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IRCER/CEA laboratories - Limoges city in France

Analytical and discrete element numerical modeling of ceramic sintering made by additive manufacturing

Context of study

CEA Le Ripault is developing digital materials engineering based on digital design tools associated with the capabilities of additive manufacturing technologies. These technologies allow the production of structures specifically designed by calculation (topological optimization for thermal, mechanical or other properties). The CEA is particularly interested in the realization of refractory porous ceramic structures in order to use them, for example, for their properties (i) of hot gas diffusers, (ii) of thermal insulation at high temperature, (iii) of solar flux absorbers, etc.

To achieve this, the stereolithography technique was chosen. This additive manufacturing technique by photopolymerization is based on the successive stacking of photopolymerized layers of a liquid resin, following its irradiation by a radiation of a specific wavelength. When manufacturing ceramics by stereolithography, the resin must be loaded with an inorganic precursor, usually ceramic powder. Once the part is printed, it is thermally treated to eliminate the organic auxiliaries (known as debinding) and also to promote the formation of bridges between the elementary particles by material diffusion (known as sintering) and thus consolidate the granular stacking. It is important to control the heat treatment parameters (temperature, heating speed and duration) because they influence the final microstructure of the ceramic object. In order to ensure the quality and reproducibility of the manufactured parts, it is important to understand their impact on the consolidation of the part and its residual porosity. Analytical modeling and numerical simulation of the physical phenomena involved must help to achieve this and will be the focus of this work.

Objectives of the study

This study, which is a collaboration between the IRCER (Institute of Research on Ceramics - UMR CNRS 7315) and the CEA Le Ripault, proposes to develop a model of the sintering behavior of a ceramic object with a specific porous architecture using the Discrete Element Method (DEM). Indeed, sintering involves complex phenomena such as rigidification, densification or granular coarsening, which depend on the granular packing. A key scale to understand these complex phenomena is the meso-structure, which involves a few thousand of grains interacting through mechanical contacts (at the beginning of sintering), gradually giving way to the formation of bridges (necks) between grains during sintering. The discrete element approach allows to predict in a natural way these granular rearrangements from the fine knowledge of the driving forces governing the interaction between grains. Particular attention will therefore be paid to the choice of these micromechanical interaction laws between the grains, which must be capable of representing the consolidation and densification kinetics. These numerical developments will be carried out within the discrete element code GranOO co-developed by the IRCER laboratory. It will then be necessary to develop a numerical approach capable of simulating the material transport paths involved in the creation of bridges between particles and to predict the apparent thermo-mechanical properties in a multi-scale approach.

The construction of the numerical model will be based on the use of a reference material whose nature and physical properties of the basic constituents are known. The study material selected is a composite consisting of a polymeric matrix (i.e. a resin) filled with spherical silica particles, having a monodisperse size distribution around a mean value of a few µm. The initial microstructure of a sample of this material, before heat treatment, will be obtained by high resolution X-ray microtomography. It will allow to build the representative numerical volume from which the simulation of the phenomena will be carried out.

In order to validate the developed model, the evolution of the simulated microstructure during the heat treatment will be compared to the microstructures tomographed at different stages of the treatment. A complete work of 3D reconstruction, image processing and analysis will be carried out to extract the statistics of the parameters of interest (contact surface, porosity...). Observations made in an environmental SEM will be able to bring complementary information on the one hand, on the main stages of morphological transformations of the granular packing and, on the other hand, on the kinetics of formation of bridges between particles in an isothermal regime. On the basis of these experimental data, it will be possible to identify the mechanism(s) responsible for the growth of the bridges and, more particularly, the diffusion pathways at the origin of the growth of the necks between the silica particles. These behavioral laws will be useful to guide the numerical simulation in the prediction of the granular stacking behavior. In addition, the size of the necks can be measured with a very good resolution by the FIB/MEB of IRCER on representative volumes of a few tens of grains in contact.

Moreover, the values of the effective properties predicted by these simulations (mechanical, thermal) will be confronted on the one hand with the experimental measurements resulting from the characterization benches of the IRCER and the CEA, and on the other hand with the properties resulting from the simulations of the codes of the CEA carried out on the same digitized structures. At the end of this work, the numerical tool developed will allow to carry out parametric studies to optimize the thermal treatment and/or the granular distribution (polydisperse and not only monodisperse) in order to control the consolidation of the parts realized by stereolithography and to minimize their residual porosity with a view to optimize the material with regard to its later applications.

Milestones and main points

This study will be conducted in 6 main phases :

1. appropriation of the subject through a bibliographical study concerning in particular (i) the analytical models of bridge growth accompanied or not by densification phenomena, (ii) the "material" parameters adapted to the description of the behavior of amorphous silica particles by the DEM.
2. acquisition of experimental data : measurements of apparent properties such as thermal conductivity and modulus of elasticity of sintered materials, measurements of thermal behavior and sintering kinetics of granular compacts by thermogravimetry (ATD/ATG/DSC) and dilatometry, then fine observation of the microstructural evolution by in situ techniques (tomography, SEM and SEM-FIB)
3. Modeling by DEM the sintering : development of interaction models to represent the inter-grain bridging, implementation of modeling strategy of the phenomena of densification and granular magnification, choice of the most suitable contact laws.
4. implementation and integration of these developments in the discrete element code GranOO
5. validation of the developed models by comparison of the simulated and observed microstructures and comparison of the simulated effective properties with those measured experimentally and/or calculated using the CEA codes
6. writing of the thesis and associated publications.

keywords

Sintering, ceramics, modeling, discrete elements, mechanics, thermics, microstructure

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