EFFECT OF THERMAL GRADIENT ON THE DENSITY DISTRIBUTION OF MACROPOROUS CERAMICS OBTAINED BY SPARK PLASMA SINTERING

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Summary

This work aims at correlating density heterogeneities observed in macroporous alumina ceramics synthetized by Spark Plasma Sintering (SPS) and thermal gradients during the sintering cycles.

Porous specimens are obtained by partial sintering of alumina spray-dried granules [1]. HP or SPS are compared. X-ray radiography and 3-dimensional X-ray computed tomography evidence that samples produced by SPS show significant density gradients. These density heterogeneities are assumed to be related to the thermal gradients in the granules packing during the sintering cycle.

In-situ evaluation of heat flow and temperature field within the sample during the sintering process is hardly possible as the measurement devices may influence the collected data. Therefore, the temperature field (inside the SPS tools and the compact) is evaluated by a coupled thermo-electric numerical model. For different sintering conditions, the density gradients, measured by 3-dimensional X-ray computed tomography, are in agreement with the thermal model.

Introduction

Porous specimens used in this work are obtained by partial sintering of controlled ceramic powder agglomerates [1]. This new method for porous ceramics synthesis lead to a hierarchical porous network that can contain up to tree levels of interconnected pores: i)- the voids existing between the agglomerates, ii)- the porosity remaining inside the agglomerates, iii)- the pores that may exist within the initial ceramic grains.

More specifically, in the present work, spray dried alumina granules are used as ceramic powder agglomerates. Granules packing are consolidated by Hot Pressing (HP) or Spark Plasma Sintering (SPS) in order to initiate “bridges” at the granules contact points (Figure 1). By comparison to HP, SPS allows a lowering in the maximum temperature cycle, which is favorable to the porosity rate [2]. However this paper highlights the heterogeneity of the so obtained specimens. Experimental density gradients are compared with a numerical model of the temperature field for different sintering conditions.

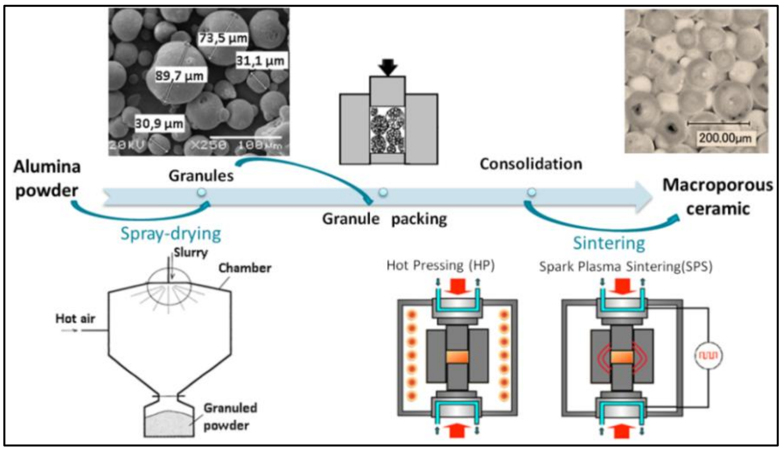

Figure 1: Principe pattern of the method

Experimental

Sample processing

Alumina granules (d_{50} = 80 µm), obtained by spray-drying of Alcan P172LSB powder (d_{50} = 0.41 µm), are used as ceramic powder agglomerates.

Partial sintering of granule’s packing is carried out by Spark Plasma Sintering with a SPS HPD10 from FCT systeme GmbH. The sintering conditions are:

- Die diameter = 40 mm
- Weight of powder = 40 g
- Heating ramp = 100°C.min^{-1}
- Maximum temperature = 1000°C
- Dwell time = 9 min
- Cooling ramp = 100°C.min^{-1}

Characterization by radiography and 3-dimensionial X-ray computed tomography (3D-CT)

Characterizations were carried out tanks to Microfocus X-ray Computer Tomography HMX 225 OIS Eng. Ltd.
This system used is composed of : a microfocus Tungsten X-ray source (5 µm diameter) 40-225 KeV; a CsI scintillation X-ray detection screen coupled with a photomultiplier and a CCD camera (1024x1024 pixels) ; a five axis manipulator (x, y, z, translations; y tilt; z rotation).
The X-ray tube setting was 80 kV for all tests. Cu filter (0.4 mm) was used to reduce beam hardening.

Measurement by conventional 2D projection radiography

At a first stage, measurements were preformed by 2D projection radiography. Average densities ρ along diameter at different heights of the cylinder and along the height at different points of the diameter were calculated (Figure 2).

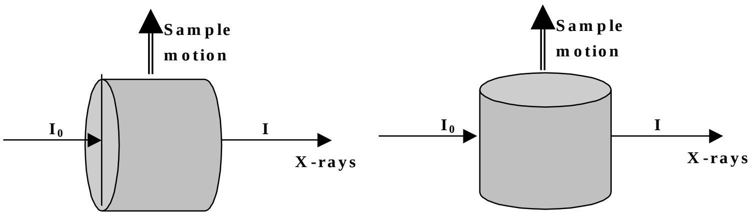

Figure 2: Principle of density measurement by 2D radiography

During a sample examination, the intensity I of the X-ray beam hitting the screen detector is attenuated with regard to the intensity I_0 when no sample is introduced along the beam path*:*

$$\frac{I}{I_0} = \exp\left[-\left(\frac{\mu}{\rho}\right)_t .\rho_t .t_t\right] \qquad \{1\}$$

t is thickness of crossed sample, $(\mu/\rho)_t$ is the mean mass absorption coefficient of sample along t.
In the CT System, the intensity I is converted into a N grey levels ranging between 0 (black, no radiation) and 4096 (white, saturation of the detector). As there is a linear dependence between the intensity and the grey level, densities can be calculated with reference to the average grey level $\overline{N}$ to which is associated the average density $\overline{\rho}$ of the sample:

$$(\rho - \overline{\rho}) = \frac{\ln\left[\frac{\overline{N}}{N}\right]}{\left(\mu/\rho\right).t} \qquad \{2\}$$

The mass absorption coefficient μ/ρ was experimentally determined on a homogenous reference specimen of known density.

Measurement by 3D-CT

In 3D-CT the grey levels associated to each voxel is proportional to the linear absorption coefficients μ = $(\mu/\rho).\rho$. Assuming a homogeneous chemical composition of the sample (μ/ρ = cste), the grey level is proportional to the local density. Calibration is done at the same time than the 3D-CT by using an internal reference of same chemical composition (same μ/ρ) but of different density. In the present case, this reference is also used as sample holder.

<u>Temperature field evaluation</u>

Direct evaluation of heat flow and temperature field within the sample is not experimentally possible. So, the temperature field is evaluated by a coupled thermo-electric numerical model of the SPS using the finite elements method. Knowing the electrical and thermal characteristics of materials and the limit conditions, this model allows the determination of the temperature field inside the tools and the sample.

Results and discussion

Figure 3 shows the average densities measured by 2D radiography ρ along 3 diameters at different heights. These measurements clearly show heterogeneities in the density with a maximum for the median plane of the specimen. These 2D measurements are in agreement with 3D-CT (Figure 4b).

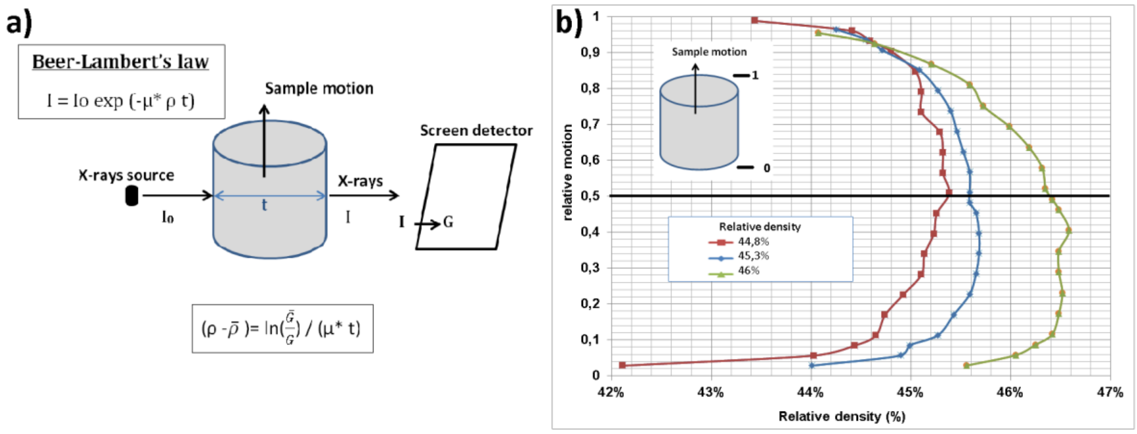

Figure 3: a) Principe of measurement of radiography; b) Relative density of SPSed samples obtained by radiography

The temperature field results given by the numerical model agree with the experimental results. This confirms our hypothesis that the gradient of density should come from the temperature gradients inside the sample. Indeed, high temperature areas predicted by the model correspond to the high density areas measured inside the sample.

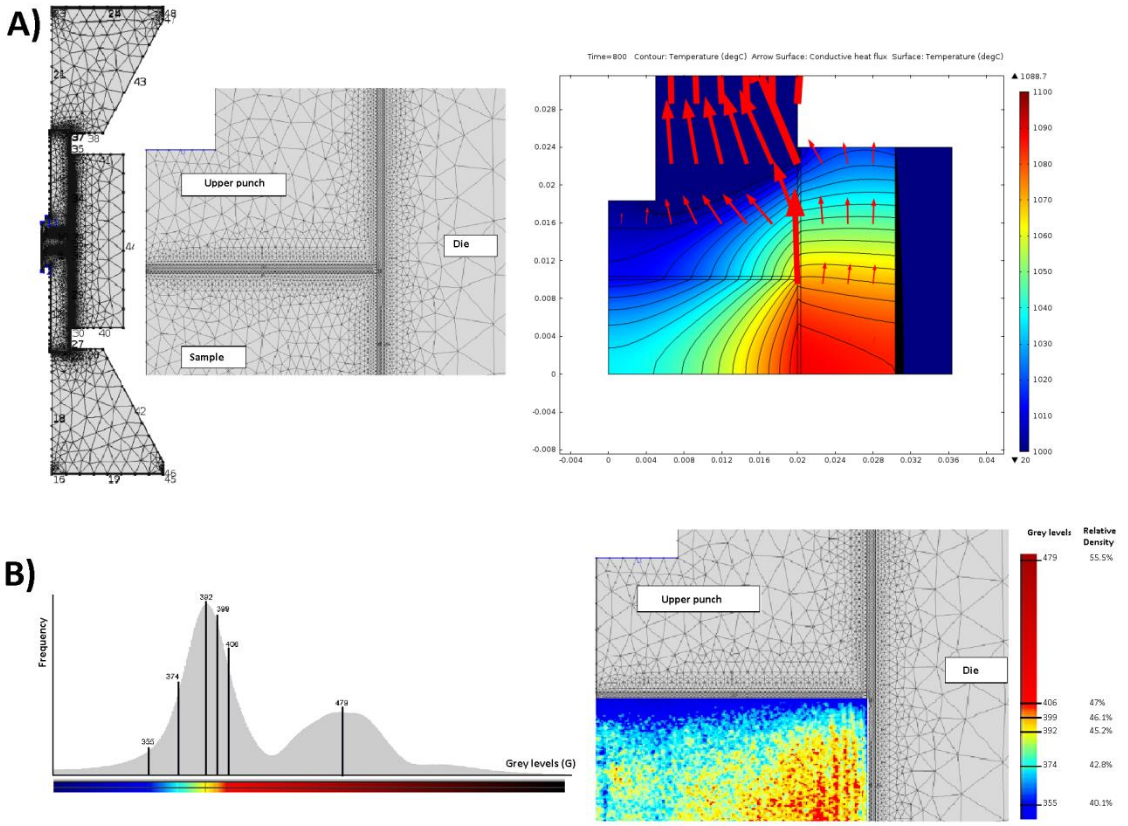

Figure 4: A) Temperature field and B) local density within the SPSed sample obtained by the numerical model and by 3-dimensional X-ray computed tomography (The first peak of the histogram corresponds to the sample while the second peak is related to the internal reference)

Conclusion

The presence of a density gradient in porous ceramics obtained by SPS has been confirmed by 2D X-ray radiography and 3-dimensional X-ray computed tomography (3D-CT). Temperature field within the sample has been evaluated by a coupled thermo-electric numerical model using the finite elements method. The results given by this model agree with the experimental measurements in density.

References

(1) G. Jean, V. Sciamanna, M. Gonon, M. Demuynck, F. Cambier*, Manufacture of Macroporous ceramics by Spark Plasma Sintering*, Proceedings of the 13th European Inter-Regional Conference on Ceramics, 2012,pp. 95-100, Barcelona, Spain

(2) G. Jean, V. Sciamanna, M. Gonon, M. Demuynck, F. Cambier*, Manufacture of Macroporous ceramics by Spark Plasma Sintering*, oral presentation, Shaping 5, 2013, Mons, Belgium

Type of presentation: poster contribution; student contest