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Study of the sintering kinetics of bimodal powders. A parametric DEM study

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ABSTRACT

This paper presents a DEM study of the sintering of bimodal powders. The influence of every type of contact between small–small, large–large and small–large particles on the macroscopic behavior is investigated. A generalized mean strain law that takes into account the contribution of the different contacts was used to compare the theoretical kinetics of the assembly with the DEM results. It came into view that the kinetics of the different contacts is shifted toward the average kinetics of the packings.

The capability of a contact type to drive the macroscopic strain was found to be not only dependent on its probability but also on its equivalent radius.

Hence, densification appears to be slower than predicted by the mean strain law when small–small contacts dominate with an important residual number of larger contacts.

1. Introduction

Solid-state sintering is a major process in ceramic and nuclear industries. It can be defined as the thermal transformation of bulk materials into compact solids below their melting point. The main phenomenon is solid state diffusion which occurs along the different interfaces of the crystals and through its vacancies.

According to the industrial applications, the sintered material has to be homogeneous and the characteristics like porosity have to be controlled. The particle size distribution (PSD) is one of the main parameters that determine the packing evolution and the microstructure properties. Sintering of different sized particles was studied on a grain scale by Wakai et al. [2] and Pan et al. [3]. Then, numerous studies investigated the impact of the PSD on the structure of packing both experimentally [4–7] and numerically [8–12]. Some general aspects of the effect of PSD are well known. For example, there is a consensus on the fact that width PSD leads to higher compacity of green packings and increases particles rearrangement [11,13].

Nevertheless, the effect of PSD on the kinetics of the different type of contacts is not well known. Various sizes of particles lead to various kinetics for contacts. The macroscopic behavior depends strongly on the initial structure of the green packings and the role of the PSD varies with the dominant diffusion mechanism [8]. This makes a general interpretation very complicated. Ting and Lin [10] predicted that there is an optimum width of the PSD to get the highest densification rate. More recently, Bjork et al. [12] showed that densification rate and final density decrease with the broadening of the distribution, which is in good agreement with Wonisch et al. [11]. Conversely, Yeh and Sacks [4] found that narrow and broad PSD lead to the same final density. Grain growth also appears to be a major reason for the decrease of densification rate of broad PSD [4,5,10,12].

Bimodal distribution is a case study which allows a simpler interpretation of data. Liniger and Raj [6] observed that bimodal assemblies allow a decrease of defect sizes compared to mono-sized. Petersson and Agren [7] studied the densification rate of mono-sized and bimodal assemblies and showed that bimodal assemblies initially show a faster densification and a slower densification in the latter stages.

However, the comparison between the kinetics of densification of polydisperse and mono-sized is generally based on volume based average radius. This only gives partial information about the contribution of the different contacts. Hence, for a more accurate description, the definition of the representative radius should take into account the contribution of every type of contacts [8]. Moreover the impact of the different type of contacts on the macroscopic strain has not been investigated so far.

This paper aims at understanding the influence of the PSD on sintering making use of the discrete element method (DEM). The later allows the simulation of thousands of particles and takes into account the motion of every particle in the packings [14–16]. Moreover, this study is based on an improved DEM called contact dynamics which does not need mass scaling for the motion law integration. In a previous study, we showed that contact dynamics gives a better representation of rearrangement compared with traditional (smooth) DEM [17]. The

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Nomenclature

α, β	Parameters which depend on the ratio of surface and grains boundary diffusion [1]
Δ_b	Grain boundary diffusion parameter
δ_b	Grain boundary thickness
δ_s	Surface thickness
$\dot{u}_t$	Particles relative tangential velocity
η_c	Viscous friction coefficient
γ_s	Surface energy
Ω	Atomic volume
ψ	Dihedral angle
τ	Characteristic time
θ	Rearrangement parameter which represents the angle between the real motion of particles in the assembly and the affine motion toward the center of the assembly
c_{ll}	Fraction of large–large contacts
c_{sl}	Fraction of small–large contacts
c_{ss}	Fraction of small–small contacts
D	Density
d	Initial distance between the centers of particles when the contact is punctual
D^o	Initial density
D_b	Grain boundary diffusion coefficient
Ds	Surface diffusion coefficient
fn	Repulsive part of the normal sintering force
f_n^{sint}	Normal sintering force
h	Particles indentation
Kb	Boltzmann constant
R	Particle radius
R^*	Equivalent radius for two particles of different size
R_1	Radius of particle 1
R_2	Radius of particle 2
s_l	Surface fraction of large particles
s_s	Surface fraction of small particles
T	Temperature
Vn	Particles center to center approach velocity
x_l	Fraction of large particles
x_s	Fraction of small particles

behavior of bidisperse powders is compared with mono-sized powders. For the bidisperse case, three different types of contact can be defined: small–small, small–large and large–large. Their individual kinetics in the packings and their contribution to the macroscopic strain are investigated for different size-ratios and fractions of small particles.

2. Numerical method

2.1. Sintering model

For any system, the minimum of surface energy is reached for a perfectly homogeneous curvature. If we consider two spherical particles in contact, the variation of curvature induces a stress gradient, given by the Laplace law, which tends to decrease the total surface area. Hence, diffusion occurs through the vacancies of the crystals, along the surface and along the grain boundary. Gas transport phenomena can also appear. In this case, the diffusion kinetics is driven by evaporation–condensation. Evaporation–condensation happen only at the surface of grain and has the same effect as surface diffusion.

Densification is due to a combination of grain boundary diffusion that brings matter from the contact area to the surface of the neck and surface diffusion that spreads out the extra matter along the surface of particles. Only these two diffusion paths are considered here.

The DEM model of sintering is based on Coble's geometrical model [18] which describes the evolution of particles under the form of truncated spheres (Fig. 1). Particles are assumed to be single-crystals and to remain spherical. Dihedral angle ψ is constant which means that mechanical equilibrium is reached at any time.

Using these assumptions, the normal force between two particles in contact can be written analytically [1,19,20]. The first member of the Eq. (1) represents the attractive force of sintering which comes from the integration of the Laplace pressure over the surface. The second term is homogeneous to a viscous force opposed to the attractive force. This force represents the grain boundary diffusion which is considered to drive densification. The ability of surface diffusion to lay out the matter brought to the surface of the neck by grain boundary diffusion is introduced under the form of two coefficients α and β. Both are functions of the ratio between grain boundary and surface diffusion [1]. It follows:

$$f_n^{sint} = -\frac{\pi\gamma_s R\alpha}{\beta} + \frac{\pi r^4}{2\Delta_b\beta} \times V_n \tag{1}$$

with: $\Delta_b = \Omega\delta_b D_b / K_b T$.

R is the particle radius, γ_s the surface energy, and V_n the center to center approach velocity. D_b is the grain boundary diffusion, δ_b the thickness and Ω the atomic volume. r is the neck radius which grows according to Coble's model given by the following equation:

$$\frac{dr}{dt} = \frac{R}{r} V_n. \tag{2}$$

For this study, we took $\alpha = 9/2$ and $\beta = 4$ which are the values given by Martin and Bordia [20] for $\delta_b\, D_b / \delta_s\, D_s = 2$.

Furthermore, the tangential velocity is defined as the relative motion of two particles parallel to their contact plane. For two particles in contact, the tangential velocity comes from interactions with neighboring contacts. Thus, for a multibody system, a tangential resistance to sliding must be considered. Assuming that sliding was driven by diffusion, Raj and Ashby [21] proposed a convenient equation for this force as a function of the contact area and of the roughness of the grain boundary. The roughness of the grain boundary is introduced in the equation under the form of a viscosity coefficient η_c:

$$f_t = -\eta_c \frac{\pi r^2 R^2}{2\beta\Delta_b} \dot{u}_t \tag{3}$$

with $\dot{u}_t$ the tangential velocity that also takes into account the rotational velocity. Nevertheless, rotations are known to be negligible because of the solid neck between particles [15].

The value of η_c varies between the different studies [16,20]. For this study, it is taken equal to 10^{-3} according to [20].

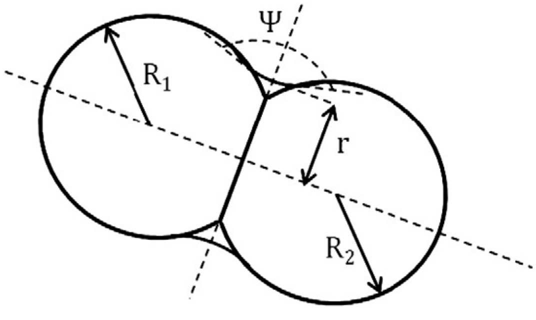

Fig. 1. Discrete element model for the simulation of sintering. R_1 and R_2 are the radius of particles, r is the neck radius, and ψ the dihedral angle.