructure obtained from the *kMC* model in time. The boundary conditions applied on the *RVE* are updated in each time step using the viscous strain rate from the macro-scale model. Fig. 1 shows a schematic flow diagram of the multi-scale modeling procedure used in this study.

Below are the details for the different parts of the multi-scale model.

2.1. Unit cell model or RVE

The kinetic Monte Carlo defines individual grains and pores on a two or three dimensional square/cubical grid, where a single grid cell is referred to as a voxel in both two and three dimensions. The model simulates grain growth, pore migration and vacancy formation and annihilation through diffusion processes. The driving force for sintering in the model is the capillarity (reduction of interfacial free energy), which is defined by the neighbor interaction energy between voxels. Only a brief description of the model is given here as it has been described in detail in previous works [22,31].

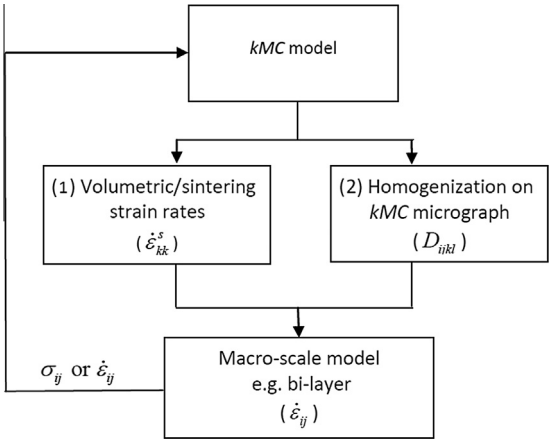

Fig. 1. Schematics of organization in the multi-scale modeling.