Phase-field simulation of sintering processes

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1 Introduction

The sintering process impacts everyday life in various ways: From the simple coffee cup which undergoes the process, over solar cells whose electrical contacting is sintered, right up to glaciers whose melting behaviour also depends on how they sinter. Whether that solar cell you have sintered is efficient or not depends on the microstructure[1], as it changes the conductivity across the electrical contact. This principle of the microstructure influencing properties spans across every imaginable material and thus predicting the microstructure after a process is of high importance. The sintering process is deceptively simple: Take a powder, compress it a bit so it doesn't fall apart under its own weight and then heat it up in an oven. What happens is that the particles which make up the powder move closer to each other while also growing at the expense of each other. These two processes are called densification and grain growth and form the fundamentals of predicting what happens during the sintering process. While simple to describe, a fully quantitative model of sintering still eludes the scientific community. In this project the focus is on building a model for the densification of a powder while resolving the individual particles. This builds the basis for simulations which predict the microstructure during the entire sintering process, allowing for improvements to the process and materials properties.

2 Results and methods

The phase-field (PF) method[2, 3] is employed in this work to resolve individual particles, which can grow, shrink, change their shape and move in space. Grain growth is roughly accounted for with the first three behaviours. Densification originates from the motion in space. In order to accurately account for this motion, a method which is closer to nature, molecular dynamics, is employed to determine how the grains move during the sintering process. By building a model which accounts for this motion, without requiring atomic information, the small time and space scales of molecular dynamics can be escaped from. This allowed the simulation of over 120 000 particles until they sintered close to 100% density, which would be entirely beyond the scope of molecular dynamics even on today's largest computing facilities. Even though it would be beyond the scope of molecular dynamics, without the processing resources at the HLRS and significant time spent in optimizing the communication and calculation structure of the PF code, the simulations would not have been possible

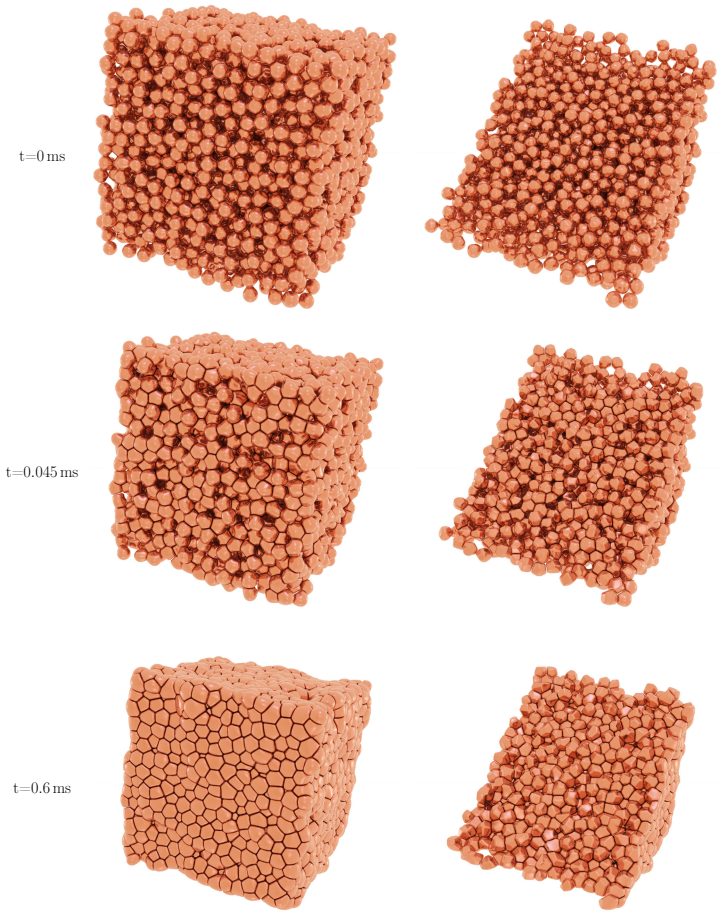
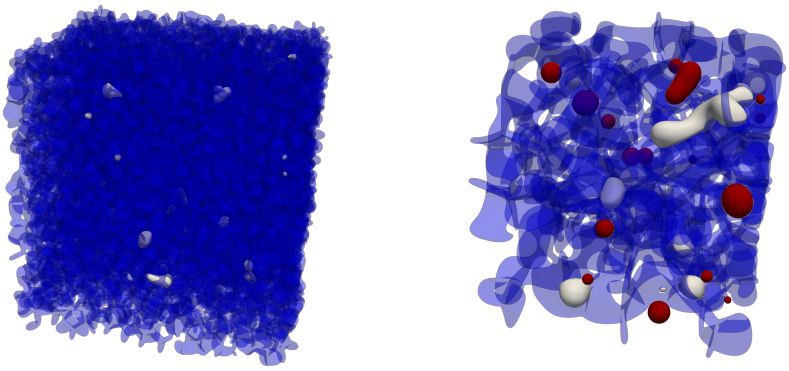


Figure 1: A full view (left) of a sintering simulation and a fracture surface view (right). This simulation is representative for quick densification without grain growth. It shows both densification and grains transforming from spheres to polyhedra.

either. This molecular dynamics inspired PF model is used to investigate the effects of particle size, materials parameters and packing shape on the sintering process. A typical simulation evolution is shown in Fig. 1 when grain growth is mostly suppressed by choosing a low grain boundary mobility. The initially cubic structure shrinks while keeping its general shape and the grains are observed to polyhedralize. Not shown are the remaining pores, which are all still attached to grain boundaries. This attachment will eventually allow the total elimination of porosity.

The situation differs remarkably when grain growth is not suppressed. Figure 2 shows a comparison of a simulation with and without suppression of grain growth, at the same relative density of 82%: On the left the earlier simulation with suppressed grain growth is shown and barely anything inside the green body is visible because the grain boundary network (blue) is very dense. The few gray



(a) no grain growth (b) grain growth + slow densification

Figure 2: The grain boundary network (blue), as well as isolated (gray) and detached (red) pores. If no significant grain growth occurs as on the left, pores stay attached to grain boundaries and can be eliminated. If grain growth occurs concurrently with densification, pores will detach from the grain boundary and limit the achievable density.

regions inside are isolated pores which are still attached to grain boundaries. On the right a simulation with high grain mobility and reduced grain boundary diffusion is shown: The high grain mobility allows grains to grow faster than the pores can move, detaching them. These detached (red) pores will not be eliminated on processing timescales and thus set a limit to the achievable density. Furthermore, these pores on their own will also influence materials properties e.g. by acting as crack starters within the material or by reducing the electrical conductivity.

3 On-going Research / Outlook

Simulations such as these form the basis of process control and computational materials design: Given that one wants to avoid certain microstructures, what kind of relations are permissible for the grain growth speed relative to the densification speed? In what way do these speeds follow from measurable properties such as grain mobility or grain boundary diffusion? By answering these questions the process can be controlled to produce desirable microstructures yielding desirable materials properties fit for various applications.

While the modelling part of this question has been advanced significantly within the project, there is still room for improvement when accounting for the locality of densification as well as the modelling of the driving force for densification. Beyond the model, a key problem is the determination of the input parameters for the phase-field simulation such that it actually approximates a certain material, with 50 or more parameters needing to be known for the full description even for a simple system[4]. This poses a challenge for experiment, theory and simulation, as none of these alone could stem the burden of giv-

ing cheap and accurate estimates for the parameters. One possibility, given an accurate model, is to employ data assimilation methods[5]: Based on experimentally observed data, the parameters of a phase-field model are adjusted to fit to the observations (e.g. density over time) in an automatic scheme. Presuming that e.g. the temperature dependence of kinetic parameters such as diffusivities is not affected by the geometry, this would allow the determination of many parameters.

References

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