Thermal characterization of high temperature reactor particle layers by photothermal microscopy

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Abstract. Thermal diffusivity of particle pyrolytic carbon layers deposited on 500 µm diameter spherical particle before irradiation has been measured using photoreflectance microscopy (PM). This technique is used to characterize of such small constituents at a microscopic scale and temperature controlled up to 1000 °C. Nevertheless, one of the layers - buffer layer - needs a particular analysis due to its porous structure. Indeed, measurements by PM on this material only permit to obtain the thermal diffusivity of the solid skeleton. These require the use of a numerical homogenization technique to estimate an effective thermal conductivity. The effect of temperature is discussed. Mapping of the thermal diffusivity of coated fuel particles provides useful data for modelling fuel performance during the nuclear reactors operation.

1. Introduction
Renewed interest in nuclear energy around the world has led to a renaissance of nuclear technology development in several countries. Two of the most promising technologies currently under study are the High Temperature Reactor (HTR) - Generation III+ - and the Very High Temperature Reactor (VHTR) - Generation IV -. The safety and efficiency of these reactor designs largely depends on the properties of coated fuel particles, making their detailed characterization and fundamental understanding of their properties vitally important.

Production and characterization R&D studies of innovative nuclear fuels are performed at the Commissariat à l’Energie Atomique (CEA). Fabrication of TRISO (TRistructural ISOtropic) HTR fuel particles are already in progress, contributing to the French VHTR fuel development and qualification program [1,2]. In the same time, new microstructural characterization methods are investigated; different thermo-mechanical properties have to be measured with appropriate devices [3].

Our thermal characterization has been made on HTR coated particle (diameter is about 900 µm) inserted in graphite element. It consists (Figure 1) of a ZrO$_2$ fuel kernel with diameter about 500 µm (simulated UO$_2$ fuel kernel) that is surrounded by several layers. Each layer in the TRISO particle design has a specific function in the fuel performance and fission product (FP) retention. A porous pyrolytic carbon buffer layer (thickness about 90 µm), provides a void volume for gaseous FP and accommodates fuel kernel swelling due to fission. An Inner dense pyrolytic carbon layer, IPyC (thickness about 35 µm), reduces tensile stress on SiC and acts as a diffusion barrier to metallic FP. A silicon carbide layer, SiC (thickness about 30 µm), ensures leak tightness to metallic FP during normal and accidental situations. An Outer dense pyrolytic carbon layer, OPyC (thickness about 35 µm), reduces tensile stress on SiC as IPyC and provides bonding surface for matrix material.
Because of the layers thickness (from 30 µm to 90 µm), conventional characterization methods are inappropriated, and only photoreflectance microscopy (PM) has the required spatial resolution to determine precisely the thermal diffusivity of such small constituents [4, 5].

The object of this paper is to present the method used to determine at temperatures up to 1000 °C the thermal properties of the HTR fuel particle pyrolytic carbon layers before irradiation, as well as results and analysis of properties measurements and estimations. The results of this work are intended to be used as input data in a numerical code able to calculate thermo-mechanical effective properties of HTR fuel.

2. Experimental Setup
Photothermal microscopy is based on the measurement and analysis of the periodic temperature increase induced by the absorption of an intensity-modulated laser beam (pump beam) [6]. By detecting the variations of the thermally induced reflection coefficient with the help of a secondary continuous laser beam (probe beam), the local temperature increase at the sample surface can be measured. Thermal parameters are then identified by fitting the periodic signal to a suited model. Unlike other photothermal methods, this contactless technique has a micrometric spatial resolution.

The experimental setup is similar to that describe in [7] and it consists of:

The pump beam is a continuous Coherent Verdi laser (wavelength 532 nm, maximum power 2 W) that intensity is modulated by a frequency generator-driven acousto-optic modulator operated at frequencies up to 2 MHz. The pump beam is then directed by a dichroic mirror and finally focused onto the sample surface in the heating stage with an optical microscope.

The probe beam is a continuous Coherent Sapphire laser (wavelength 488 nm, power 20 mW) that travels through a quarter-wave plate and the dichroic mirror, and then is focused onto the sample surface with the same optical microscope. After reflection, it travels through the quarter-wave plate again and is then sent to the photodiode by a beam splitter cube. An optical filter prevents any pump beam photon from reaching the detector. Finally, a lock-in amplifier extracts the amplitude and phase of the thermoreflectance periodic signal collected by the photodiode.

A PC controls the dichroic mirror orientation and consequently the distance between the probe and pump beam locations. A heating stage can be used to characterize sample thermal diffusivity in temperature up to 1500 °C under a vacuum level better than $10^{-3}$ mbar. The heating stage window, made of fused silica, transmits 93% of the intensities of the two beams. An objective (x40) with a large work distance specially designed to correct the spherical aberrations due to the heating stage window is used. A mirror polishing of the sample is necessary to ensure a good reflection.
3. Pyrolytic carbon layers characterisation

The layer’s characterized zones are chosen after microscopic optical observation which allows us to know if they can be considered homogeneous at the PM spatial resolution scale.

Using our photothermal microscope and the described thermal parameter extraction scheme of [6], we determine the thermal diffusivity of each pyrolytic carbon layers. Figure 2(a) shows a typical result obtained at 100 kHz after a two-dimensional mapping of the buffer surface. The solid skeleton of the buffer does not present privileged direction of the heat transfer. In spite of a small increase of the phase shift in the left lower part of the cartography - that one can suppose this is due to the reflection of the thermal wave on a pore present in the vicinity of the characterized zone - the isophases have a circular form characteristic of an apparently isotropic behavior. Figure 2(b) shows the results of a least square adjustment between several isotherms and the equation of an ellipse. The thermal anisotropy determined is 1.01 which confirms the thermal isotropic behavior of the buffer skeleton and the thermal diffusivity identified value is equal to $5.3 \times 10^{-6}$ m$^2$/s.

![Figure 2](image.png)

**Figure 2.** (a) Two-dimensional phase shift scan at 100 kHz, (b) Extracted isophase lines fit result.

Figure 3 represents the attenuation (on the left) and the phase shift (on the right) of a measured signal at 100 kHz (dot points) and its related least square adjustment (lines).

![Figure 3](image.png)

**Figure 3.** One-dimensional scanning of the buffer skeleton at 100 kHz and its related least square adjustment.

The statistical processing of a series of measurements makes it possible to obtain an average value of the thermal diffusivity of the buffer skeleton equal to $5.2 \times 10^{-6}$ m$^2$/s, with a satisfying standard deviation equal to $0.5 \times 10^{-6}$ m$^2$/s.

Last, we can note that the diffusion length is around 4 μm at 100 kHz. That means that this frequency is (i) high enough to consider that the periodic increase temperature is confined inside the skeleton and (ii) low enough to average sub-micrometric pores influence when thermal diffusivity is determined.
The same procedure for other pyrolytic carbon layer is used and the results are compared to previous ones on table 1.

| Layers | Thermal diffusivity \(10^{-6} \text{m}^2/\text{s}\) | Standard deviation \(10^{-6} \text{m}^2/\text{s}\) |
|--------|---------------------------------|------------------|
| Buffer | 5.2                             | 0.5              |
| IPyC   | 7.6                             | 3.2              |
| OPyC   | 3.7                             | 0.2              |

4. Result in temperature

Figure 4 illustrates thermal diffusivity evolution versus temperature for the skeleton buffer of another particle. The thermal diffusivity decreases quite rapidly from 200 °C to 600 °C and very slowly until 1000 °C. We can also notice that, at room temperature, the thermal diffusivity of these particles is 20% lower than the previous diffusivity measurements. It could be due to the different heat treatments applied to this sample when it has been elaborated, which weren’t exactly the same.

When the evolution of volume specific heat with temperature of the buffer will be known, the next step will be to determine the buffer effective thermal conductivity with the help of our numerical homogenization technique applied to 3D-X-ray tomography.

5. Numerical homogenization technique

As described in [7], the buffer layer conductivity can be determined from micrographs of the polished buffer 2D surface by a numerical homogenization technique. However, polishing may change the buffer microstructure, which can have a significant impact on its thermal conductivity. To overcome this problem, 3D-X-ray tomography of a buffer shaving removed from a particle was taken at the European Synchrotron Radiation Facility (ESRF) with a spatial resolution of 0.3 µm. Thermal conduction numerical experiments, described in [7] were applied to a volume extracted from the tomography to determine the effective conductivity of the buffer layer. Each element (voxel) is considered as homogeneous and isotropic, and is attributed local thermal properties corresponding to the buffer skeleton (measured diffusivity by PM, \(\rho C\) known [3]) or to the gas (air) confined to the pores.

Figure 5 represents a temperature field calculated at 20 °C in the circumferential direction from which the effective thermal conductivity in this direction is deduced: \(5.3 \times 10^{-1} \pm 5.21 \times 10^{-2} \text{W/m/K}\). In the same way, the effective thermal conductivity in the radial direction has been estimated to \(3.5 \times 10^{-1} \pm 4 \times 10^{-2} \text{W/m/K}\). The results underline the thermal anisotropy of the buffer layer due to the fabrication process (anisotropy degree \(\approx 1.5 \pm 0.3\)). Indeed, the coating is performed by
chemical vapour deposition in fluidized bed. During the process, the particles collide and rotate which involves a stratified structure. This numerical technique allows us to obtain the thermal conductivity of the whole buffer layer that we can’t measure.

6. Conclusions and perspectives
The measurements of thermal diffusivity on the different pyrolytic carbon layers show that IPyC is more diffusive than the solid skeleton of the buffer, itself more diffusive than OPyC. A particular characterization approach has been developed to determine thermal buffer properties at temperatures up to 1000 °C. This approach, coupling local measurement by PM and numerical homogenization technique, has been validated for thicker buffer layer especially elaborated [7].

The results obtained for HTR particle layers are to be correlated to the microstructural studies carried out by [8]. With regard to the buffer, the low density of its skeleton contributes to reduce its thermal diffusivity whereas the many heat treatments that it undergoes, during elaboration process, tend to increase it. The competition between these two phenomena leads the buffer skeleton to have an intermediate thermal diffusivity between IPyC and OPyC ones.

In addition, an evaluation of porosity by images analysis carried out on another study, showed different pores sizes distribution and a more important porosity in IPyC layer than in OPyC layer. These measurements remain to be confirmed but can explain the important dispersion of the thermal diffusivity estimated on IPyC where the probability that measurement is disturbed by pores is stronger.

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