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## Non-destructive characterization of refractories by mirage effect and photothermal microscopy

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**Abstract :** In this paper we will demonstrate how thermal waves have turned out to be a good probe for the thermal parameter determination, at various spatial scales for heterogeneous materials such as ceramics. After the detailed description of the two setups we have used ( mirage and photothermal microscope), we will explain how to proceed to achieve theoretical calculations either in a 1D geometry or in a 3D one. Then, we will illustrate our purpose with examples of refractory studies. Because these samples are porous and multiphase materials, they have to be investigated at macroscopic scale as well as at a microscopic scale. Finally, we will show results on the temporal behaviour of heat diffusion which can reveal how heat diffuses inside the sample.

### Introduction

Thermal wave probing has turned out to be a very interesting way to study various samples without much preparation. In this paper we want to demonstrate that this technique can be very fruitful for the investigation of ceramics. These heterogeneous materials are complex and their optimization requires the knowledge of physical parameters at various scales. It is well known that the macroscopic behaviour is strongly dependent upon the microscopic properties. Photothermal experiments seem to be good candidates for these studies.

In the first part, we will recall how to create thermal waves, how they can propagate in different media, and how they can be detected. Then, we will describe briefly how to solve the heat diffusion equation in various cases in 1D or cylindrical geometries.

In the second part, we will show results on various ceramics (carbon bonded magnesia and aluminium nitride) obtained with the two experimental setups described in the first section. Thermal diffusivity evaluations as well as photothermal imaging will be presented.

We will conclude with an extension of these photothermal experiments towards the study of heat diffusion dynamics.

### I Thermal parameter determination with a photothermal experiment.

#### a- photothermal experiment setups

A photothermal experiment is the "combination" of:

- illumination of the sample with a pulsed or modulated pump beam.
- detection of the surface or volume temperature variation related to the transformation of this radiant energy into heat.

This detection can be done with different experimental setups depending upon the physical parameters under investigation : refractive index, infrared emissivity, acoustic waves or local deformation... In this paper, we will use mainly two kinds of optical detection : the mirage effect for 1D experiments / 1 / and the photoreflectance for 3D configuration setups / 2 /, which both allow the detection of the surface or the bulk temperature variation with a very good sensitivity.

Fig.1 describes the mirage setup which allows the recording of the induced temperature variation of the sample. The sample is installed on a "mirage cell" / 3 / and illuminated with the pump beam ( laser beam or monochromator ). The heating of the surface leads to a temperature gradient which spreads in the transparent surroundings (air or transparent liquid) and in the sample. The temperature gradient is associated to a refractive index gradient, which can be measured by the deflection of an HeNe laser probe beam crossing it .The angular deflection of the probe beam is detected by a silicon position sensor. It is possible to reach the photon noise with this setup, (  $10^{-11}$  rad.Hz<sup>-1/2</sup> for the angular deviation which corresponds to  $10^{-5}$  °CHz<sup>-1/2</sup> for the sample surface temperature variation in air or  $10^{-7}$  °CHz<sup>-1/2</sup> if the surrounding medium is a transparent liquid such as CCl<sub>4</sub> ). With this kind of detection it is possible to detect thermal waves up to 100 kHz.

Fig. 2 shows a photoreflectance setup. The sample is illuminated with a focussed excitation spot and the probe beam is reflected at the surface of the sample, in the heated area. In order to obtain easily heated spots as small as a few micrometers, we use a commercial microscope to focus the heating beam. The probe beam is produced by a 670 nm diode laser. After reflexion on the area of modulated heating, the probe beam is received on a silicon photodiode coupled with a high-frequency lock-in amplifier. This experiment requires high modulation frequencies in order to prevent heat from diffusing on a large area, if an experiment on a small area or a photothermal image of good resolution are wanted. In this case, the measurement is performed by following the variation of the refractive index of the sample by means of the reflexion coefficient R. It is possible to achieve a sensitivity of  $10^{-6}$  in  $\Delta R/R$  with a frequency range spreading up to 20 MHz / 2, 4 /.

#### b - definition of thermal waves

In order to explain what is called a thermal wave, we have to solve the heat equation diffusion in a semiinfinite medium with a plane modulated heat source located at  $x = 0$ . The solution is the following :

$$T(x,t) = T_0 \exp(-x/\mu) \exp(j [\omega t - x/\mu ] ) \quad (1)$$

where  $T(x,t)$  is the temperature at a distance  $x$  from the source and  $T_0$  is related to the amplitude of the heat source;  $\mu$  is the thermal diffusion length which characterizes the heat propagation in the medium and which can be controlled by the modulation frequency  $f = \omega/2\pi$  of the heat source.  $\mu$  is related to  $k$  (thermal conductivity), to  $\rho$  (density),  $C$  (specific heat) and  $f$  by the following expression:

$$\mu = ( k / 2 \pi f \rho C )^{1/2} = ( D / 2 \pi f )^{1/2}$$

where  $D$  is the thermal diffusivity. Expression (1) could describe the propagation of an optical wave for which the real part and the imaginary part of the refractive index would be equal. So, although a thermal wave is strongly attenuated, it can reflect, diffract and propagate.

Moreover, eq (1) clearly indicates that measuring the phaselag of the complex temperature allows the localization of heat sources in the sample.

To know the temperature distribution in the sample and in its surroundings we have to solve the heat equation diffusion in all the media (fluid, sample and backing). If the sample is a complex one (multilayered sample) we have to consider separately each layer of the sample and of the surroundings. At each interface, the incident thermal amplitude ( $A_0$ ) is transmitted ( $A_1$ ) and

reflected ( $B_0$ ) according to the thermal properties of the media. The analogue of the refractive index is then the thermal effusivity  $e = (k/D^{1/2})$ . We must underline that a strong thermal mismatch can be useful for the photothermal investigation of a layered sample.

The two experimental setups we have described require a focussed heating spot. Therefore, in order to interpret the results, the heat equation diffusion has to be solved in a cylindrical geometry ( $T(r,z,t)$ ). Fourier and Hankel transforms allow to run easily calculations in the case of modulated heat sources. Let us simply outline that in the case of a heat point source, the result is given by:

$$T(R,t) = T_0 / R \exp(-R/\mu) \exp(j[\omega t - R/\mu])$$

for the heat diffusion equation in a homogeneous material. We have a spherical thermal wave.

## 2- Photothermal experiments on ceramics

### a) determination of the thermal diffusivity

In order to understand the behaviour of complex materials such as functional ceramics or refractories, it is important to know not only the physical parameters at a macroscopic scale but also the parameters of each constituent. Photothermal experiments are able to determine the thermal diffusivity at different scales from the micrometer to the millimeter by varying the modulation frequency / 5 /.

With the two setups we have already described, to measure the thermal diffusivity we have to explore the heated area. The width of the latter is correlated to the diffusivity. After a modelization taking into account all media in which heat can diffuse and the geometrical parameters of the experiment, the thermal diffusivity can be extracted with a precision better than five percent.

In a mirage experiment, the heating spot diameter is 50 to 100  $\mu\text{m}$  and the heated area is explored on a few millimeters depending upon the sample's diffusivity and the modulation frequency. The phase of the mirage signal is the pertinent parameter to record, since it is this less sensitive to the imperfections of the surface. Two values of the deviation can be measured parallel or perpendicular to the surface, because of the shape of the thermal gradient in this cylindrical geometry. Figure 3 is an example of such normal deviation for the binding of a carbon bonded magnesia refractory sample. The thermal diffusivity of this ceramic is measured to be  $0.2 \text{ cm}^2\text{sec}^{-1}$ .

The thermal diffusion length decreases with increasing frequency. So, at high modulation frequencies the heated area is only a few tens of micrometers wide. Figure 4 shows a measurement at 100 kHz on a gold sample for validation of our apparatus and on a magnesia grain in carbon bonded magnesia refractory coated with gold (to prevent light scattering inside the magnesia grain). The thermal diffusivity of this aggregate is measured to be  $0.09 \text{ cm}^2\text{sec}^{-1}$ .

### b) photothermal imaging

The photothermal microscope makes it possible to make thermal images with a micrometer scale resolution. In order to obtain such images the sample is usually scanned by the pump spot and the probe spot superimposed. But in the case of an irregular sample surface, we find better to leave the probe spot at rest on an area of good reflectivity, while the heating spot is scanned on the surface. Figure 5 is an example of such a photothermal image recorded in the binding of the carbon bonded magnesia refractory. By comparing this photothermal image with the optical image obtained by recording the d.c. component of the reflected pump intensity, the anisotropy of the heat propagation is clearly evidenced.

Figure 6 is an example of thermal barrier between two grains of AlN ceramic, revealed by a photoreflectance experiment run at 1 MHz. The modelization of this experiment requires the resolution of the heat equation diffusion with a boundary without cylindrical geometry. When the barrier is vertical it is possible to solve the equation analytically. When the barrier is slanted we have to use a numerical approach. The knowledge of these thermal barriers at a microscopic scale allows one to understand the macroscopic behaviour of the ceramic / 6 /.

## c) heat diffusion dynamics

The photothermal experiments also allow one to follow the propagation of heat inside the sample when a pulse or a step of heat are sent on the sample surface. It is possible to follow either the front surface temperature or the back one, according to the sample thickness. An IR detector is suitable to detect such surface temperature variations. Figure 6 shows the surface temperature variation of a carbon bonded alumina refractory during a step of heating. We can easily see that the behaviour of the sample is strongly dependent upon the time of the recording. In view of the large heterogeneity of the sample this kind of behaviour is not surprising.

In conclusion, this paper evidences the strong abilities of photothermal techniques such as mirage effect and photothermal microscopy for studies of thermal properties of complex materials such as ceramics. The determination of the thermal diffusivity at various scales is of importance for the understanding of the thermal behaviour of refractories.

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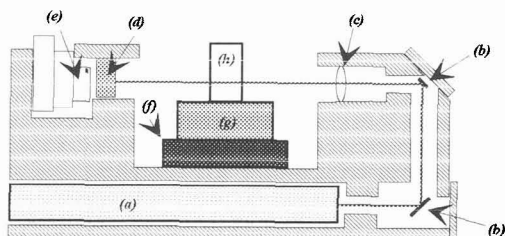


Fig 1 : Mirage cell a) He-Ne Laser; b) Mirrors; c) lens; d) interference filter; e) position sensor; g) micropositioners; h) sample

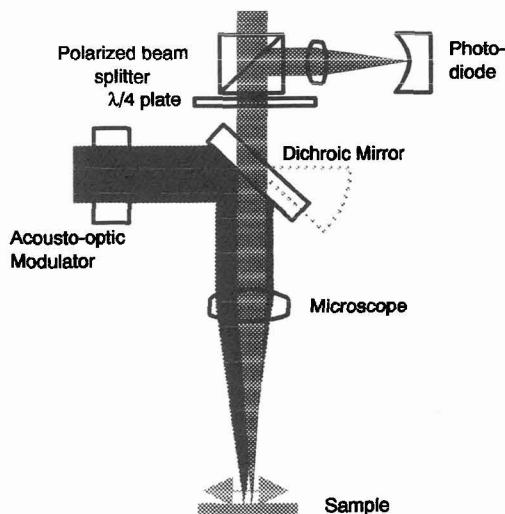


Fig 2 : Photoreflectance setup

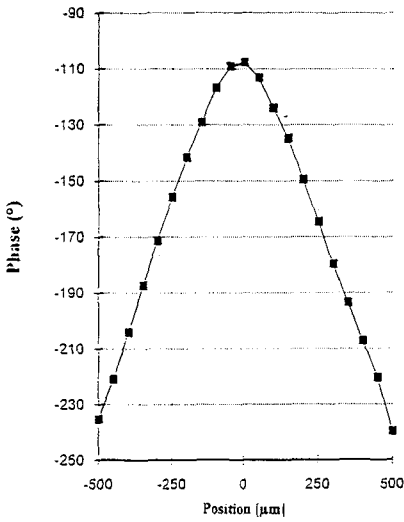


Fig 3 Phase signal versus position for the normal deviation in a mirage effect measurement at 330 Hz (carbon binding of a carbon bonded magnesia ceramic)

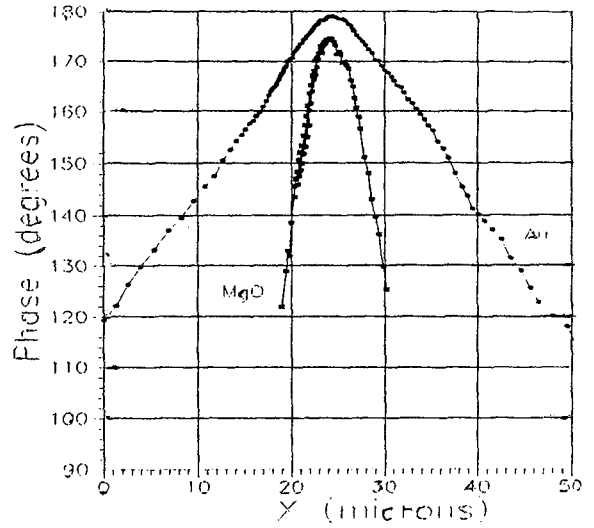


Fig 4 Phase versus position in a photothermal microscope measurement in gold (top) and MgO (bottom); Modulation frequency 100 kHz

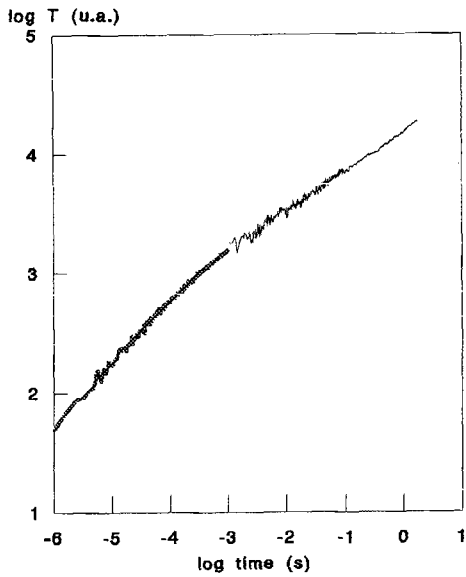


Fig 7 : Surface temperature response versus time at a step heating carbon bonded alumina

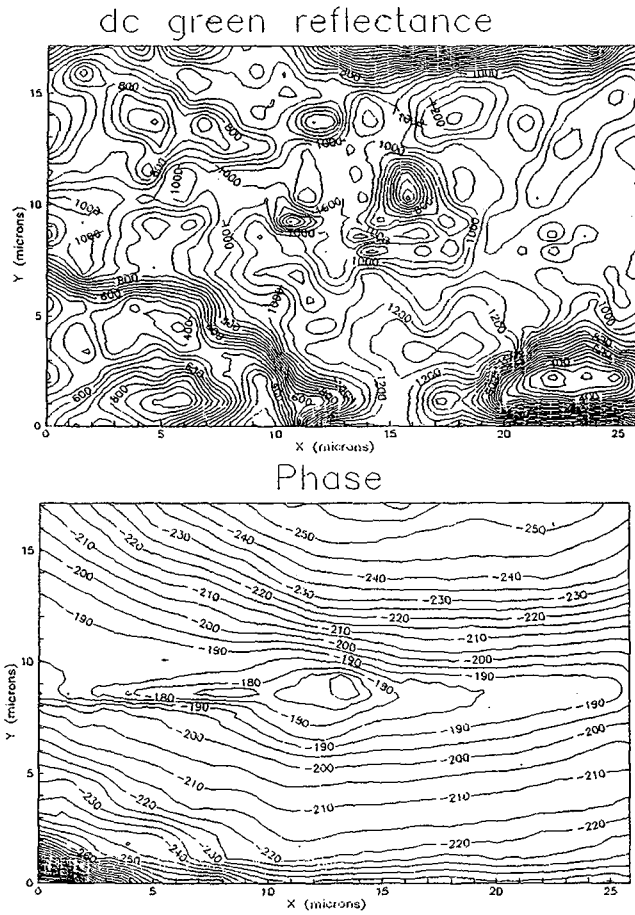


Fig 5 Optical (top) and photothermal (bottom) microscope images of the same area, in the carbon binding of carbon bonded MgO ceramic. The optical image essentially shows micron scale surface irregularities while the photothermal image clearly evidences strongly anisotropic heat diffusion. Frequency 20kHz; detection spot at the center

Fig 6 : Photothermal microscope sweep across an AlN grain, clearly evidencing the thermal barriers at the two boundaries of the grain

