DEVELOPMENT OF EXPERIMENTAL PLATFORM FOR ANALYSIS AND IMAGING OF FUEL PELLETS HEATED 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, gases 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 gases and 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 kinetics 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 Uranium dioxide samples, we demonstrate the performance of the system in the range 20-2000°C with 10 µm resolution on a field of 1cm, 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.

1. Introduction

Several experimental 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. By improving this knowledge, it is possible to improve nuclear reactor safety and to precisely define specific on-site emergency plans in the event of an extreme event. The necessity for such an approach was once again highlighted by the Fukushima accident where the need to better assess the source term became painfully evident. One of the most useful ways to do that is to perform appropriated annealing tests in order to measure both the absolute level and the time dependence of the released fission products [1,2,3,4] together with the corresponding fuel micro-structural changes during representative thermal transients, since experimental knowledge of fission gas release alone is not efficient enough [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. Up to now, predicting correctly the fission gas (*FG*) and fission product (*FP*) releases (i.e. the source term) from nuclear fuels at off-normal conditions, such as power transient, reactivity-initiated accident (*RIA*) and loss-of-coolant accident (*LOCA*), remains a significant and important challenge.
In this context an annealing device, referred as *MERARG* (Moyen d'Etude pour Recuit et Analyse des Relâchements Gazeux - Study Method for Annealing and Gaseous Release Analysis) [6,7,8], has been developed. It allows homogeneous annealing at high temperature, - usually between 1200°C and 1600°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 gases and fission products released during the annealing test. It is devoted to power transient and *LOCA* type simulation. 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 kinetics and fuel fragmentation, as infrared pyrometry to monitor and control the fuel temperature.
We describe in this paper the selection of the instrumental options based on the field and resolution to be imaged, of the operating distance, of the optical windows, of the line of sight, etc…the development and assembly of the imaging system coupled to an image processing software for the detection and monitoring of the restructuring 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, 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.

2. Material and Methods

2.1 MERARG

The main objective of MERARG is to carry irradiated fuel at temperature levels which allow the extraction of all or part of the gas inventory contained therein. This device has been developed and patented by the CEA Cadarache [9] to simulate the thermal level seen by the fuel pellets in Pressurized Water Reactors (PWR) in the case of LOCA type accident, at the hot cell laboratory level.

The fuel pellet to be characterized (usually with its cladding) is placed in a metal crucible that can be tungsten, molybdenum or platinum according to the experimental needs. It consists of two chambers and is positioned at the center of the oven induction coil powered by a high frequency power generator. The crucible is a cylinder with a diameter of 20mm and 30mm as high. 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 TC in the lower chamber of the crucible. The furnace, MERARG has many other uses than the two manipulations which will be presented hereafter, for instance gamma spectrometry sighting to monitor on-line the fission gases release kinetics during a dedicated annealing sequence.

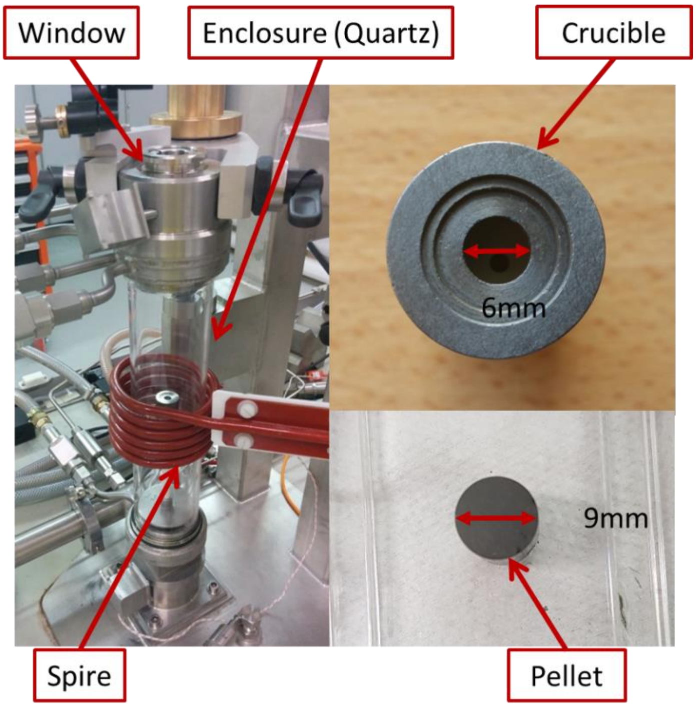

Fig 1. Photo of Merarg (left), of the crucible (top right) and of a UO_2 pellet (bottom right)

2.2 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 hot laboratory environment, although at this point we report only on the implementation in a mock-up experiment of MERARG, operating on non-irradiated fuels but totally equivalent. It has been conceived based on the geometrical constraints of the MERARG system that can be summarized as follows. The sample is placed in a Tungsten crucible with a 6mm aperture on the upper part to allow sample observation (Fig.1). This crucible is itself placed in a quartz tube (Fig.2) sealed on the upper part by an optical window located at 250mm from the crucible, with a clear aperture of 25mm. 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 3µm with 0.5µm wavelength in case of diffraction limited optical system (0.61λ/NA). In such conditions the depth of field can be estimated to 100µm suggesting the need for fine adjustment of the whole optical assembly with respect to the sample plane. The field of view being 6mm in the object plane, a magnification in the range 1 to 2 is required to image the whole area of interest on classical industrial camera sensor (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 displayed in Fig. 2

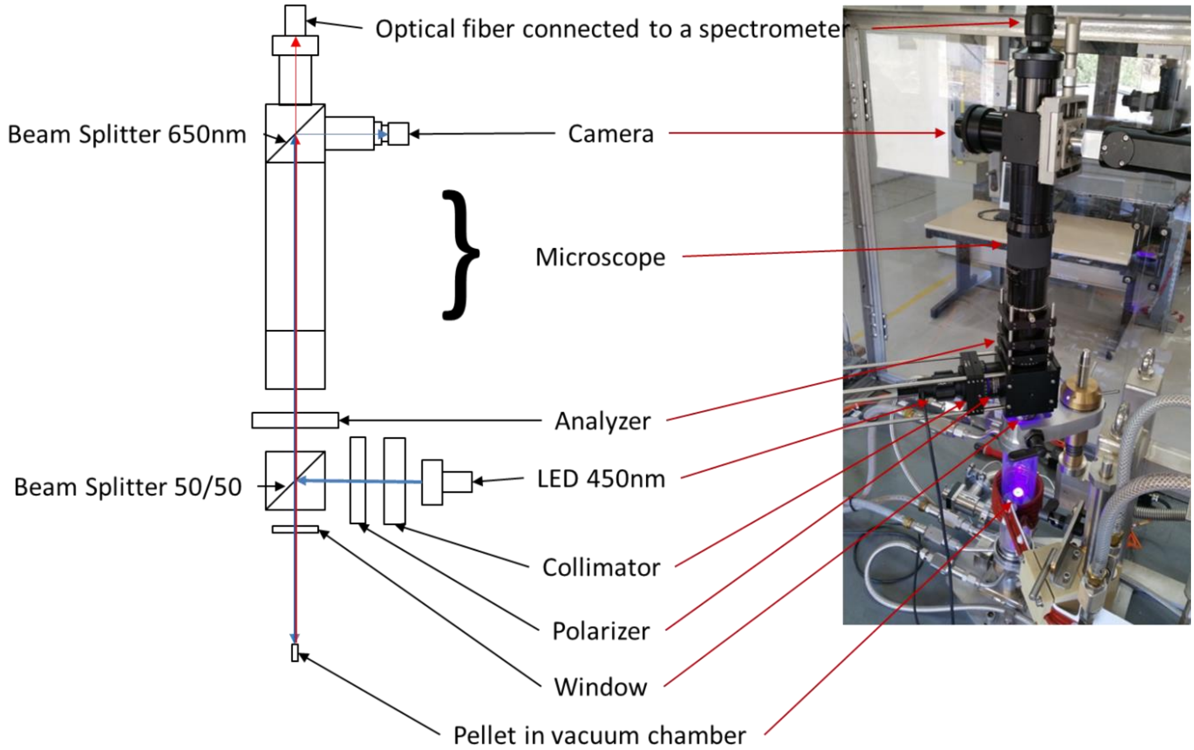

Fig 2. Schematic and photo of the optical device operating on the Merarg mock up experiment

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 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 50mW, corresponding to a power density of $1.7W/cm^2$ and resulting in negligible heating of the sample. Additionally, as the system should also allow thermal radiation measurements, a 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 5.5µm 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 the dynamics of transient events such as fracturation. We report on Fig.3, on the observation of a UO_2 sample on the MERARG mock up experiment with the optical system, at ambient temperature. The black part corresponds to the top of the crucible in this image. As shown in the Figure, fine details such as a fracture on the sample can be resolved. A color camera has been chosen 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.

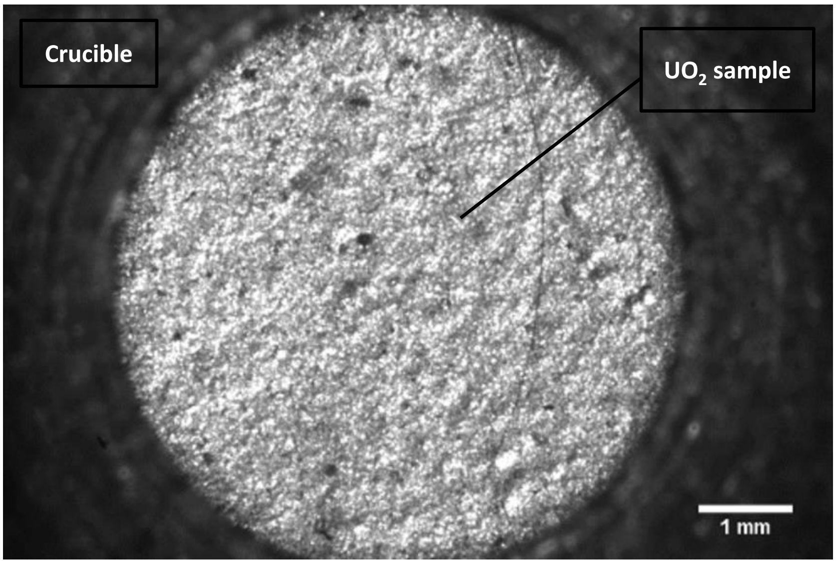

Fig 3. Observation of a UO_2 pellet with the optical system

2.3 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 requires the knowledge of the sample emissivity to retrieve the temperature from radiance measurements. Reliable data are available for the total emissivity of UO_2, as reported for instance in the review by Fink on the thermophysical properties of UO_2 [10]. The reported emissivity varies little with temperature and is only a weak function of wavelength and a value of 0.85+/-0.05 is recommended for UO_2.This agrees with the value of 0.83 which was determined by Manara et al. for the normal spectral emissivity of UO_2 [11]. 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, but 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 [12] and are suitable for operation in extreme environments encountered in the nuclear field [13]. Such techniques were employed successfully by Manara and al to measure thermograms and spectral emissivity of oxides such as UO_{2+x} [14]. Therefore, we have coupled such a diagnostic to the optical system we developed for MERARG.

Obviously, some compromises had to be found on the targeted temperature range and we have restricted the range of measured temperature to 800-2000°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. As a beam splitter to separate the imaging and specral pyrometry port we have used a long pass dichroic mirror with 650nm cutoff (Thorlabs DMLP650R). A silica optical fiber with 1mm core is used for collection (Thorlabs M35L02), which corresponds to 1.5mm 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 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 [15] which is favorable to this approximation.

To determine the spectral response of the device (imaging system, fiber and spectrometer) we have used a Tungsten-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 considered for measurements. An example of the acquired raw spectrum measured on a UO_2 pellet heated at 1200°C is shown on Fig.4 (curve in black), as the treated signal considering the transfer function (curve in blue).

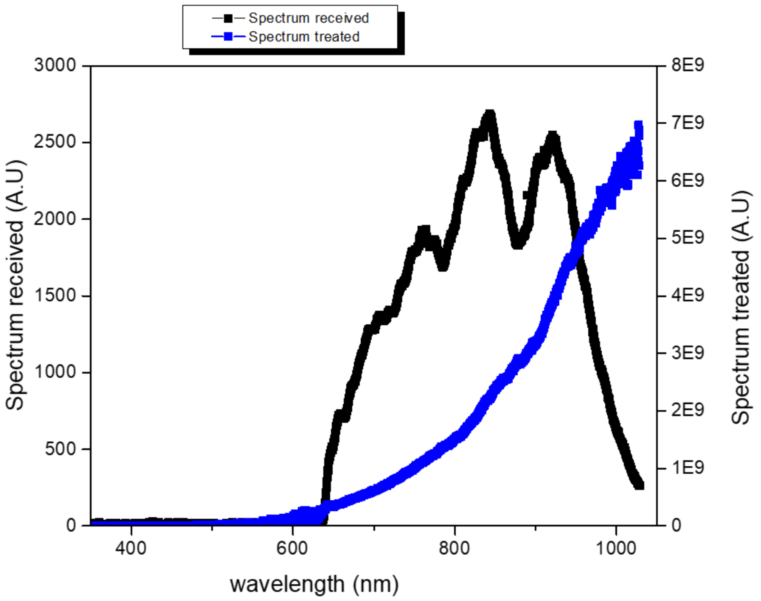

Fig 4. Spectrum as received from the spectrometer of a UO_2 pellet heated at 1200°C (black) and after processing with the inverse transfer function (blue)

3 Results

3.1 Experiments

In order to test the system in realistic conditions and assess the performances 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 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).

We have a good agreement between experimental values and theorical ones (Fig. 5). Indeed, the two curves are very similar and tend to overlap.

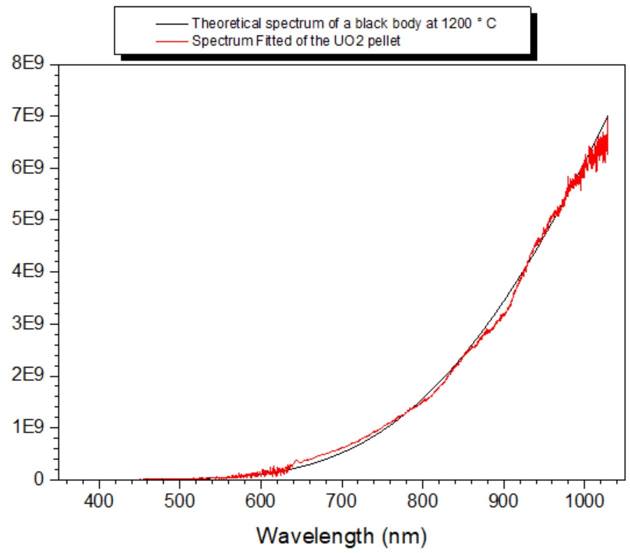

Fig 5. Measured spectrum of a heated UO_2 pellet (red) and fitted with Plank's law at 1200°C (black)

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 30 seconds plateaux to ensure that the temperature reached a steady-state regime.
Measurements were taken from 800°C, at this temperature the pellet begins to radiate sufficiently to obtain a usable spectrum, and then every 200°C. up to 1600°C. At 1700°C. the spectrometer saturated even at the lowest integration time. To make measurements above 1600°C, a neutral density filter will need to be introduced in the system. We report on Fig. 6 the temperature determined with the multispectral system which is compared to the crucible temperature (pilot TC) and the estimated sample temperature, as determined through the thermal qualification phase described previously. The data are in reasonable agreement and suggest a temperature determination with an uncertainty of ±50°C at high temperature. This range could be refined after further work on the calibration, which would require access to a black body at very high temperature.

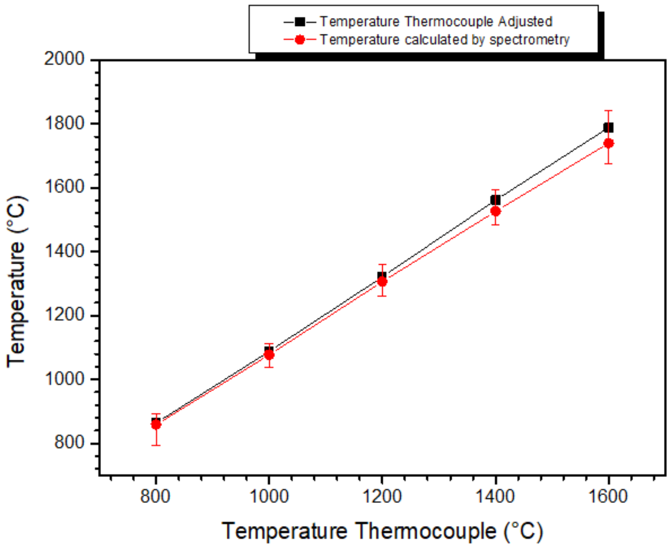

Fig6. Temperature of the sample from the thermal qualification phase and as determined by multispectral pyrometry.

3.2 Imaging

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 large range of temperature, from ambient to more than 2000°C. Indeed at low temperature there is no thermal emission in the visible and a blue LED is used as the illumination to acquire images in reflection mode. However, at high temperature the thermal radiation overcomes any illumination system that could be implemented, and the sample need to be imaged in emission mode. Experiments were conducted up to temperatures of 1800°C and by adjusting the camera exposure, it was found no limitations on the imaging system. Imaging at higher temperatures can be done by reducing further the integration time.

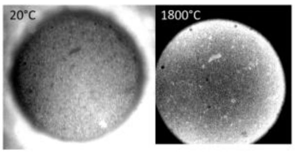

Fig 7. Observation of a UO2 sample at ambient temperature and at 1800C.

The ability of the system to capture transient events is also shown of Fig8 with the observation of cracking of the pellet during a rapid cooling sequence.

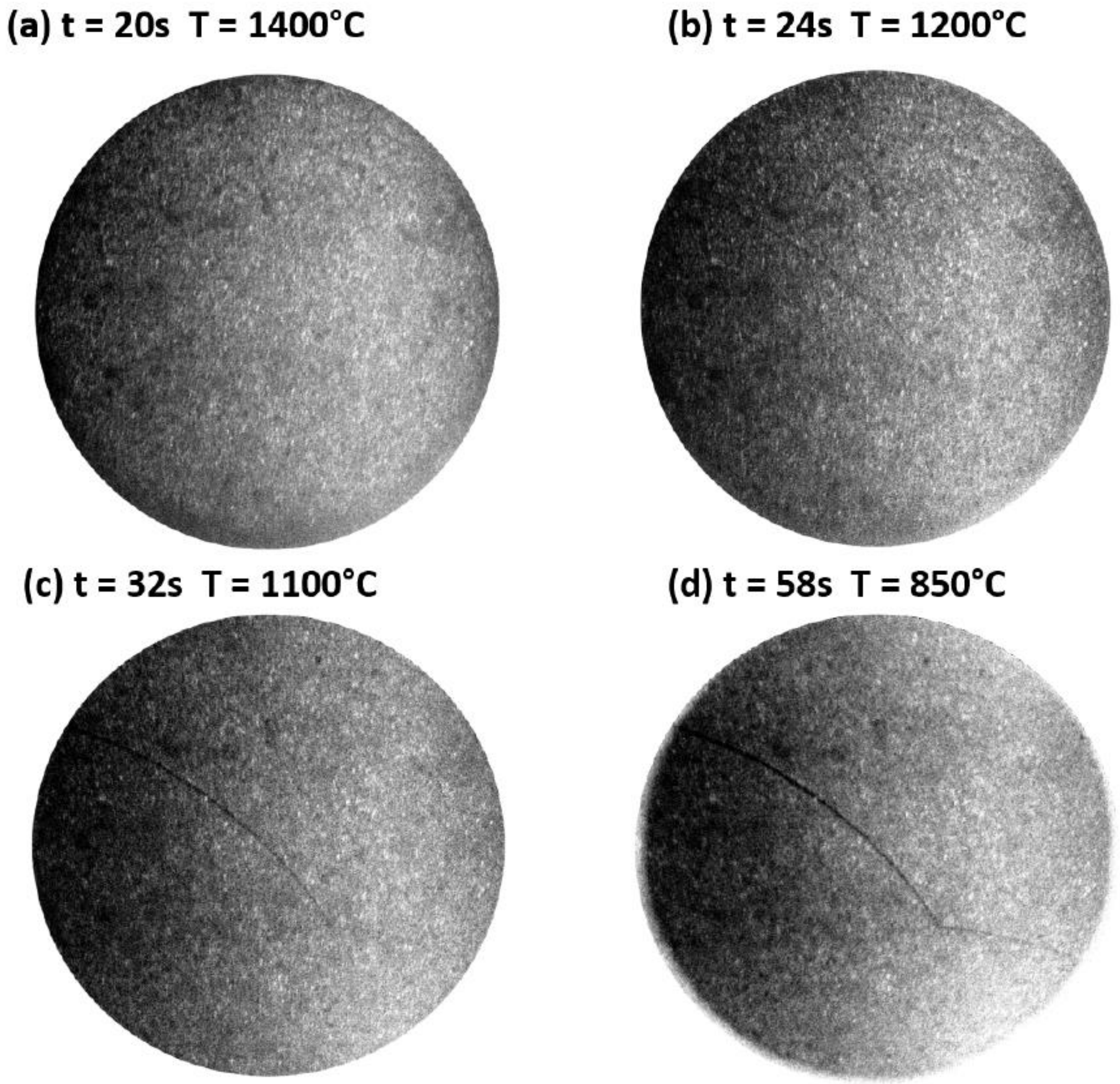

Fig 8. Image sequence illustrating the cracking of the pellet during a cold phase

4. Conclusion

The optical system proved its ability, to observe a UO2 pellet in the Merarg enclosure, and those of an initial ambient temperature at a maximum temperature of 1800 ° C, while simultaneously measuring the temperature of the pellet on the observed surface. Once this system put on a hot cell, the CEA, will be able to when the cracks happens and at which temperature the pellet was, and crossed this data with the gas γ spectrometer. This will provide additional information for instance on the micro structure evolution kinetics and fuel fragmentation, as infrared thermography to monitor and control the fuel temperature.

5. Acknowledgments

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6. References

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