Sintering during constrained forging and isostatic pressing: the influence of the interstitial stress.

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SUMMARY. The importance of the interstitial stress exerted by the pressure of the gas against the grains during sintering of pre-compacted nano or micro powders is determined in this paper for different loading modes.

1 INTRODUCTION

Sintering pre-compacted powders (ceramic or metallic) is still of great industrial interest, especially after the latest wide employment of grain sizes of the order of a few nanometers.

Unlike previous contributions [3], this paper, based on ongoing work [4, 5, 6], faces the issue of accounting for the interstitial stress (often called either Laplace pressure or sintering stress) exerted by the pressure of the gas against the grains. This is not only important for free sintering but also in all of the cases in which moderate external loading are applied during the process. Furthermore, the necessity of considering the effect of such a stress gets amplified whenever nanopowders are utilized.

For the sake of illustration, cylindrical samples are considered in the sequel undergoing four different loading modes, such as forging, constrained forging, isostatic pressing and free sintering. In particular, the second mode is studied here and in [4, 6] for the first time.

The error on the sintering time evaluated by neglecting the Laplace pressure is determined for each loading mode (but for free sintering). Henceforth, threshold pressures remain associated with any admissible value of the calculated error.

Moreover, for the first time, the effect of the interstitial stress is here determined on the residual porosity detectable at the end of any sintering process.

A general stability analysis for each loading mode is performed; in particular this becomes crucial at the occurrence of a critical porosity. This may arise whenever the stress produced by external loading would equate the Laplace pressure. Indeed, the stability of such a situation would entail the achievement of a limiting porosity below which the process could not proceed. This circumstance actually may arise in the case of constrained forging, where the lateral expansion is prevented, for which the balance between the applied stress and the Laplace pressure represents then a dangerous situation. For instance this stability does not occur during isostatic pressing, where a desired instability owes the continuation of the process.

2 THE MODEL

The mechanical response of a porous body with nonlinear-viscous behavior is described by a constitutive relation that interrelates the components of a stress tensor σ_{ij} and a strain rate tensor $\dot{\varepsilon}_{ij}$[1].

$$\sigma_{ij} = \frac{\sigma(w)}{w}[\varphi \dot{\varepsilon}'_{ij} + \psi \dot{e} \delta_{ij}] + p_l \delta_{ij}. \tag{1}$$

where $\dot{\varepsilon}'_{ij}$ denotes the deviatoric strain rate tensor, and w is the effective equivalent strain rate. This is connected with the current porosity and with the invariants of $\dot{\varepsilon}_{ij}$:

$$w = \frac{1}{\sqrt{1-\theta}}\sqrt{\varphi\dot{\gamma}^2 + \psi\dot{e}^2}, \tag{2}$$

where

$$\dot{e} = tr\dot{\varepsilon} = \dot{\varepsilon}_{ii}, \tag{3}$$

$$\dot{\gamma} = \sqrt{\dot{\varepsilon}'_{ij}\dot{\varepsilon}'_{ij}}, \tag{4}$$

i.e. $\dot{\gamma}$ is the second invariant of the deviatoric strain rate tensor.
The dependence of effective equivalent stress $\sigma(w)$ on the effective equivalent strain rate w determines the constitutive behavior of a porous material.

Following Ashby [9], a power-law mechanism of deformation is assumed:

$$\frac{\sigma(w)}{\sigma_0} = A\Big(\frac{w}{\dot{\varepsilon}_0}\Big)^m, \tag{5}$$

where A and m are material constants (A is the temperature dependent, $0 < m < 1$), σ_0 and ε_0 are the reference stress and the reference strain rate, respectively. Two limiting cases corresponding respectively to the ideal plasticity and linear viscosity are given by $m = 0$ and $m = 1$.

The quantities ϕ and ψ represent, respectively, the non-dimensional shear and bulk moduli. In literature several studies relative to the determination of these moduli and their dependence upon the porosity are present.
The quantity p_l represents the interstitial pressure produced by the gas contained in the pores (in the sequel we shall refer to p_l as either the "Laplace pressure" or the "sintering stress").
The effective Laplace pressure p_l is the result of collective action of local capillary stresses in a porous material. A variety of approaches can be found in literature (these contributions are summarized in [1]. We shall consider two possible derivations of the expression for the Laplace pressure (Figure 1 shows a comparison between the different models):

- 1. Sintering stress derived by using a stochastic approach [7],

$$p_l = \frac{3\alpha}{r_0}(1-\theta)^2, \tag{6}$$

- 2. Sintering stress derived by an averaging approach involving the dissipation at the microlevel [3]

$$p_l = \frac{2\alpha}{r_0}\sqrt{\frac{3}{2}\psi(\theta)\frac{\theta}{1-\theta}}, \tag{7}$$

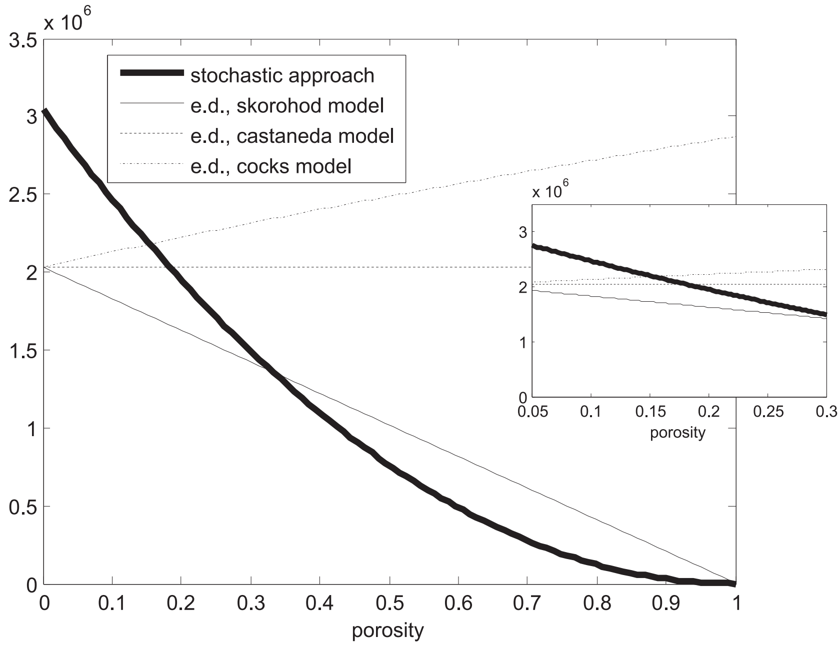

Figure 1: the Laplace pressure as function of porosity

where α is the surface tension and r_0 is the characteristic radius of particles.

Both the Laplace pressure and the stress due to the applied load can be considered as "driving forces" of the process. In previous contributions, either the influence of the interstitial stress is neglected (see [2]) or the free sintering case is considered (see [3]). In [4], [5], [6], the effect of the presence of both the Laplace pressure and the stress due to loading of pre-compacted (micro/nano)-powdered cylindrical specimen is analyzed .

Four loading modes, are here considered (see Fig.2): (i) forging, (ii) constrained forging, (ii) isostatic pressing and (iv) free sintering (this mode can be regarded as a particular case of (iii)). In the sequel, we will focus on modes (ii) and (iii).

For the four different loading modes, it is possible to obtain the evolution law of the porosity θ:

- forging [4, 6]

$$\dot{\theta} = \dot{\varepsilon}_0(1-\theta)\Big(\frac{|\sigma_z - p_l|}{A\sigma_0}\Big)^{\frac{1}{m}}\Big[\frac{\psi}{1-\theta}\Big(\frac{6\psi}{\varphi}+1\Big)\Big]^{\frac{1-m}{2m}}[3\psi]^{\frac{-1}{m}}. \tag{8}$$

- constrained forging [4, 6]

$$\dot{\theta} = -\dot{\varepsilon}_0\Big(\frac{|\sigma_z - p_l|}{A\sigma_0}\Big)^{\frac{1}{m}}(1-\theta)^{\frac{3m-1}{2m}}\Big(\frac{2\varphi}{3}+\psi\Big)^{\frac{-(1+m)}{2m}}. \tag{9}$$

- isostatic pressing [4, 5]

$$\dot{\theta} = -\dot{\varepsilon}_0(1-\theta)^{\frac{3m-1}{2m}} \left(\frac{|\sigma - p_l|}{A\sigma_0}\right)^{\frac{1}{m}} \psi^{\frac{-(1+m)}{2m}}. \tag{10}$$

- free sintering [3, 4, 6]

$$\dot{\theta} = -\frac{p_l}{\sigma_0 A}\dot{\varepsilon}_0 \frac{1}{\psi}(1-\theta). \tag{11}$$

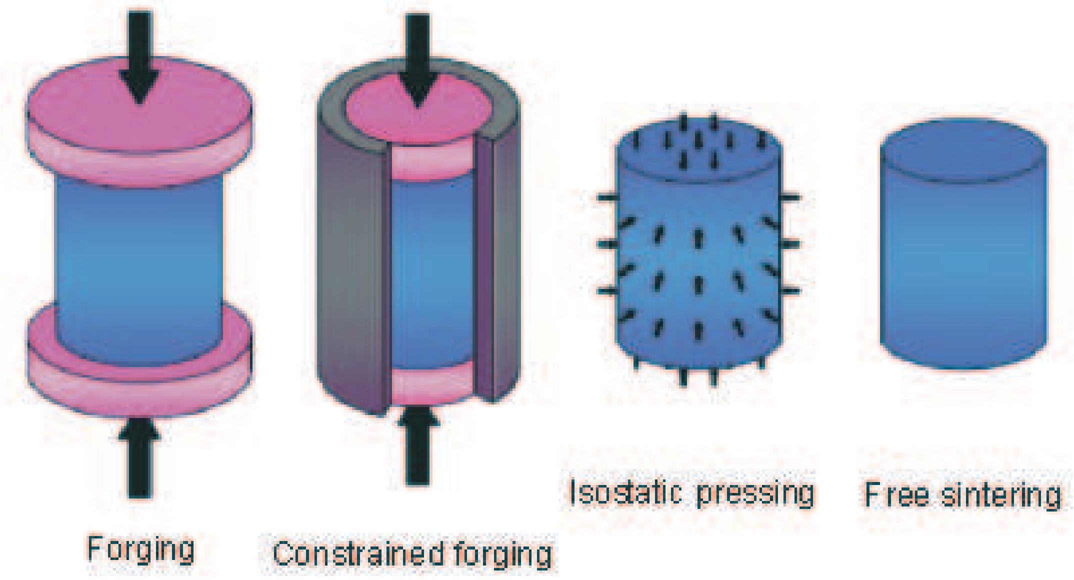

Figure 2: Different loading modes: forging, constrained forging, isostatic pressing

In the sequel, the time-evolution of the porosity will be studied. Comparisons of the achieved results with the ones obtained by neglecting the effects of the Laplace pressure will be performed. Obviously, the neglection of the sintering stress is equivalent to consider non-pressurized voids. Henceforth, no pressure could act against stresses caused by external loading. During sintering, the stress due to the applied load, σ (or σ_z, depending on the loading mode), may vary; on the other hand, the Laplace pressure may evolve according to either relation (6) and (7) (see 1).
Furthermore, the rate of change of the porosity is proportional to the quantity $|\sigma - p_L|$. It is therefore possible that, for a definite range of external loads, the equality condition $|\sigma - p_L| = 0$ may be achieved. We then may distinguish three cases:

1. *$\sigma > p_l$ during the whole process.*
   Here the force driving the sintering is the external loading, whereas the Laplace pressure act against the process.

   Consequently, the time decay of the porosity obtained by neglecting the interstitial stress would be faster than the real one (see Figure 3, referring to he isostatic pressing case).

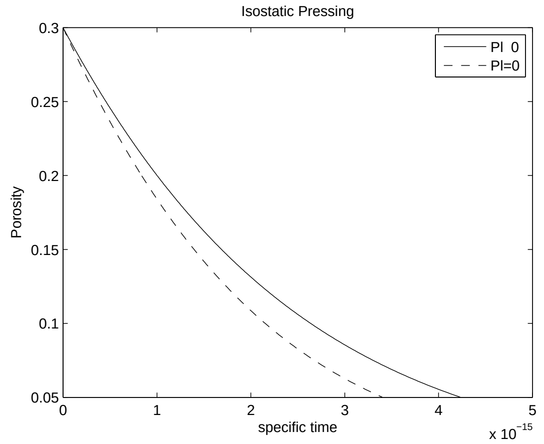

Figure 3: Evolution of porosity- $\sigma > p_l$ during the whole process

2. *$\sigma - p_l$ changes sign during the process* .
   When the condition $|\sigma - p_L| = 0$ is achieved, by equations (8, 9, 10, 11) the rate of change of the porosity is null, $\dot{\theta} = 0$, and the porosity remains constant, at a value that we may label θ^*. Such a θ^* may be called *critical porosity*. For the sake of illustration see e.g. Fig. 4.

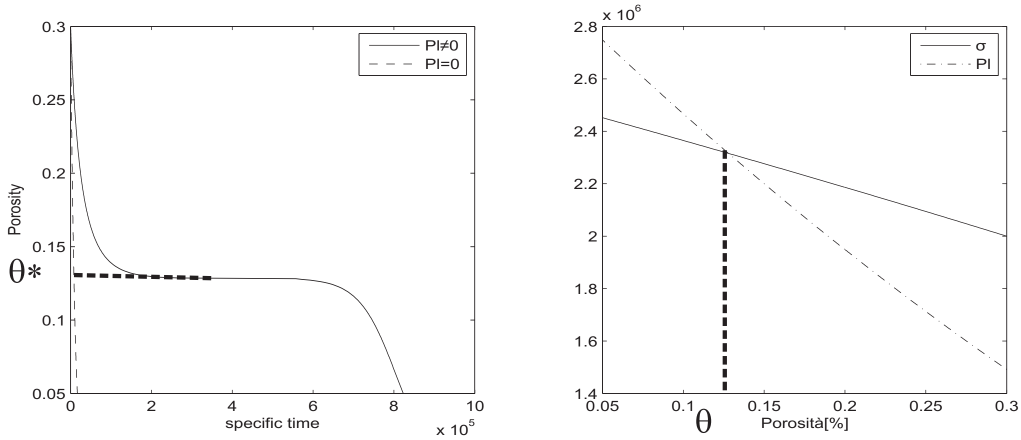

Figure 4: Evolution of porosity- $\sigma - p_l$ changes sign during the process

3. *$\sigma < p_l$ during the whole process.*
   This case may be summarized by analyzing Figure 5. Here, the gap between the curves is remarkable. In this case, the force driving the process is basically the Laplace pressure, simply because it is higher than the externally imposed stress.

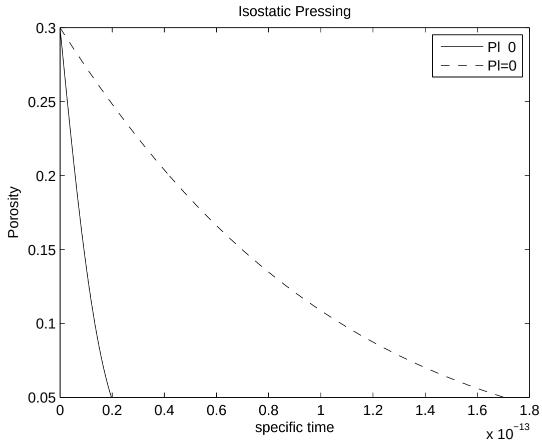

Figure 5: Evolution of porosity- $\sigma < p_l$ during the whole process

2.1 *Effect of the interstitial stress on the sintering time and on the residual porosity*

It is evident that the interstitial stress influences the evolution of the porosity and, in particular, the sintering time. It is worth emphasizing that threshold pressures may be defined. They are meant to be the values below which the sintering stress is actually not negligible; the duration of the process is indeed heavily affected by such a stress. In turn, such a duration would be underestimated otherwise. Fig. 6 shows the threshold pressures for a specific metallic alloy (aluminum-zinc-magnesium-copper alloy) for $1\mu m$ powder [8].

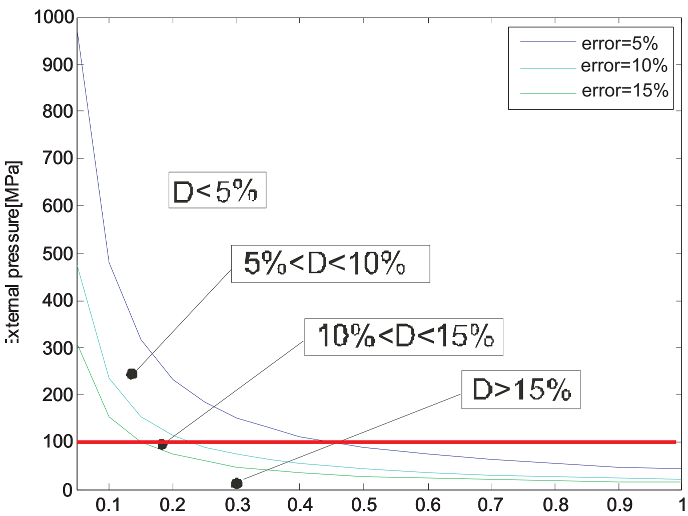

Figure 6: Threshold pressure p^{*}, for $1\mu m$ powder

Furthermore, industrial processes often entail loading pressures lower than the thresholds mentioned above, especially of "small" grain sizes.

Moreover, the sintering stress influences the value of the residual porosity, defined to be the one still present after a given time (roughly speaking, the duration of e standard process for the considered alloy). Such a value of the porosity is a fundamental feature of the actual material, because it is a key parameter that determines the mechanical properties of a sintered specimen. Figure 7 shows the error on the evaluation of the residual porosity, for a thirty minutes sintering process, with external loading pressure of 100MPa for a standard specimen of the given alloy, for different values of the grain size.

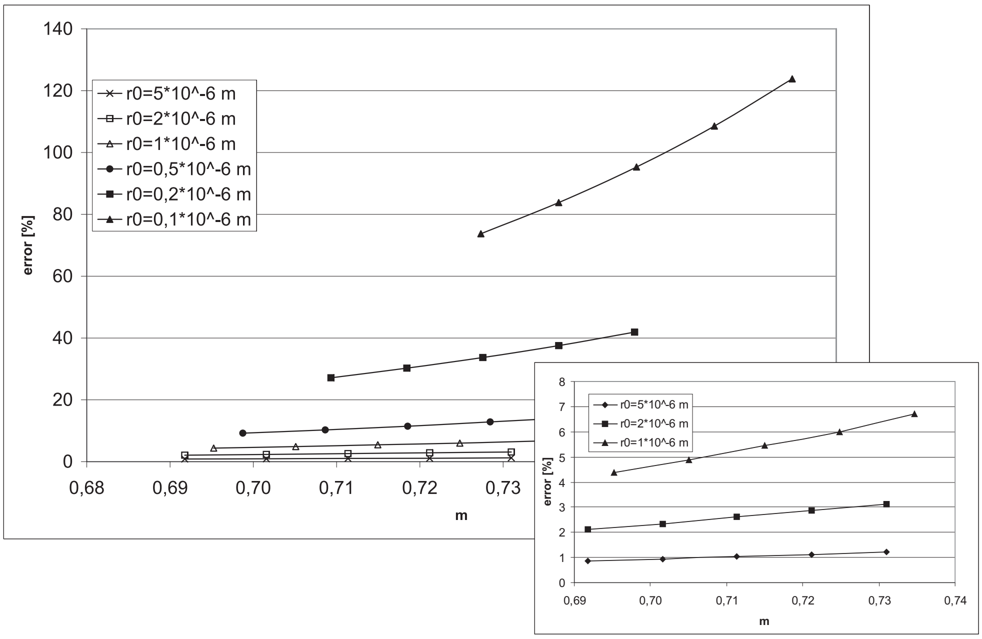

Figure 7: Errors $e\%$ on the residual porosity for different values of the grain size r_0

2.2 *Stability analysis*

As we discussed above, it is evident that the loading parameter may be tuned in such a way that, at some stage of the process, its value may equate the interstitial stress, leading to a constant value of the porosity. The stability of such a situation is studied through a perturbative approach, following [3]: a perturbed solution is considered in the form:

$$\begin{cases} \theta(t) = \theta_0(t) + \delta\theta \exp(\lambda(t-t_0)), \\ \sigma(t) = \sigma_0(t) + \delta\sigma \exp(\lambda(t-t_0)), \\ S(t) = S_0(t) + \delta S \exp(\lambda(t-t_0)), \\ p_{l0}(t) = P_{l0_0}(t) + \delta P_{l0} \exp(\lambda(t-t_0)); \end{cases} \tag{12}$$

where S is the cross-sectional area of the specimen, the symbol δ denotes the perturbation of the amplitude of the considered item and t_0 is the initial time for the process. By substituting (12) in the evolution law (10), in the relation between the cross sectional area and the porosity, and in the expression of the Laplace pressure (6) or (7), after linearization about the fundamental solution ($\theta_0(t)$, $\sigma_0(t)$, $S_0(t)$, $p_{l0_0}(t)$) we have, for the case of isostatic pressing:

$$\begin{bmatrix} 0 & S & \sigma & 0 \\ \frac{\lambda}{\dot\theta} + \frac{\frac{d}{d\theta}G(\theta)}{G(\theta)} & 0 & \frac{1}{S}\left[1 - \frac{\lambda}{\dot\theta G(\theta)}\right] & 0 \\ \frac{1}{\dot\theta}\frac{\partial f(\theta,\sigma,p_{l0})}{\partial\theta} - \frac{\lambda}{\dot\theta} & \frac{1}{\dot\theta}\frac{\partial f(\theta,\sigma,p_{l0})}{\partial\sigma} & 0 & \frac{1}{\dot\theta}\frac{\partial f(\theta,\sigma,p_{l0})}{\partial p_{l0}} \\ \frac{\partial p_l(\theta,p_{l0})}{\partial\theta} & 0 & 0 & \frac{\partial p_l(\theta,p_{l0})}{\partial p_{l0}} \end{bmatrix} \begin{bmatrix} \delta\theta \\ \delta\sigma \\ \delta S \\ \delta p_{l0} \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad (13)$$

where

$$\begin{cases} G(\theta) = \frac{2}{3(1-\theta)} \\ , f(\theta,\sigma,p_{l0}) = \dot\varepsilon_0(1-\theta)^{\frac{3m-1}{2m}}\left(\frac{|\sigma_z - p_l|}{A\sigma_0}\right)^{\frac{1}{m}}\psi^{\frac{-(1+m)}{2m}}. \end{cases} \quad (14)$$

Such a system has non-trivial solutions if and only if the determinant of the matrix in (13)is equal to zero. This condition leads to a second order characteristic equation with respect to the normalized perturbation growth rate $\frac{\lambda}{\dot\theta}$. Roots of such an equation are shown in the following figures for the different load modes.

- Isostatic pressing.

  It is easy to show that, for cases 1 and 3, the process is always stable. Figure 8 shows the roots $\frac{\lambda_1}{\dot\theta}$ and $\frac{\lambda_2}{\dot\theta}$ for the remaining case, i.e. the one in which the quantity $\sigma - p_l$ changes sign during the process. It is immediate to note that, whenever $\theta = \theta^*$ (which is achieved for $\sigma = p_l$) such roots have an asymptote; close to it, the roots λ_1 and λ_2 become real, both greater than zero. This means that, at the critical value of the porosity θ^*, the process is unstable. This condition allows for sintering to keep on going. Henceforth, in order to have stability of sintering either the loading parameter must be high enough with respect to the interstitial stress or zero, leading to (stable) free sintering.

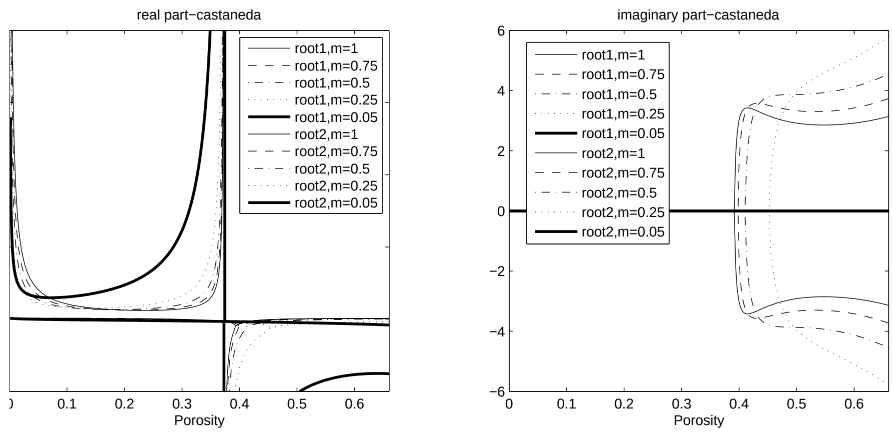

Figure 8: High order stability analysis, isostatic pressing

- Constrained forging.

  Since forging of samples is performed against rigid walls, there is no change of the cross section, leading to a first order problem with only one root. This turns out to be positive for the considered range of porosity and, hence, the process is linearly stable. It is evident from the graphs 9 that there is no asymptote correspondent to $\sigma = p_l$. In the case in which the quantity $\sigma - p_l$ changes sign during the process, the stability analysis allows us to conclude that at θ^* the process is stable (see figure 9.
  Then, such a value represents a critical threshold, below which the sintering process cannot proceed. The critical porosity, in this case, represents the lowest threshold under which the (averaged) longitudinal strain cannot evolve.

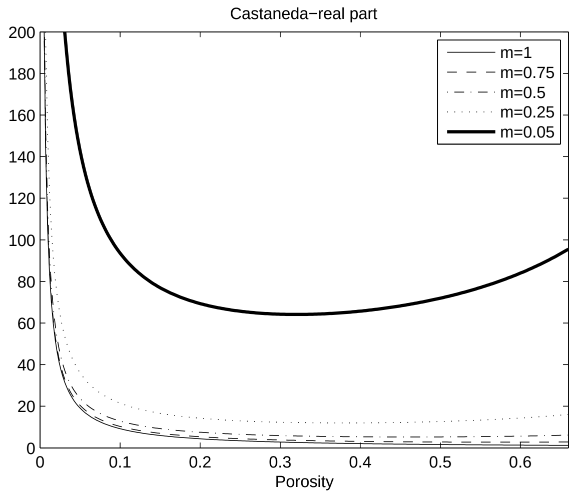

Figure 9: High order stability analysis, isostatic pressing

References

[1] Olevsky, E.A., Theory of sintering: from discrete to continuum. *Invited Review. Mater. Sci. Eng. Rep. Rev.* **23** (1998) 41-100.

[2] A. Molinari & E. A. Olevsky, Kinetics and stability in compressive and tensile loading of porous bodies.

[3] Olevsky, E.A., Molinari, A., Instability of sintering of porous bodies. *Int. J. Plast* **16** (2000) 1-37. *Mechanics of Materials* **38** (2006) 340-366

[4] L. Galuppi, Effects of the Laplace pressure during sintering of cylindrical specimen. In preparation.

[5] L. Galuppi & L. Deseri, Effects of the Laplace pressure on isostatic pressing and free sintering of cylindrical specimen. In preparation.

[6] L. Galuppi & L. Deseri, Effects of the Laplace pressure on free and constrained forging sintering processes of cylindrical specimen. In preparation.

[7] Skorohod, V.V., Rheological Basis of the Theory of Sintering. *Naukova Dumka, Kiev.* (1972) .

[8] E. Olevsky, H.J. Dudek, W.A. Kaisser: Hiping conditions for processing of metal matrix composites using the continuum theory for sintering-II. Application to fibre reinforced titanium alloys. *Acta Materialia*, Vol. **44**, No. 2, (1996) 715-724.

[9] Ashby, M.F., Background Reading, HIP 6.0. *University of Cambridge, Cambridge, UK.*