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Numerical and Experimental Study of Polymers Microwaves Heating

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Abstract. Heating polymers by microwaves is not common. In order to quantify the ability of microwaves to heat different type of polymers, a bench measuring the dielectrics characteristics of polymers is built, and a numerical modeling of the waves- polymers interaction is achieved.
Finite elements method is introduced to solve the Maxwell equations [6-8] and the energy equation. A series of numerical simulations test are performed: convergence tests are realized and the heating power distribution in the sample is obtained. Using the power distribution, we studied a transient heating process of a polymer (CAPA) sample by microwave.

In order to validate the numerical approach, we set-up an experimental bench to heat the sample by microwave and measure temperature distribution, using optical fiber and pyrometers. A vector analyzer (VNA) [6, 9-12] is used to check the quality of the microwave cavity.

The results of the numerical simulations of the wave propagation are presented and the amplitude of the electric field is compared to the experimental measurement: good agreement is observed. In terms of polymer heating, numerical results of temperature field and experimental measurements are also compared: an efficient heating is observed.

Introduction

The conventional heating method actually used in polymer processing such as a ceramic band heater is inefficient and energy-consuming, improving polymer heating has thus gained increasing attention over the past two decades. One of the solutions could be the use of microwaves to heat polymers. The electromagnetic (EM) wave has the ability to penetrate and even pass through most polymers while inducing a form of energy transformation due to molecular polarization and leading to rapid volumetric heating. Studies on various materials [1-5] have shown reduction of energy consumption compared to the traditional method on the one hand and on the other hand improved microstructures and material properties. Consequently, the study of microwave heating of polymers becomes an important topic. The main problem related to microwave heating processes lies in the non-uniform temperature distribution within the processed material [13]. This non-uniformity highly depends on the physical and geometrical properties of the product and also on its dielectric properties [14].

Experimental Study

The objective of this work is to numerically model the heating of a small piece of polymer irradiated for several minutes with different microwave powers. Pyrometers and optical fiber are used to monitor the temperature at the surface of the sample, and the measurement are used for the validation of the numerical model. The reflection coefficient will also be measured and compared to numerical cases on the entry point of the microwaves.

The measurement sample will be of three types named here only by three codes PCL (Polycaprolactone) 6100, 6250 and 6500. The samples are in the form of a thin disc, placed on a cylindrical Teflon support. *Details are not given here for confidential reasons.*

The polymer sample will be placed on a cylindrical PTFE support. For an optimal heating, the position of all the polymers in the waveguide will be where the electric intensity would be maximum (on a peak of electric fields as a function of the guided length of the wave λ_g. (Fig.1)

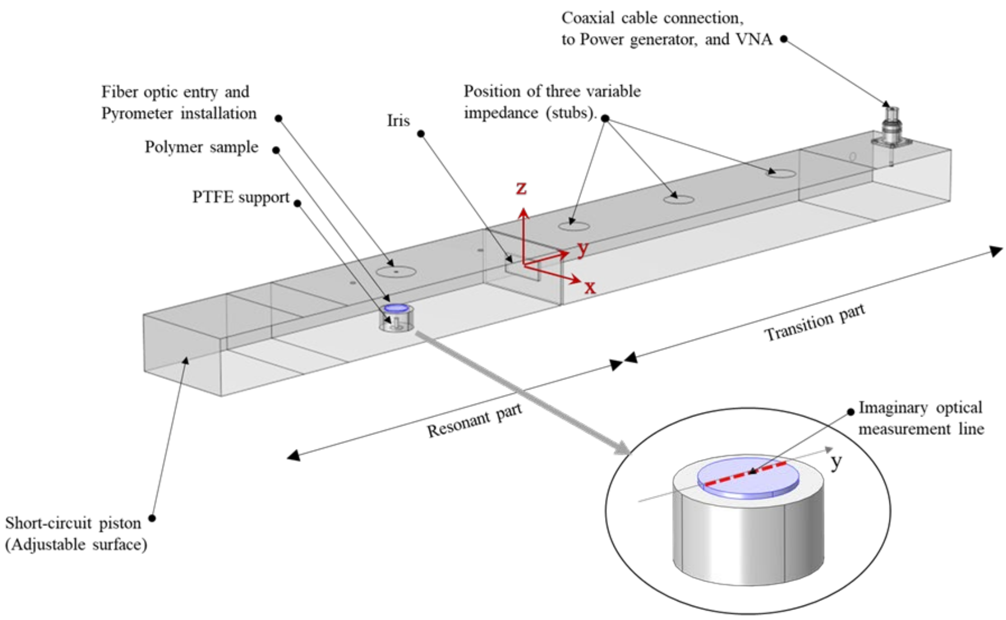

Fig.1 Experimental bench of the single-mode resonant cavity.

In order to guarantee a minimum temperature gradient in the material, a thin polymer disc is used, which imply us to assume that there will not be a large temperature gradient inside the polymer relative to the surface. This will give more confidence in the temperature measurement and allow comparison with the numerical results, as all the temperature measurements for validation are taken on the sample surface by radiative technics. The calculated Biot number for sample and sample holder, in the measurement configuration are 0.015 and 0.08, respectively, which satisfies the condition of a *thin thermal body*. Table 1.

Governing Equations

In this study, the objective is to model heat transfer in polymers under the influence of an EM field. We assume the heating of a sample of polymers, a Polycaprolactone (PCL) with three different concentration levels, positioned inside an Electric field propagated in a waveguide. The polymer will be considered as dielectric with measured complex EM property.

Numerical model of microwaves propagation in a dielectric medium is defined by Maxwell's equations. We opt for the hypothesis of a harmonic propagation in order to separate the dependence of the phenomenon on time. Accordingly, we can use the Helmholtz equation for a frequency regime.

Electromagnetic Modeling. The Maxwell's equations for electromagnetic wave, in its harmonic and time independent form [6, 7, 18], in the used cavity configuration can be written as,

$$\nabla \times \left(\mu_r^{-1} \nabla \times \tilde{\vec{E}}\right) - \gamma^2 \tilde{\vec{E}} = 0 . \tag{1}$$

where γ is the constant of propagation given as:

$$\gamma^2 = \left(\omega\sqrt{\varepsilon_0 \mu_0}\right)^2 . \left(\epsilon_r' - \frac{j\sigma}{\omega\varepsilon_0}\right) . \tag{2}$$

with ε_0 and μ_0 are respectively electrical permittivity and permeability of vacuum. Electric conductivity σ is linked to the pulsation ω of the wave, relative permittivity ε_r and permeability μ_r of dialectic medium (lossy material) are defined by:

$$\sigma = \omega\varepsilon_0\varepsilon_r''. \tag{3}$$

$$\varepsilon = \varepsilon_0\varepsilon_r, \quad \mu = \mu_0\mu_r. \tag{4}$$

The dielectric properties are function of temperature. (Cf. Fig.3).

$$\varepsilon_r(T) = \frac{\varepsilon'(T)}{\varepsilon_0} - j\frac{\varepsilon''(T)}{\varepsilon_0}. \tag{5}$$

<u>*Boundary and initial conditions:*</u> the cavity structural material is generally Aluminum, with electrical conductivity $\sigma = 3.03e7\ [S/m]$. Assuming the walls of the rectangular cavity are perfect electric conductors hence;

$$\vec{n} \times \tilde{\vec{E}} = 0. \tag{6}$$

the tangential component of the electric field was set to zero, also at time zero, the electrical field inside the cavity was initialized at zero. The source of electromagnetic power could be simulated with Eq.7 in one side of the wave guide. [8, 16]

$$\tilde{\vec{E}}_{input} = \tilde{\vec{E}}_0 \cos\frac{\pi x}{L_x}, \tag{7}$$

x represents the location in the cavity larger dimension:

$$\tilde{\vec{E}}_0 = \sqrt{\left(\frac{4Z_{TE}P_{in}}{L_xL_z}\right)}\ . \tag{8}$$

The dimensions of the waveguide section, $L_x \times L_z = 86.36 \times 43.18\ [mm^2]$. Z_{TE} is the wave impedance, and $\tilde{\vec{E}}_0$ electric field amplitude being a function of input power, according to the Poynting theorem (Conservation of energy in electromagnetic field).

Heat transfer modeling. The numerical resolution of heat transfer, unlike from Electromagnetic, was limited to the polymer sample and its PTFE support. The general energy equation has been used to describe the heat transfer in the system.

$$\rho c_p\frac{\partial T}{\partial t} = \nabla.(\lambda\nabla T) + Q_{EM}. \tag{9}$$

$$Q_{EM} = \frac{1}{2}\omega\varepsilon_0\varepsilon_r''.\left|\tilde{\vec{E}}\right|^2\ . \tag{10}$$

Where ρ, Cp and λ are respectively the density, the heat capacity and the thermal conductivity of the different mediums under consideration. (here polymer sample and PTFE support). Q_{EM} The heat generation due to microwaves is determined from the local electric field, computed and updated at each thermal time step. Note that, considering the permittivity of materials, this quantity for Teflon will be low.

The temperature evolution within the polymer sample is described from the general heat equation which depends on electromagnetic properties of the sample.

Boundary and initial conditions: At time zero, we considered a homogenous distribution of temperature inside the polymer and support: $T_0 = 30\ °C$. On all the external walls of the polymer and PTFE, we impose a Robin boundary condition Eq.11, (convection) with $h = 6\left[\frac{w}{m^2.K}\right]$ and $T_\infty = 18\ °C$ (considering natural convection for a flat surface, at atmospheric pressure and at temperatures close to ambient.).

$$-\lambda\frac{\partial T}{\partial \vec{n}} = h(T - T_\infty). \qquad (11)$$

Thermo electromagnetic parameters

Generally, the simulations of microwave heating are performed with the assumption of isothermal thermo electromagnetic properties, but in the study of the polymer we must precisely characterize this dependence; the variation of heat capacity and permittivity as a function of temperature, because during phase change of the polymer we will have an abrupt change which can influence the temperature follow-up on the whole sample. To that end, for three different concentration levels of PCL (CAPA6100, 6250 and 6500), the dielectric properties of the samples as function of temperature were evaluated previously via cavity perturbation method [9] and the heat capacity, is obtained using isothermal and no-isothermal DSC experiments, (Fig. 2). The thermal conductivity was obtained from literature.

We observe a strong variation of permittivity and heat capacity versus temperature, especially during the phase change range of temperature. (See Fig.2, Fig.3).

Indeed, in order to model this part of phase change in the energy equation, either we have to add another source of energy in the form of kinetics of crystallization, or by inducing an apparent thermal capacity. Using the DSC diagram data and doing a spline interpolation, we considered the dependence of Cp on temperature [15].

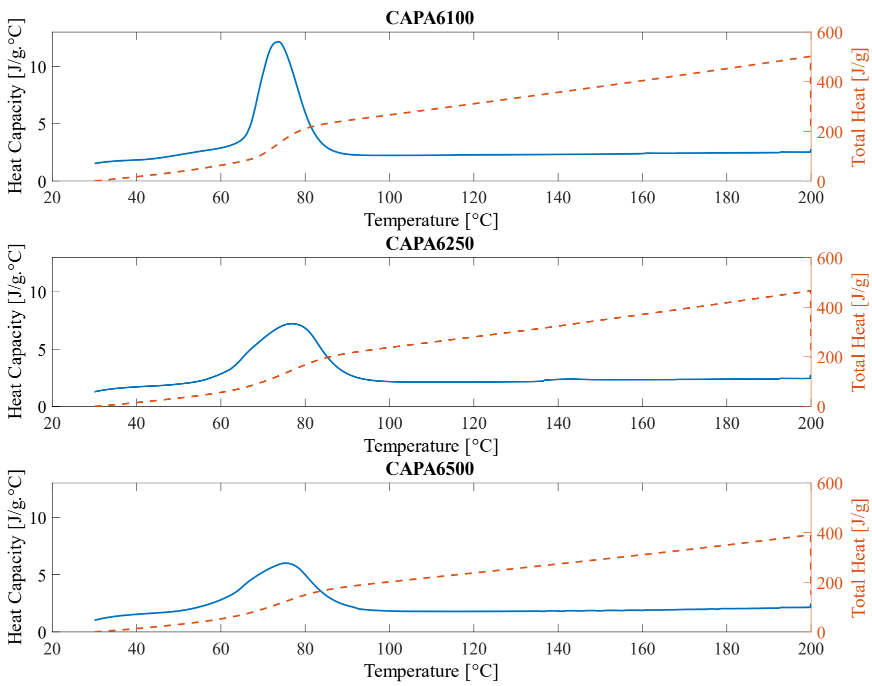

Fig.2 Result of DSC measurements on three different PCL.
In order to obtain the variation of Cp (T).

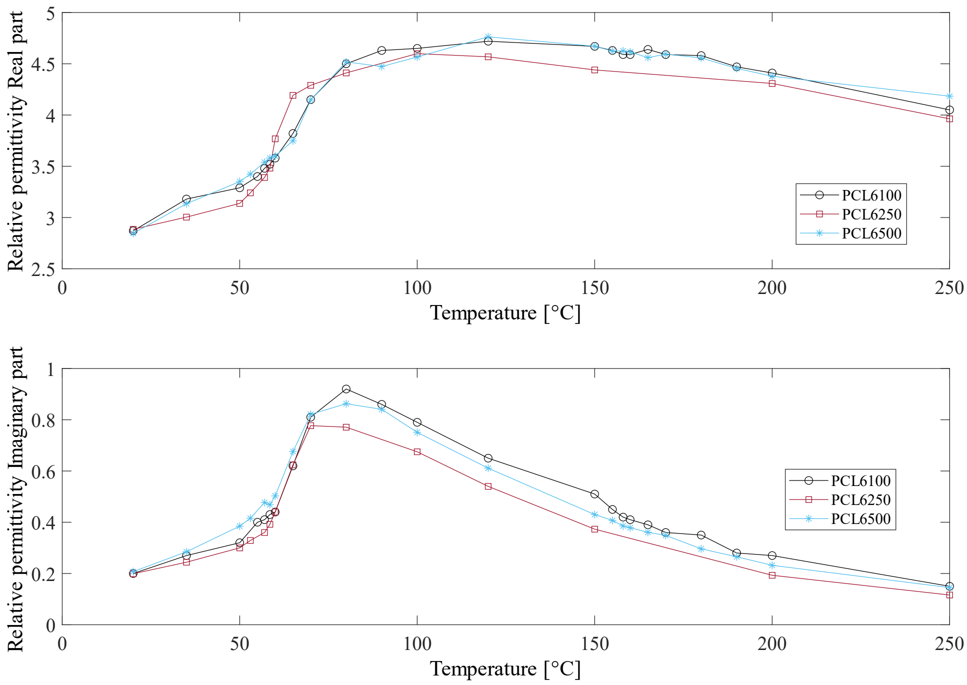

Fig.3 Dielectric properties of three kinds of PLC as function of temperature.

The other thermophysical properties of the polymer are considered constant. The detail was showed in Table 1.

Table 1. Thermo electromagnetic properties of the polymers and PTFE.

Properties	PTFE	PCL 6100, 6250, 6500
Density $\left[\frac{Kg}{m^3}\right]$	2180	1145
Heat capacity $\left[\frac{J}{kg.K}\right]$	1000	(Fig. 2) Cp(T)
Thermal conductivity $\left[\frac{W}{m.K}\right]$	0.25	0.18
Permittivity ε_r'	2.1	(Fig. 3) $\varepsilon_r'(T)$
Permittivity ε_r''	4e-4	(Fig. 3) $\varepsilon_r''(T)$
Permeability μ_r	1	1

Numerical Solution

The electromagnetic and the energy equation model were fully coupled. The flow chart of coupling procedure was showed in (Fig. 4). The Maxwell's equations Eq. 1-4 are solved using the finite elements methods (harmonic mode) with boundary conditions given by Eq. 6–8. The maximum element sizes (spatial resolution) of each tetrahedral element are limited to 0.05 of the free space wavelengths at 2.45 GHz (0.122 m), by defining the following relation [16, 17],

$$Max.element < \frac{0.122}{20*\sqrt{\varepsilon_r}}. \tag{12}$$

The time step for the resolution of the energy equation has been fixed with the BDF solver uses backward differentiation formulas with maximum order of 2. In order to insure the best numerical precision, the convergence criterion was fixed at 0.001.

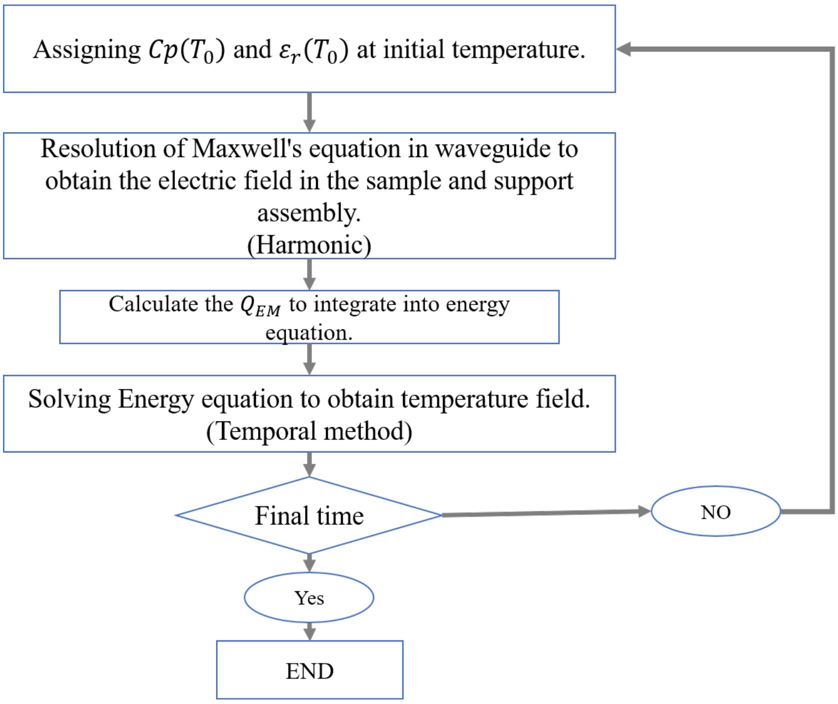

Fig.4 Flow chart for coupling of electromagnetism and heat transfer

Result and Discussion

Considering that the wave propagation direction in the cavity is Y (all along the waveguide), the electric field component with the dominant amplitude will be in the E_z direction. Therefor the electric field strength following the microwave propagation is thoroughly analyzed in order to quantify the influence of the polymer on the electric field distribution comparing with the empty cavity. (see Fig.5). The numerical results of electrical field are presented in terms of absolute value (norm).

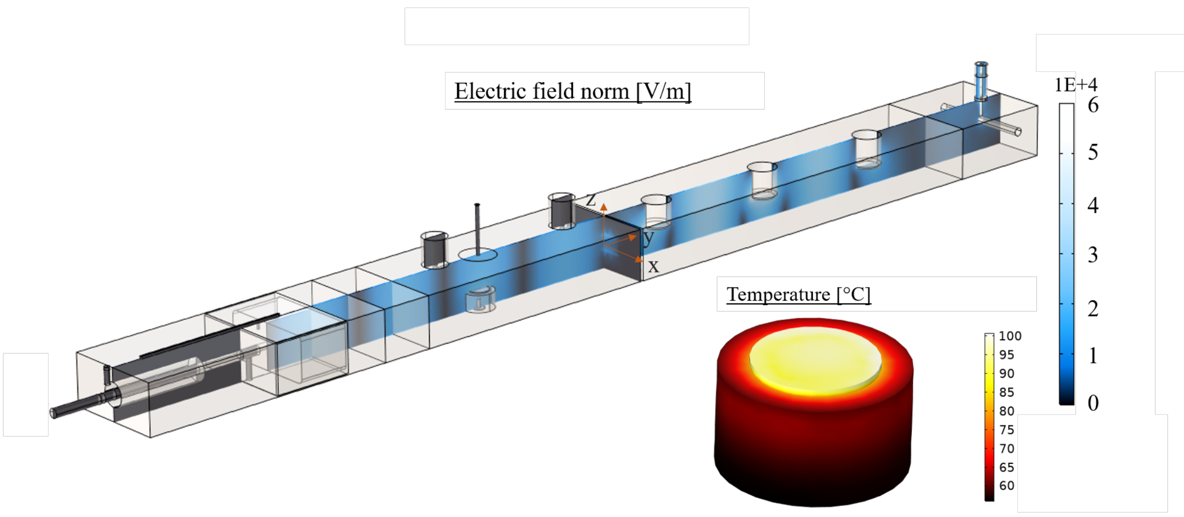

Fig.5 Electric field distribution [v/m], all along the YZ plane, with the presence of a sample in the resonant part. f=2.46 [GHz], P_{in}=20watt

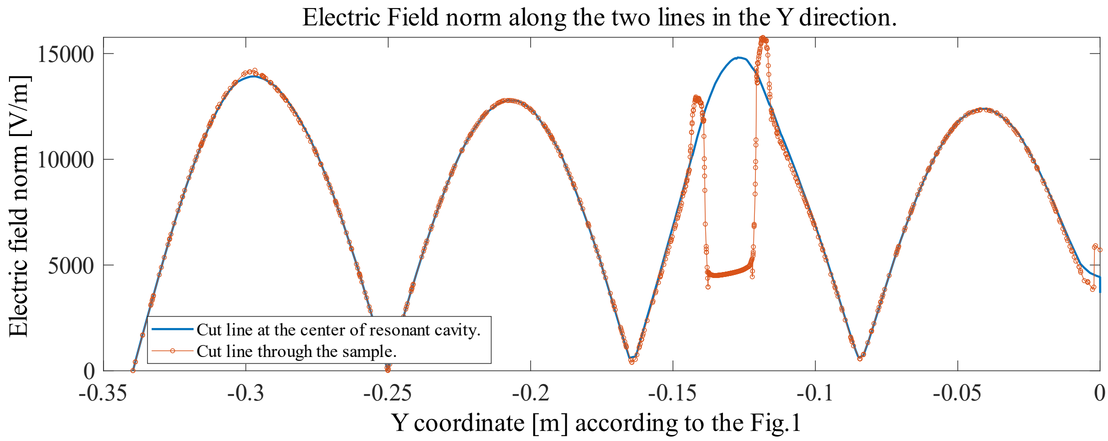

Fig.6 Cut lines crossing the center of the cavity between the iris and the short circuit piston. at two different heights, f = 2.46 GHz, P_{in}=20 Watt.

Overall the electric field strength near the top surface of polymer is attenuated (magnitude reduced approximately 40%), due to the high relative permittivity of the sample with respect to Teflon and also air (see Fig.6). Using a VNA, the reflection coefficient S_{11} was measured on the port (the coaxial cable input) and compared to the numerically calculated one. Consequently, the electric field amplitude inside the cavity will be close to that of the numerical model because we have found a good agreement between the two values. ie; Lower than S_{11} = -20 dB. (Fig.7).

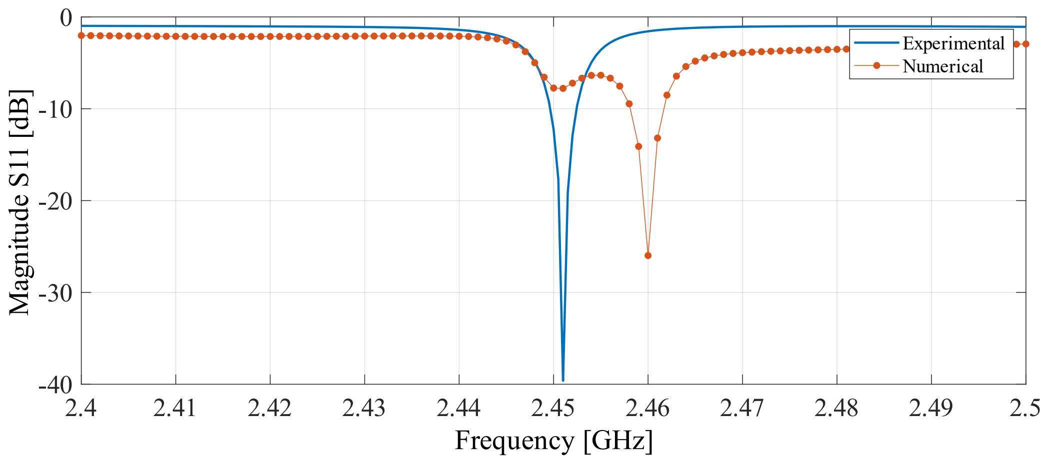

Fig.7 Calculation of reflection coefficient on the power input port in both numerical and experimental cases.

Several series of measurements were carried out on the three studied polymers. For two different incidence powers 20 and 60 Watt, the temperature was recorded using an optical fiber positioned on the sample center and thus the diagonal temperature profile, on a line shown in (Fig. 1) was evaluated and presented in Fig. 8, Fig. 9 and Fig. 10, with their respective comparison to the calculated ones.

Simulated temperature evolutions for Pin = 60 W are quite similar to the experimental result but a deviation is manifested in all cases during the melting step, probably due to poor precision in permittivity measurements at this step and also on the surface emissivity of the material during this phase, which can affect strongly the temperature estimated by the pyrometers, as the radiative emissivity is needed for the temperature conversion.

Considering that, to correctly measure the temperature by pyrometer, one would need a prior estimation of the apparent emissivity of the polymer which also varies during melting, we had carried out a calibration as a function of temperature taken by optical fiber. However, this step of measurement is always approximate and strongly related to the position of the thermal apparatus. Work is in progress to better master these evolutions.

Nevertheless, the comparison between numerical results and experimental measurements gives a fairly good agreement. The maximum temperature is located numerically near the center which agrees with the profile of EM Field. (Fig.6).

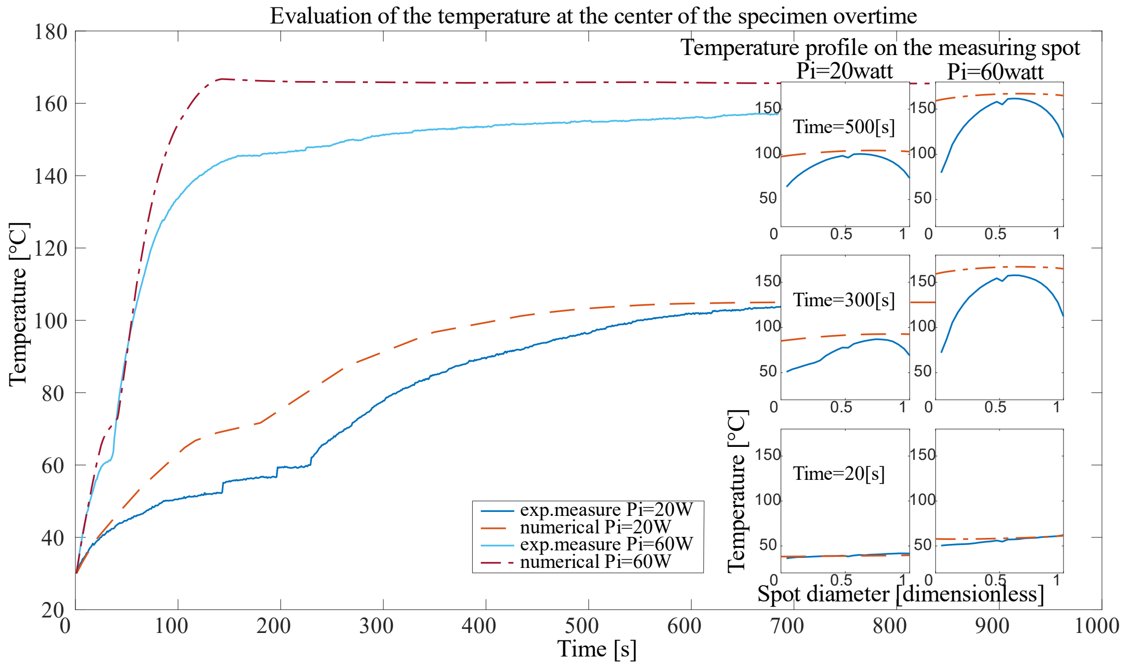

Fig.8. Temperature monitoring at center of the CAPA6100 sample and temperature profiles on the sample diameter for two heating power 20 and 60 watt compared with numerical simulation.

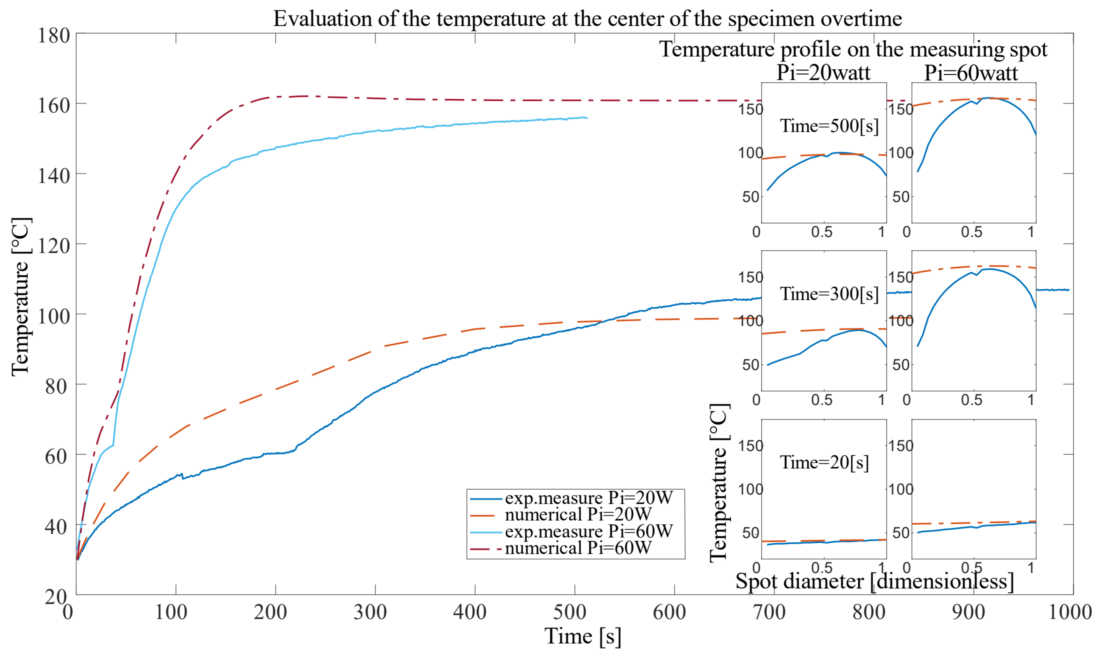

Fig.9. Temperature monitoring at center of the CAPA6250 sample and temperature profiles on the sample diameter for two heating power 20 and 60 watt compared with numerical simulation.

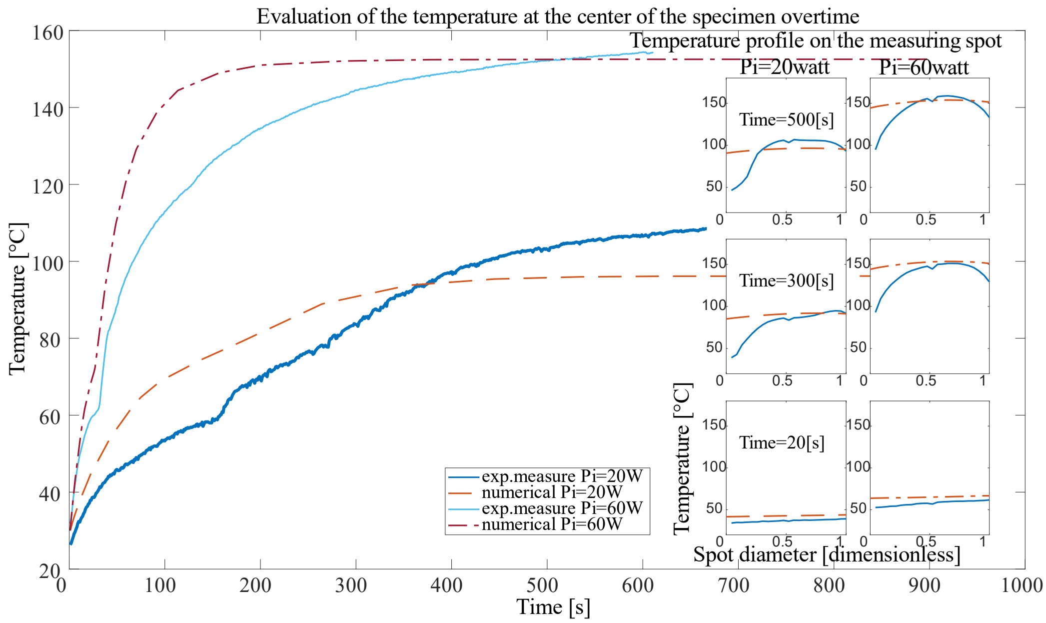

Fig.10. Temperature monitoring at center of the CAPA6500 sample and temperature profiles on the sample diameter for two heating power 20 and 60 watt compared with numerical simulation.

Conclusions

The most important information of this study can be summarized by the following points:

A model of coupling between electromagnetic and heat transfer was carried out in the case of a rectangular cavity. The two measurable parameters of the system in the experimental case were used in order to establish a comparison between the numerical and experimental results. The parameter S_{11} has been measured and confirms the absorption and the interaction of polymer with the wave.

The sample temperature is estimated at the center and also on a cross line passing through the center of the sample, by pyrometer and optical fiber.

To correctly model the temperature monitoring and especially the melting part, we opted for an apparent thermal capacity model. The EM properties of the three samples were previously obtained via the cavity perturbation method. These two parameters vary drastically during melting; it is crucial to define or identify a function describing the parameters evolution versus temperature, in order to better model the temperature evolution.

The sample size has been chosen such that it satisfies the condition of a *thermally thin element*, which allows us to consider that there will be a low temperature gradient in the polymer. However, the edge effect is present on the polymer disc which gives a difference in the measurement and model. The Robin condition of the convection at the edges was not sufficient to overcome this effect.

Finally, this study enables to build a bench helpful to compare theoretical or numerical models to experimental analysis of microwaves-polymer interaction, and constitutes a way to master the heat transfer phenomena in polymer under EM fields. Complementary results comparing to the experimental bench and based on our operating conditions.

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