Multiphysics Simulation of Microwave Sintering in a Monomode Cavity

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The simulation of sintering of a ceramic powder in a monomode microwave cavity has been carried out with COMSOL finite element software. At a given time, a stationary calculation provides the electromagnetic field in both the cavity and the compact when we assume an incident power, and when we suppose that dielectric permittivity and thermal parameters depend on relative density and temperature. From the electric field in the compact, the value of the generated heat is deduced, and a transient thermal calculation is then run with this value as a heat source and with radiating losses at the boundaries of the compact. The density of the compact is updated through a prescribed densification law, and finally, temperature and density kinetics are obtained. The developed tool is used to analyze the influence of the insulation device on heating. The effect of introducing a susceptor in the cavity for the purpose of hybrid heating is also investigated.

Introduction

Microwave heating of dielectric materials results from the absorption by molecule polarization of part of the energy transported by an oscillating electric field. As compared to conventional heating, it results in shorter heating times and thus may slow down unwanted microstructural changes arising during sintering, such as grain growth in fine grain ceramics. However, microwave sintering is a complex process, and is much more difficult to control than conventional sintering. Even such a basic issue as measuring the temperature of the material undergoing sintering is a problem. Also, insulation and positioning of a compact may be critical. Computer models representing the process as a whole would be of great help in the effort of understanding the microwave sintering process and bringing it to an industrial scale.

Microwave sintering involves several phenomena that are strongly coupled to each other: electromagnetism, heat transfer and sintering. For example, electromagnetic energy absorption that controls heating (and thus also controls sintering) depends on temperature-dependent material parameters and on the electromagnetic field, which changes as the sintering progresses. Taking into account such coupling effects is necessary for realistic modeling of microwave sintering. Macroscopic scale simulations of microwave sintering, coupling electromagnetism and heat transfer, have been presented in the literature [1-3]. These models are mainly based on either the finite element method or finite difference time domain techniques, and most of these studies do not introduce densification. Notable exceptions are the model by Birnboim and Carmel [4], who calculated density gradients in complex shape components, and the model by Riedel and Svoboda [5], who found density and grain size distributions in a cylindrical compact surrounded by a susceptor inside an axisymmetric resonant cavity.

We present in this paper several results of a 3D finite element simulation of microwave sintering in the monomode cavity furnace designed at Grenoble INP. This simulation takes into account electromagnetism, heat transfer and densification, as well as, in part, coupling of these phenomena. The simulation procedure is first described. Next, to demonstrate the interest of modeling, several examples of calculation are shown, including a simulation of sintering of a compact of zirconia, a dielectric material that absorbs microwave energy moderately at room temperature but significantly at sintering temperature. Emphasis is put on the influence of the insulation on temperature and density fields, and the question of hybrid heating is also discussed. More simulations can be found in [6].

Model Description

The model attempts to closely represent the conditions of a monomode cavity microwave furnace similar to the one developed at Grenoble INP by Charmond et al [7]. In this equipment, a rectangular waveguide transports microwave radiation to a rectangular TE_{10p} cavity of 86 x 43 mm cross-section. Located at one end of this cavity is a movable short-circuiting piston (an electric conductor) to reflect the radiation and create a standing wave; at the other end of the cavity (on the same side as the waveguide) is a movable iris consisting of a copper sheet with a vertical slot. When the distance between the short-circuiting piston and the iris is set correctly, backward radiation is reflected by the iris, and the stacking of forward and multiply-reflected microwaves results in an increase of the electromagnetic energy in the cavity (a resonance phenomenon). When the cavity is empty, the proper distance between the iris and the piston is a multiple of the half-wavelength in the waveguide. When the cavity contains a dielectric sample, the wavelength is modified, and thus this distance changes depending on material permittivity and sample dimensions. In this study, we simply simulate a standing wave in the cavity with a value of electromagnetic energy adjusted so that the sample reaches a prescribed temperature in the steady state; we do not introduce the coupling iris.

The iterative simulation process is described is as follows. At a given time, a stationary calculation provides the electromagnetic field everywhere in the cavity, including within the compact, and uses the electric field within the compact along with the imaginary part of the material permittivity to calculate the energy absorbed by the compact. This energy is considered as an internal heat source; taking into account heat flux out of the compact according to the assumed boundary conditions, we then calculate the resulting heterogeneous temperature variation for the next time increment. As a consequence of local temperature, a corresponding change in material density results; this change is calculated by integrating a known densification equation. This increase obviously results in a deformation of the compact – however, for the sake of simplicity, we assume that the dimensions of the compact do not change during the process. This means that the relative density is a local parameter that describes the microstructure of the material but does not affect the geometry of the sintering compact. We do not calculate the stresses and strains resulting from heterogeneous density variation.

This calculation procedure has been carried out in 3D with COMSOL Multiphysics finite element software. Three "models" have been used:

- Electromagnetics Waves (COMSOL model reference ("rfw") in harmonic propagation,
- Heat Transfer by Conduction ("ht") in transient analysis (in the compact only),
- Partial Differential Equations, General Form ("g") in transient analysis with the relative density as a variable.

We suppose that the complex permittivity in the compact is a function of temperature, T, and that the thermal conductivity of the compact is a function of the relative density, ρ_r. The densification rate is expressed as a function of temperature and relative density, and we suppose that it supposed roughly describes the behavior of submicronic zirconia powder.

The calculations presented in this paper have been run on a standard PC with 1.8 GHz CPU and 2 GB RAM. The computing time for the later calculations that involve all the phenomena and coupling was around 5 min.

Process and Material Parameters

The following parameters have been introduced in the simulation. Some of them have been taken from the literature, while others have been estimated intuitively. It should be emphasized that such dielectric parameters as permittivity are very poorly known: indeed, values with different orders of magnitude can be found in the literature. This, of course, is a serious issue for modeling and should be considered in future work, when we seek a quantitative description of microwave sintering.

Electromagnetic parameters:

- Relative permittivity of the air in the cavity: 1.0
- Relative permittivity of the compact: $\varepsilon_r = 10 - i0.1e^{0.0017.(T-293)}$, i.e., the real part of ε_r is constant and the imaginary part increases from 0.1 to 1.0 when the temperature increases from room temperature to 1,700 K.
- Input power in the cavity entry section is constant in every simulation, adjusted by trial and error to reach a prescribed temperature.
- Cavity walls are perfectly conducting.

Heat transfer parameters:

- Conductivity: $k = 30\rho_r$ W/(mK)
- Weight density: $\rho = 6{,}000\rho_r$ kg/m^3
- Heat capacity: $C_p = 900$ J/(kg K)
- Initial temperature: $T_0 = 293$ K
- Boundary conditions: radiative loss with emissivity equal to 0.1 or 0.9 (see below)

Densification parameters:

- Densification law: $\dfrac{d\rho_r}{dt}\dfrac{1}{\rho_r} = 200\,e^{-\frac{20000}{T}}\left(\dfrac{1-\rho_r}{\rho_r - 0.64}\right)^2$, describing significant densification around 1,700 K.
- Initial density: $\rho_{r0} = 0.65$

Process and Material Parameters

Figure 1 presents the electric field distribution in the cavity containing a 1 mm tall cylindrical sample of diameter 1 mm. For this simulation, a constant relative permittivity, $\varepsilon_r = 10 - i0.3$, has been assumed. Prior simulation of the empty cavity showed regular nodes and antinodes along the wave direction, as expected; the compact has been positioned at one of these peaks so that it absorbs as much energy as possible. Presence of the compact does not significantly change the positions of nodes or antinodes, but the electric field inside the compact is very low (about 15%

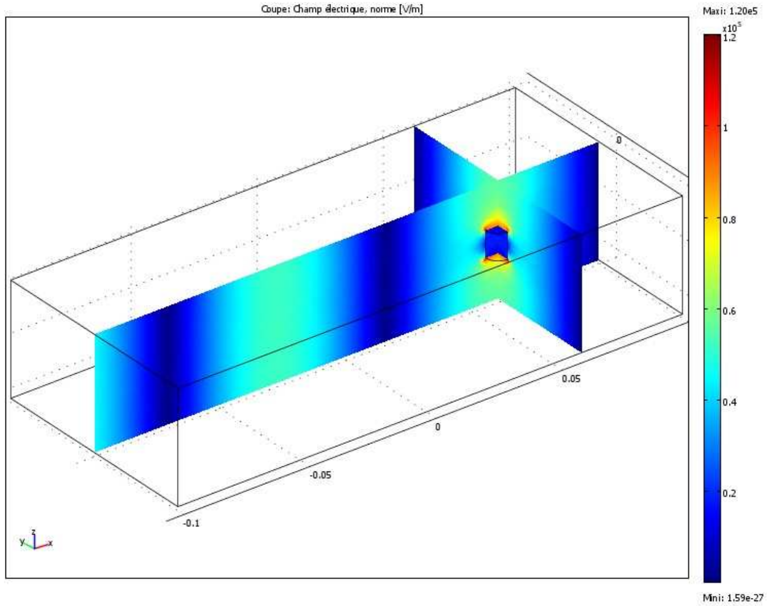

Fig. 1. Electric field norm distribution in the plain cavity containing a cylindrical compact. The electric field ranges from 0 to 1.2 10^5 V/m.

of the field found in the empty cavity) due to the high relative permittivity of zirconia compared to that of air, and to the strong gradient observed in the air below and above the compact. This means that the energy absorbed by the compact will be much lower than from the energy absorbed in an empty cavity situation; thus, a higher electromagnetic power will be required in the cavity to heat the compact, which is practically obtained due to the resonance. The gradient may also lead to detrimental phenomena, such as plasma formation.

Next, we will show results of multiphysics simulations. We investigated the effect of compact insulation on temperature and density gradients. In the first case, the 1 mm height, 1 mm diameter cylindrical compact is insulated on every side (an emissivity of 0.1 is assumed), whereas in the second case it is insulated everywhere except on its upper surface (an emissivity of 0.9 is assumed). The second case corresponds to experiments of Charmond et al. [7], during which the temperature in the upper surface is measured by an infrared camera. The input power in every experiment is adjusted so that the average temperature after 30 min is about 1,680 K. Figure 2 shows the temperature on the surface of the compact in both cases. With full insulation, the temperature at the center of the compact is found to be higher by 16 K than the temperature of the edges of the upper and side surfaces. With partial insulation, the temperature distribution is asymmetrical, with the maximum temperature in the lower part of the compact being 57 K higher than the minimum in the upper part. This second case has been more deeply investigated. We plotted in Figure 3 the changes of temperature and relative density during sintering in three points of the compact: the center of the lower section (called "bottom"), the center of the compact ("center"), and the center of the upper section ("top"). It can be observed that the heating rate is about 200 K/min and the temperature reaches a peak before a slight decrease. This variation results from the assumed constant input power, and could be controlled by adjusting this power throughout the simulation. Figure 3 shows that the bottom and center temperatures are about equal, and the temperature at the top separate around 1,300 K. This difference leads to a variation of about 1% in density.

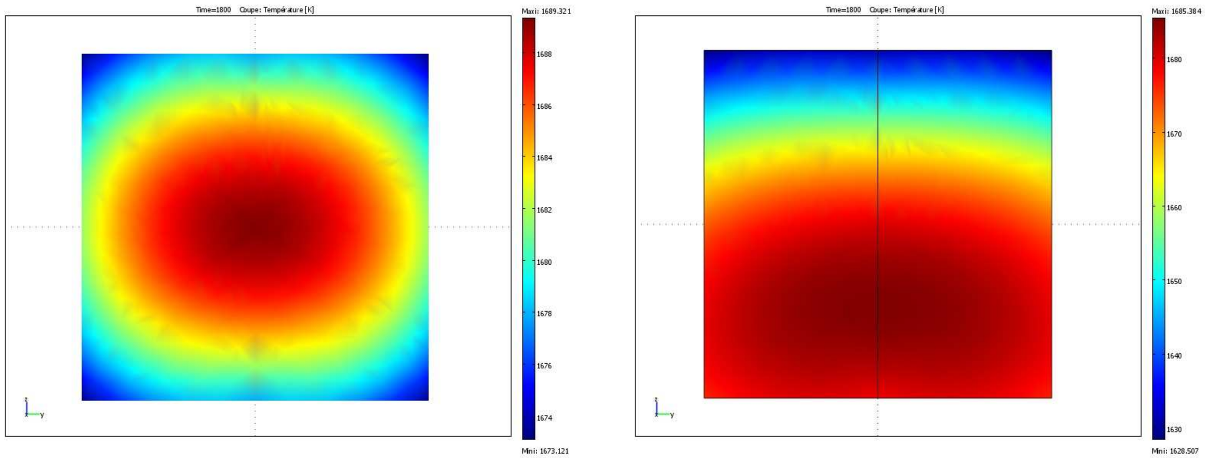

Fig. 2. Temperature distribution in a vertical cross-section of the compact after 30 min heating. The compact surrounded by thermal insulation on every side (a), and on every side except the upper surface (b). The temperature ranges from 1,673 to 1,693 K in (a) and from 1,628 to 1,685 K in (b).

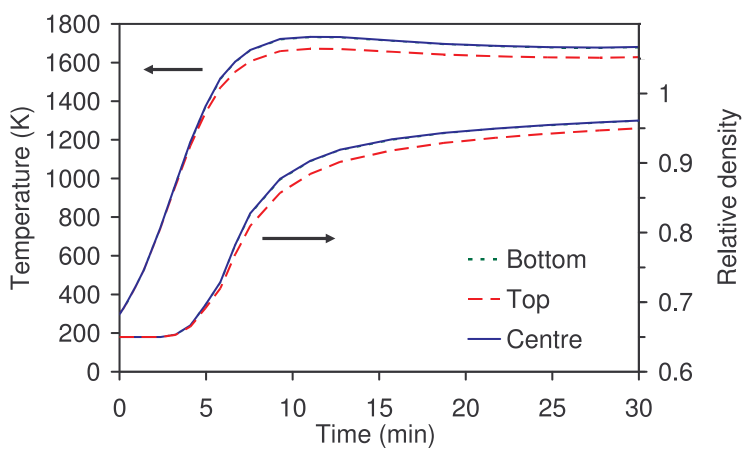

Fig. 3. Temperature and relative density changes during sintering of the partially-insulated compact. The curves corresponding to “center” and “bottom” temperature and relative density coincide.

Finally, we present results of simulating sintering of a SiC cylindrical susceptor (height: 15 mm, external diameter: 30 mm, thickness: 5 mm, relative permittivity: 100 – *i*100) surrounding the compact. Such a configuration is frequently used in experimentation to preheat materials with low permittivity at room temperature, and to sinter materials in case of poor coupling with microwaves in sintering temperature range. A relevant question is whether the susceptor shields the compact from the electric field, which would prevent hybrid sintering. Figure 4 shows that the electric field is lower in the susceptor than in the compact due to the higher real permittivity of SiC. The dissipated power in the susceptor is much higher than that in the compact (296 W vs. 2.3 W) due to the higher imaginary component of electric permittivity of SiC. Our calculation shows that the susceptor does not shield the compact from the field. Thus the compact can undergo hybrid heating, i.e. radiation coming from the susceptor and coupling with the microwaves.

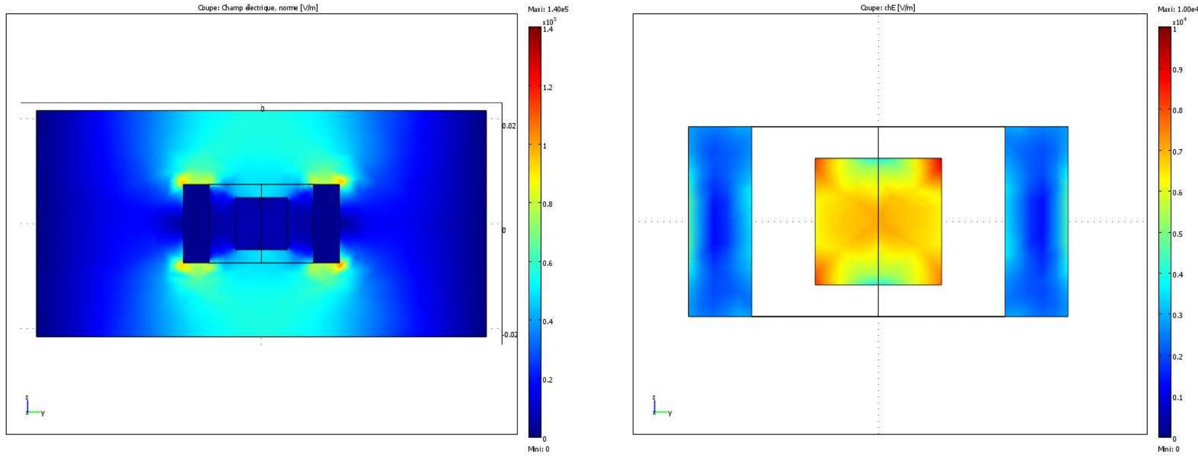

Fig. 4. Distribution of the electric field norm in a plane normal to wave propagation in the whole cavity (a), and in the compact and susceptor only (b). Range of electric field: 0-1.4 10^5 V/m in (a) and 0-10^4 V/m in (b).

Conclusion

Interest in using finite element simulation to aid in better understanding microwave sintering in monomode cavities has been demonstrated. We first calculated the distribution of electromagnetic field in the cavity containing a typical cylindrical ceramic compact; next we showed the temperature and density gradients in a compact with complete or partial thermal insulation, and finally, we investigated the electric field with a susceptor surrounding the compact and found that the compact can successfully undergo hybrid heating. We are currently working on simulating such hybrid heating by taking into account radiation from the susceptor to the compact.

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