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Optical system for real-time monitoring of nuclear fuel pellets at high temperature

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Abstract

Understanding the behavior of nuclear materials regarding fission gas release in relation to the different thermal loads to which they can be subjected requires appropriated annealing tests in order to measure both the absolute level and the time dependence of the released fission products together with the corresponding fuel micro-structural changes during representative thermal transients. In this context, we describe in this paper the development and qualification of an experimental platform coupling heat treatment, gas release analysis and optical systems, for the study of fuel pellets at high temperature. This system, which is mainly devoted to power transient and LOCA (Loss Of Coolant Accident) type simulation, is based on an induction furnace to heat the pellet at high temperatures (up to 2000°C with up to 50°C/s temperature ramps) in controlled atmosphere. The device is coupled with dedicated analysis loops that are designed to identify and quantify on line the gaseous fission products released during the annealing test. The purpose of the optical system is the real time monitoring of the sample surface to provide additional information (for instance on the micro structure evolution and fuel fragmentation). It is coupled with non-contact temperature measurements by thermal radiation analysis to monitor and control the fuel temperature. Based on experiments conducted on non-irradiated Uranium dioxide samples, we demonstrate the performance of the system in the range 20-1800°C with 10 μm resolution on a field of 1cm², with simultaneous temperature measurements obtained by multispectral pyrometry. We also discuss the limitations of the system as well as further developments for integration in a hot cell for application to irradiated fuels.

Keywords — Nuclear fuel, Instrumentation, Non destructive examination, High temperature

I. INTRODUCTION

Several programs exist at the international level which aim to improve our understanding of the behavior of nuclear materials in relation to the different thermal loads to which they can be subjected, i.e. from nominal operation through extreme conditions. Usual studies in this field involve dedicated annealing tests in order to monitor both the absolute level and the time dependence of the released gas [1, 2, 3, 4], together with the corresponding analysis of fuel microstructural changes during representative thermal transients. Indeed, experimental knowledge of fission gas release alone is not efficient enough where the structural or mechanical integrity of the fuel pellet is at stake or interest [5]. This approach requires not only to extend the database for transient analyses to a broader range of test conditions and materials, but also to quantify properly the uncertainties relative to the database to make its use more relevant. This corresponds to the driving force of the present work. Up to now, predicting accurately the Fission Gas Release (FGR) from high burn up UO_2 and/or MOX (Mixed Oxide) fuels at off-normal conditions, such as power transient, reactivity initiated accident (RIA) and loss-of-coolant accident (LOCA), is still a significant and important challenge.

In this context an annealing device, referred as *MERARG* (french acronym for Means of Study for Annealing and Gaseous Release Analysis) [6, 7, 8], has been developed. It allows annealing at high temperature, - usually between 1200 °C and 1600 °C but can reach peak of 2800 °C - on irradiated nuclear fuel pellets with slow (between 0.05 and 0.5 °C/s) and moderate (maximum 20-30 °C/s) temperature ramps thanks to an induction furnace. This device is coupled with dedicated analysis loops that are designed to identify and quantify the gaseous fission products released during the annealing test. The development of additional optical diagnostics for implementation on this system is the intent of the present work. It should be used to provide additional information for instance on the micro structure evolution and fuel fragmentation kinetics, together with fuel temperature monitoring by infrared pyrometry.

We describe in this paper the selection of the instrumental options on the basis of the field of view and resolution to be imaged, of the operating distance, of the optical windows, of the available optical access, etc... . Then we describe the development and assembly of the imaging system for the detection and monitoring of the microstructural evolution of the sample. We also detail the integration of a visible spectrometer to perform a multispectral pyrometry measurement at the

surface of the observed sample, in order to acquire simultaneously temperature and image at the same location.

The section 2 is devoted to a quick presentation of *MERARG* facility followed by a description of the optical system and associated instrumentation and protocols. Third section describes the results obtained during the qualification phase, based on experiments performed on Uranium dioxide samples. We conclude with the future improvements and next objectives for hot cell integration of the system.

II. MATERIAL AND METHODS

II.A. *MERARG* Presentation

The objectives of *MERARG* (without the optical device) loop that has been introduced previously are :

- To heat irradiated fuels at temperature levels appropriate to extract all or part of the gaseous inventory it contains,
- To collect the released gases in a circuit swept by a neutral or weak oxidising atmosphere,
- To analyse online the released gases by gamma spectrometry (gamma emitters such as Kr, Xe, . . .) and micro-gas chromatography (He),
- To collect all the gases in the storage capacities in order to obtain samples for offline quantifications.

This device has been developed and patented by the CEA Cadarache [9] mainly to simulate the thermal level seen by the fuel pellets in Pressurized Water Reactors (*PWR*) in the case of *LOCA* type accident. It is implemented in a hotcell at the *LECA/STAR* hot laboratory.

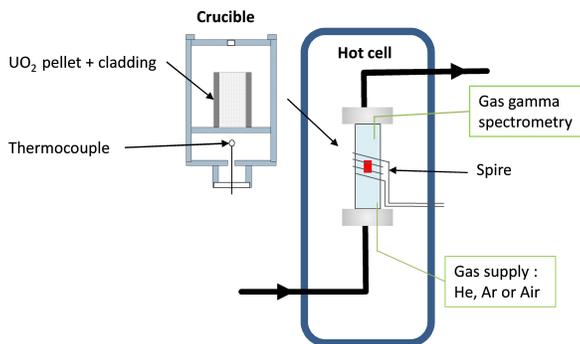


Fig. 1. Simplified schematic of *MERARG*

The fuel pellet to be characterized (usually with its cladding) is placed in a metal crucible that can be tungsten, or platinum according to the experimental needs(Fig. 1): Tungsten is used for high temperatures and platinum for oxidizing atmosphere. It consists of two chambers and is positioned at the center of the induction coil oven powered by a high frequency power generator.

The crucible is a cylinder with a diameter of 20 mm and 30 mm height. The enclosure (2.5 mm-thick Quartz tube) can be filled with dry air, Ar or kept under vacuum. *MERARG* has a thermocouple for monitoring the inductive furnace. The temperature of the sample is regulated by a thermocouple called "local control thermocouple" in the lower chamber of the crucible. As said previously, the *MERARG* facility has many other uses than the two instrumentations which will be presented hereafter, for instance gamma spectrometry and mass spectrometry sighting to monitor on-line the fission gas release kinetics during a dedicated annealing sequence.

In order to test the new optical system in realistic conditions and assess the performance and limitations of the system, we have conducted experiments on the *MERARG* mock-up experiment that is intended for all "cold" qualification phase before integration in the hot cell. The temperature of the sample is regulated through a PID controller (Proportional-Integral-Derivative controller) for adjustment of the power of the HF generator. The regulation system uses as an input the signal from the thermocouple (type C) at the bottom of the crucible shown on Fig. 1 (pilot TC), giving T_{CC} (Crucible lower Chamber Temperature). However in order to connect the real sample temperature to the crucible temperature a thermal qualification phase is conducted as a standard procedure on this system. It consists of using a second thermocouple inserted in the core of a pellet, giving T_{CP} (Centre Pellet Temperature) to determine the correction factor to apply to the pilot TC as a function of the heating conditions ($T_{CP} = SF * T_{CC} \pm 10\%$). A schematic is given in Fig. 2 for better understanding of the configuration and more detailed information can be found at this reference [10]. At the end of this calibration phase the pellet temperature is correlated to the crucible lower chamber temperature by a correction factor (Scaling Factor - SF). This corrected temperature is given and used in the following of this document.

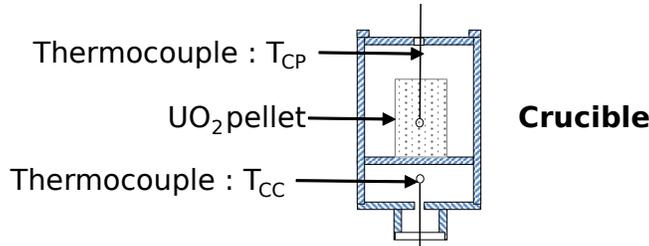


Fig. 2. Schematic of configuration for the thermal qualification phase

The experiments were conducted on UO_2 pellets, made by the CEA. A pellet is 10 mm high for a radius of 4.8 mm and was sintered from natural UO_2 (not enriched or irradiated) powder. The

temperature was increased with slow linear ramps of 1 to 5 °C/s and stabilized at different plateaux for few minutes in order to perform experiments at constant and homogeneous temperature in the sample. We report (Fig. 3) an example of a heating sequence during one of the experiments, where the temperature of interest ranges from 850 °C to 1900 °C (T_{CP}).

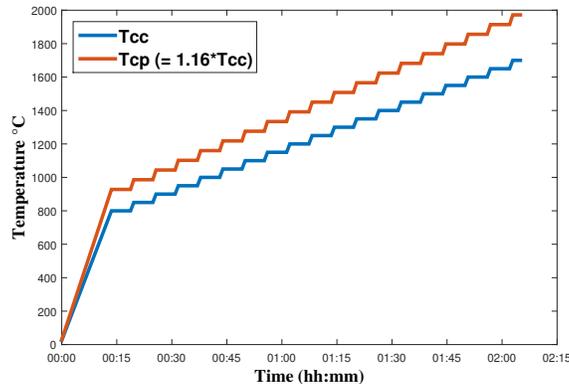


Fig. 3. Example of a heating sequence during one of the experiment, $S_F=1.16$

II.B. Imaging optical system

The optical system has been developed to allow the visualization and the measurement of the fuel pellet in real-time during thermal annealing conducted on MERARG. It is intended to operate in a so-called "hot lab" environment (i-e. working on irradiated materials), although at this point we report only on the implementation in a mock-up experiment of MERARG, operating on non-irradiated fuels but totally identical to the MERARG system.

The sample is placed in a Tungsten crucible with a 6 mm aperture on the upper part to allow sample observation (Fig. 4). This crucible is itself placed in a quartz tube (Fig. 4) sealed on the upper part by an optical window located at 250 mm from the crucible, with a clear aperture of 25 mm. Considering this configuration the working distance of the optical system must be greater than 250mm with a maximum field of view of 6mm on the target. On this basis, the numerical aperture of the optical system is limited to 0.1. Accordingly the image resolution in such conditions can be estimated to 3 μm with 0.5 μm wavelength in case of diffraction limited optical system ($0.61 \lambda/NA$). In such conditions the depth of field can be estimated to 100 μm suggesting the need for fine adjustment of the whole optical assembly with respect to the sample plane. The field

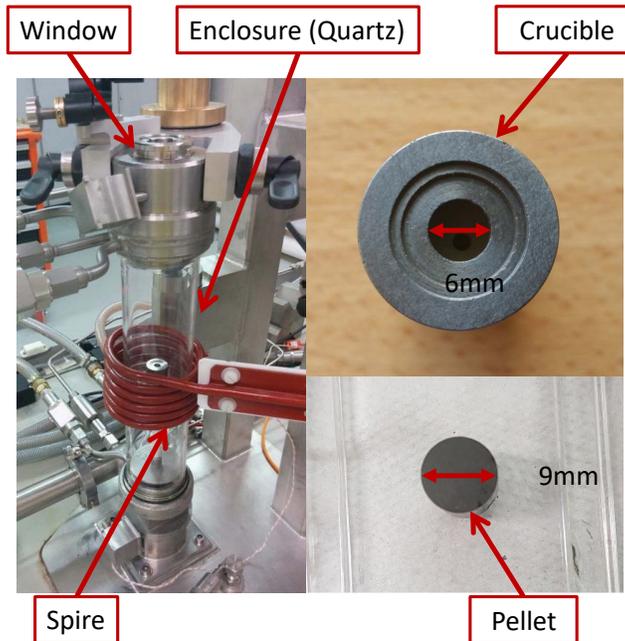


Fig. 4. Photo of MERARG (left), of the crucible (top right) and of a pellet of UO₂ (bottom right)

of view being 6 mm in the object plane, a magnification in the range of 1 to 2 is required to image the whole area of interest on classical industrial camera sensors (1 inch). On this basis we have chosen a K2 DistaMax long distance microscope coupled with a CF1 objective (Infinity, USA). The implementation of this microscope, along with an adapted illumination system that we have developed, on the MERARG platform is described on Fig. 5.

A high power collimated LED source is used for illumination, operating at 455nm with 900mW (Thorlabs M455L3 with SM2F32-A collimator), combined with a pellicle beam splitter for the visible (Thorlabs BP245B1). The shortest wavelength possible has been chosen for illumination in order to be able to image the sample in condition of thermal radiation from the sample and crucible. A corresponding band pass filter (Thorlabs FB450-40) is placed in the camera port. A cross-polarizer configuration has been used to reduce parasitic reflections from the metallic (crucible, mechanical parts) and glass parts (windows) of the system while not affecting the scattered light from the sample. The effective received optical power on the sample has been measured to 50 mW, corresponding to a power density of 1.7 W/cm^2 and resulting in negligible heating of the sample. Additionally, as the system should also allow thermal radiation measurements, a

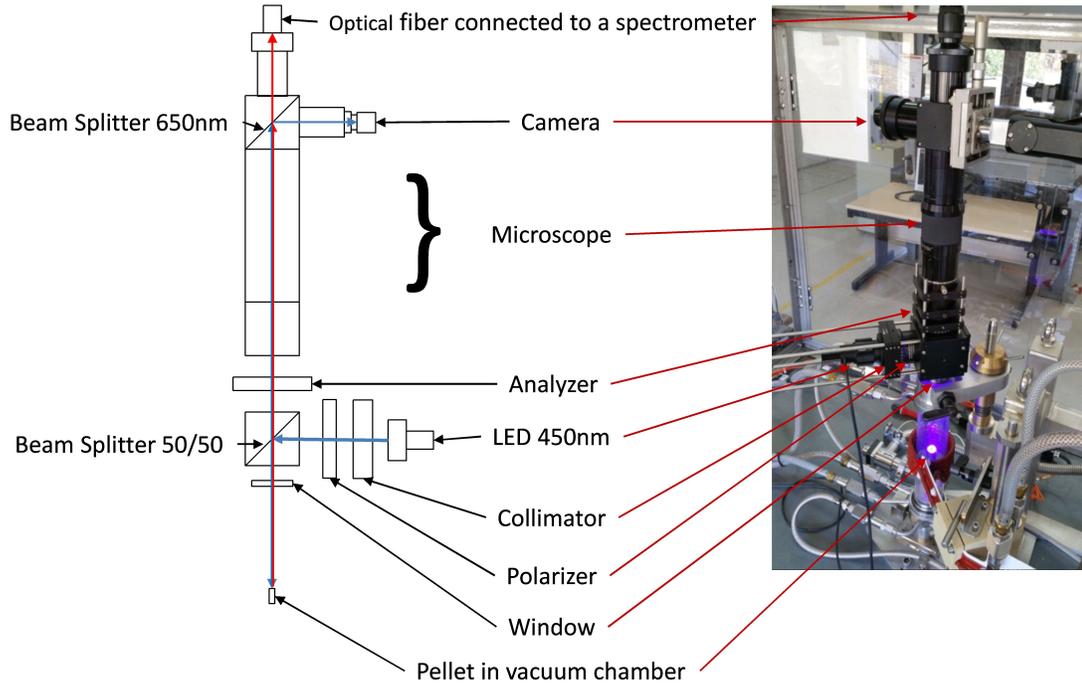


Fig. 5. Schematic and photo of the optical device operating on the MERARG mock up experiment

two ports configuration is used: one port dedicated for the video camera and the other one for spectral measurements (described below). A longpass filter is used as the beam splitter (Thorlabs DMLP650R, cut-off wavelength at 650nm).

For imaging we have used a standard industrial camera (Grasshopper 3-U3-41C6-C, FLIR) with a definition of 2048x2048 pixels, a pixel size of $5.5 \mu m$ and 1-inch sensor size. The frame rate is limited to 90 fps but a high-speed camera could easily be used on the system to observe and monitor the dynamics of transient events such as fracturation. We report (Fig. 6) on the observation of a UO_2 sample on the *MERARG* mock up experiment with the optical system, at ambient temperature. In this image, the black part corresponds to the top of the crucible. Fine details such as a fracture on the sample can be resolved (Fig. 12 & 13). A color camera has been chosen in order to be able to operate simultaneously in different spectral bands defined by the color filters (RGB) and take benefit for imaging in a large range of temperature, as it will be shown in the next section.

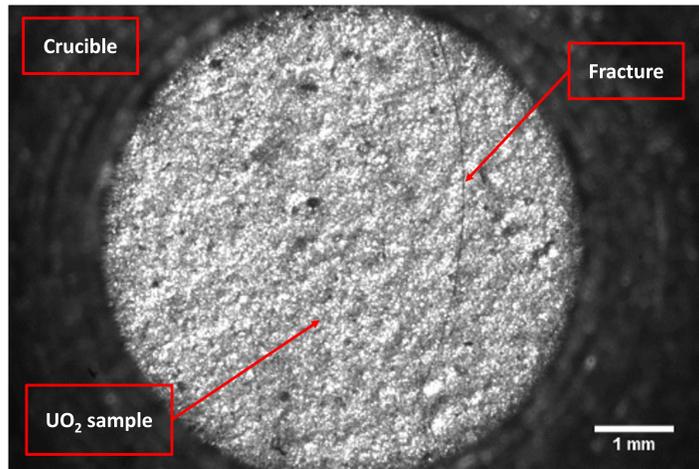


Fig. 6. Observation of a UO₂ pellet with the optical system

II.C. Multispectral pyrometric measurement

During the annealing tests, if the temperature of the metallic crucible can be monitored with a thermocouple, the only way to measure the sample temperature is with optical techniques through the crucible aperture. Pyrometric techniques for non-contact temperature measurements are mainly based on monochromatic methods that require the knowledge of the sample emissivity to retrieve the temperature from radiance measurements. Reliable data are available for the total emissivity of UO₂, as reported for instance in the review by Fink on the thermophysical properties of UO₂[11]. The reported emissivity varies little with temperature and is only a weak function of wavelength. A value of 0.85 ± 0.05 is recommended for UO₂. This is in agreement with the value of 0.83 which was determined by Manara et al. for the normal spectral emissivity of UO₂ [12]. However, in the perspective of working on irradiated fuels at high temperature, the spectral emissivity needs to be measured and cross-checked with complementary techniques during the tests. Bichromatic or two-color methods are of interest since they do not require the knowledge of emissivity but of the emissivity ratio between the two measurements wavelengths. Nevertheless this ratio is still to be determined with great accuracy. To go further, multispectral pyrometric techniques require only the wavelength dependency function of the emissivity to retrieve the surface temperature [13] and are suitable for operation in extreme environments encountered in the nuclear field [14]. Such techniques were employed successfully by Manara et al to measure thermograms

and spectral emissivity of oxides such as UO_{2+x} [15]. Therefore, we have coupled such a diagnostic to the optical system we developed from *MERARG*.

In our case the temperature of interest lies in a range from ambient to 2000 °C. We recall below (eq. 1) the Planck's law that describes the relation between spectral radiance, wavelength and temperature:

$$R(\lambda, T) = \epsilon(\lambda, T) \frac{2 \cdot h \cdot c^2}{\lambda^5} \times \frac{1}{e^{\frac{h \cdot c}{\lambda k_B T}} - 1}. \quad (1)$$

ϵ the emissivity

R , the spectral radiance

λ , the wavelength

T , temperature

k_B , is the Boltzmann constant

h the Planck constant

c , the speed of light in the vacuum

Since the multispectral pyrometric measurement is intended to be integrated in the optical system described before, it needs to operate in the visible/NIR range for compatibility with the spectral transmission of the optical elements. Therefore a visible/NIR spectrometer based on silicon detector array (sensitivity in the range 200-1100nm) is adapted to this measurement. We have plotted in Fig. 7 the spectral radiance in the case of black body ($\epsilon=1$) in this range of wavelength.

Obviously, some compromises had to be found on the targeted temperature range if we want to operate in the visible/NIR range. We have therefore restricted the range of measured temperature to 850-2250 °C. We have worked with a standard fiber optic spectrometer (Avaspec-ULS2048, Avantes). The entrance of the optical fiber is placed in the second port of the optical imaging system, in the center of the image plane so that the only radiance at the central part of the image is collected by the fiber. A silica optical fiber with 1mm core is used for collection (Thorlabs M35L02), which corresponds to 1.5 mm diameter in the sample plane. It is assumed that the sample has a homogeneous temperature distribution on its surface. The effect of thermal radiation by the crucible on the measurement is neglected for several reasons. Firstly, due to the imaging configuration, the optical fiber receives only the direct emission from the pellet and

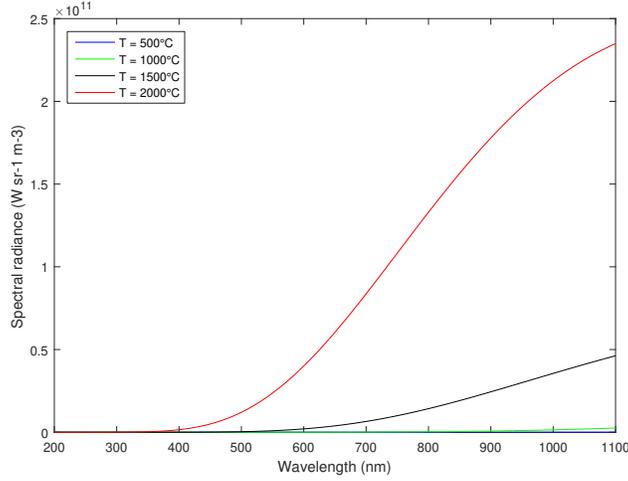


Fig. 7. Theoretical radiance of a black body

no direct emission from the crucible. Only scattered light from the crucible’s emission on the UO_2 pellet could be collected, which is negligible in first approximation considering the estimated absorption of UO_2 material (0.85). Secondly the emissivity of tungsten in the visible to NIR domain is considerably lower than that of UO_2 [16] which is favorable to this approximation.

To determine the spectral response of the device (imaging system, fiber and spectrometer) we have used a Tugnsten-Halogen calibration light source (DH-2000-CAL, Ocean Optics). The procedure consists in imaging the output of the source with the optical system defined previously under similar conditions to the MERARG conditions, ie through the viewport of the quartz vacuum chamber, and collect the light on the spectrometer port. From the theoretical spectrum and the received signal, the transfer function can be calculated and taken into account for measurements. An example of the acquired raw spectrum measured on a UO_2 pellet heated at 1200 °C is shown on Fig. 8 (curve in blue). Also shown on the Figure is the treated signal taking into account the transfer function (curve in red).

In order to determine the temperature of the sample surface from the acquired and treated spectrum, we use an algorithm based on the least-squares method through the minimization of the following error function:

$$\chi^2 = \sum |S(\lambda_i, T) - R(\lambda_i, T)|^2 \quad (2)$$

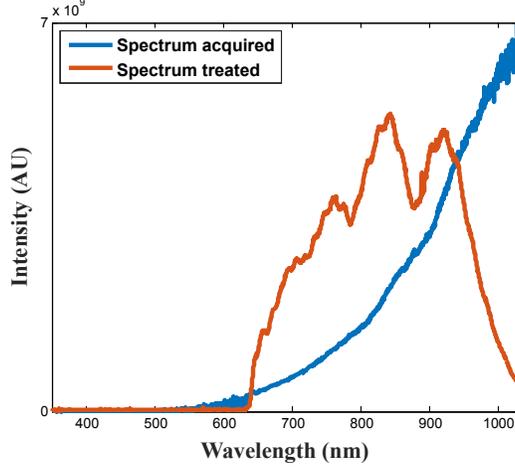


Fig. 8. Spectrum as received from the spectrometer of a UO_2 pellet heated at 1200° (blue) and after processing with the inverse transfer function (red)

with S the measured Spectrum, λ_i the wavelengths channels on the spectrometer, and R the spectral Radiance defined by Plank's law (Eq. 1). An essential condition for the successful application of such methods in multi-wavelength pyrometry systems is the choice of an adequate emissivity function [13]. As a first step in our developments we have used the hypothesis that the emissivity of UO_2 is independent of the wavelength over the analysis range (650-1000nm) [11]. However such choice could be revised when working on irradiated fuels, which is out of the scope of this paper. Equation 2 is solved by the Levenberg-Marquardt method with a Matlab script [17]. An example of application is given in Fig. 9 which corresponds to the case of the UO_2 pellet heated at 1200°C shown previously on Fig. 8.

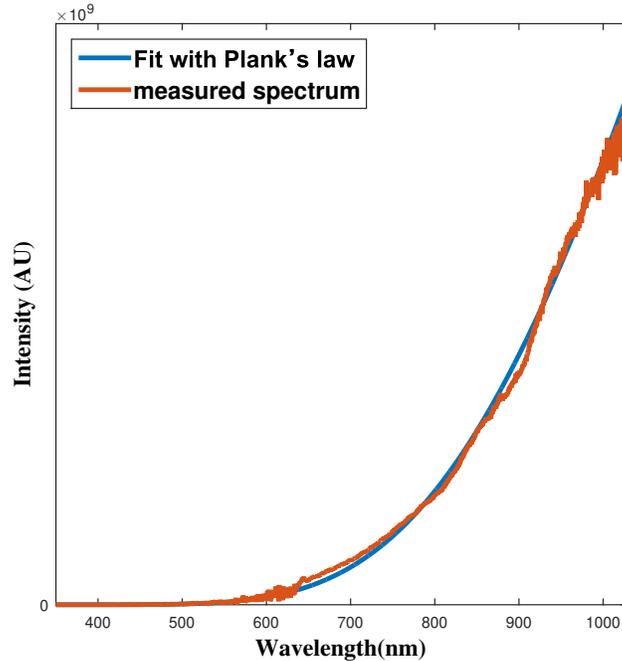


Fig. 9. Measured spectrum of a heated UO_2 pellet and fitting with Plank's law at a temperature of $1200\text{ }^\circ\text{C}$

III. RESULTS AND DISCUSSION

III.A. Imaging of heated UO_2 pellets

The main issue in the development and the implementation of the imaging system was to maintain the image quality and operation of the system in a large range of temperatures, from ambient to more than $2000\text{ }^\circ\text{C}$. Indeed at low temperature there is no thermal emission in the visible range and a blue LED is used as the illumination to acquire images in reflection mode as detailed previously and illustrated in Fig.6. However at high temperature the thermal radiation overcomes any illumination system that could be implemented, and the sample needs to be imaged in emission mode. In order to be able to image the sample in a large dynamic range, we have, as mentioned in part II.B, used a color camera, which by construction acquires 3 images in different wavelength bands referred as Red, Green, Blue. At low temperature with the 450nm LED illumination only the Blue channel is of interest. However at high temperature the LED can be switched off and image acquired in the Red, Green or Blue band depending on the radiance

level. In our experimental condition the transmission from one mode to the other (Reflection to Emission) has been found to be optimal around 1500 °C (T_{CP}). This is illustrated in Fig. 10 with the simultaneous acquisition in Green and Blue channels. For temperature greater than 1500 °C (T_{CP}) the pellet radiates so much that it makes the LED illumination useless and it can be switched off.

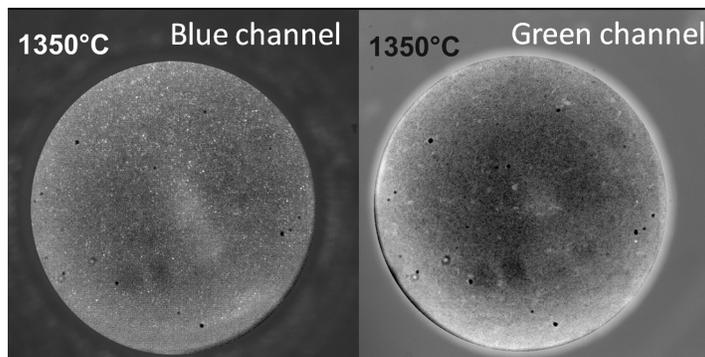


Fig. 10. Observation of a UO_2 sample at 1350 °C (T_{CP}) with 450nm LED illumination. Left: Blue channel of the camera showing the sample illuminated by the high power LED (Reflection mode). Right: Green channel of the camera showing the thermal radiation from sample (Emission mode).

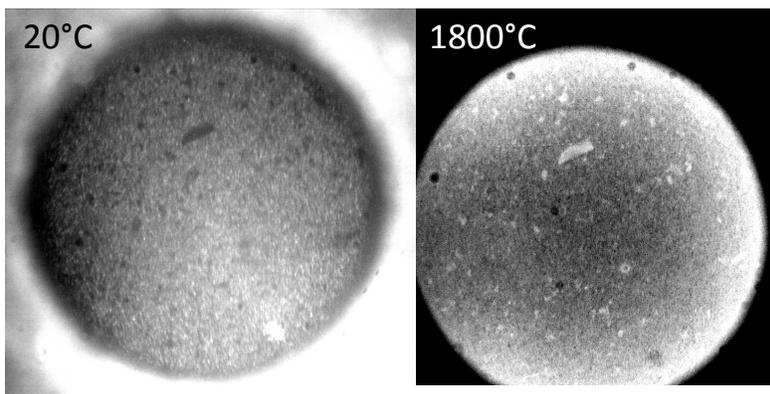


Fig. 11. Observation of a UO_2 sample at ambient temperature and at 1800 °C (T_{CP}).

Experiments were conducted up to temperatures of 1800 °C (T_{CP}) and by adjusting the camera exposure down to 0.016 ms. It was found no limitations on the imaging system. Imaging at higher temperatures can be done by reducing further the integration time. An example of observation of a UO_2 sample at 1800 °C (T_{CP}) is reported on Fig. 11. The ability of the system

to capture transient events is also shown in Fig. 12 with the observation of cracking of the pellet during a dedicated sequence, or the complete fracturing of a pellet (Fig. 13).

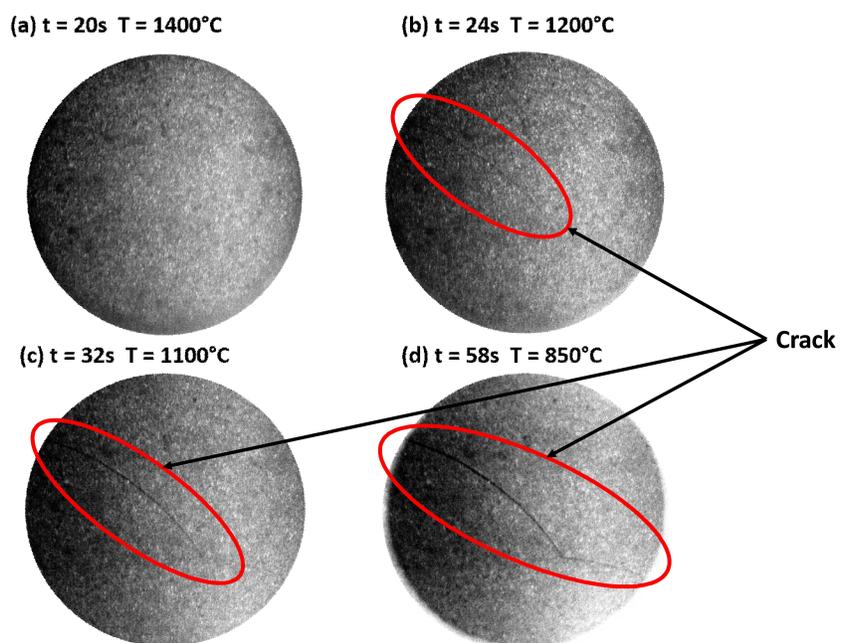


Fig. 12. Images sequence illustrating the cracking of the pellet during a cooling phase.

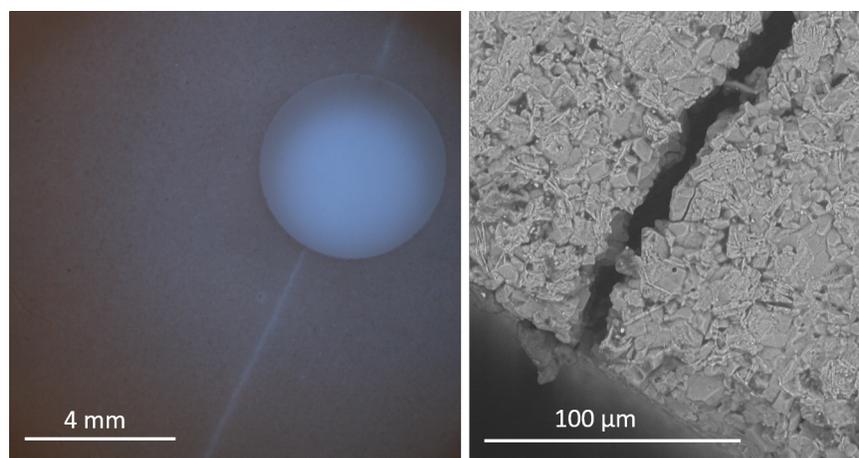


Fig. 13. Fracturing of a pellet observed during a heating phase (left). Same fracture observed with Scanning Electron Microscopy (right) after the experiments showing that the width is approximately 10 microns wide.

III.B. Temperature measurement with multispectral pyrometry

The spectrometric measurement associated to fitting procedure makes it possible to carry out a temperature measurement on the upper surface of the pellet, which may differ from that given by the thermocouple. Spectrometer measurements were taken at the end of 30 seconds plateaux to ensure that the temperature reached a steady-state regime. Measurements were taken from 850 °C, temperature at which the pellet begins to radiate sufficiently to obtain a usable spectrum and then every 200 °C, up to 1800 °C. At 1800 °C the spectrometer saturated even at the lowest integration time (3 ms). To make measurements above 1800 °C, a neutral density filter will need to be inserted in the system. We report, on Fig. 14, on the temperature determined with the multispectral system which is compared to estimated sample temperature, as determined through the thermal qualification phase described previously. It can be seen that the data are in good agreement, and suggest an increase as a function of temperature of the difference between the core of the pellet and its surface due probably to a low axial thermal gradient inside the crucible. This range could be refined after further work on the calibration, which would require access to a black body at very high temperature.

IV. CONCLUSION

The optical system proved its ability to observe a non-irradiated UO_2 pellet in the *MERARG* enclosure, from ambient temperature up to 1800 °C, while simultaneously measuring the temperature of the pellet on the observed surface. Ongoing work focus on the integration of this optical system in a hot cell. This will provide additional information for instance on the microstructure evolution kinetics and fuel fragmentation, as infrared thermography to monitor and control the fuel temperature. Some limitations of the system above 1900 °C were discussed in this paper, therefore other work must be carried out to reach the 2200 °C / 2400 °C which would be the final objective. For this purpose, the integration of a variable attenuator in the optical system to cover a larger range of temperatures may be necessary. Another evolution of this setup will consist in adding a spectrometer operating in the infrared, in this way one could have the top of the peak of the spectrum curve, this would have a significant advantage over the quality and accuracy of the results. The final goal of the device development and qualification described above is to be able to

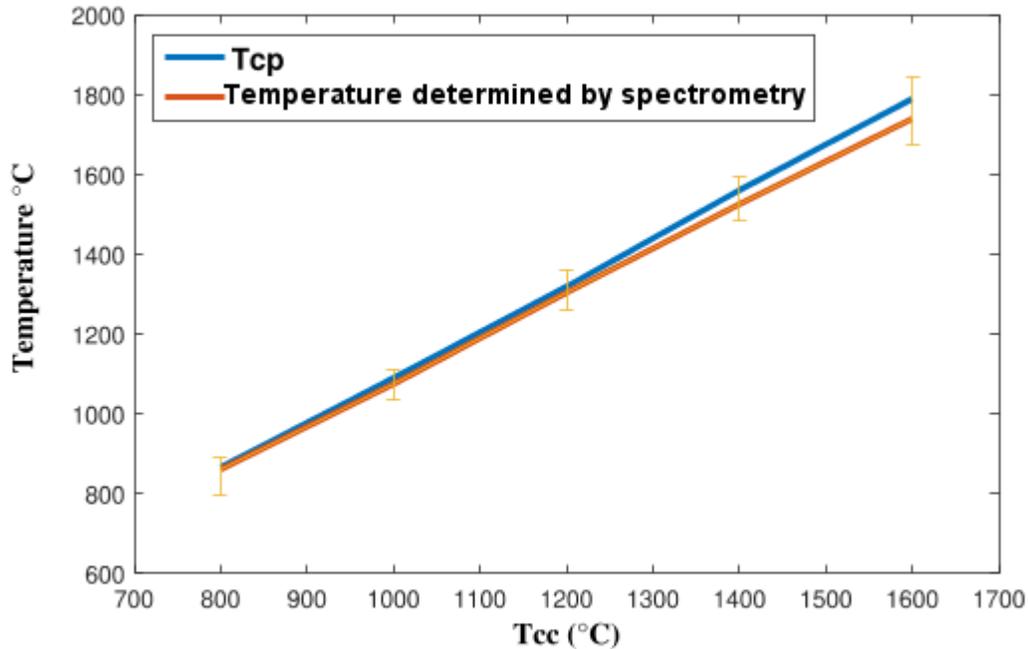


Fig. 14. Temperature of the sample from the thermal qualification phase and as determined by multispectral pyrometry

implement all this new apparatus onto the MERARG Loop (at the LECA-STAR Facility) in order to be used with irradiated fuels. That is the reason why all the technical choices have been done in order to be compatible with this final goal (i.e. used in hot cells). Thanks to the qualification tests performed on UO_2 and surrogate materials, it is possible to transfer directly the apparatus as it is. This next step is currently scheduled in the beginning of 2020. This will allow, for instance, determining fuel fragmentation process in the range of a LOCA type temperature history (i.e. up to 1200 °C) according to different thermal treatment parameters (temperature ramp, final temperature plateau, ...) or to various fuel samples used (burnup and nature (UO_2 or MOX)). Besides, these data could be crossed with the gas γ spectrometer ones which will give crucial information regarding mechanism involved in fission gas release.”

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