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# Optical pyrometry measurement on oxidized Zircaloy-4 cladding

**B Bouvry<sup>1\*</sup>, L Ramiandrisoa<sup>1</sup>, C Cheymol<sup>1</sup>, N Horny<sup>2</sup>, T Duvaut<sup>2</sup>, C Gallou<sup>1</sup>,  
H Maskrot<sup>1</sup>, C Destouches<sup>3</sup>, L Ferry<sup>4</sup> and C Gonnier<sup>5</sup>**

<sup>1</sup> Den – Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France

<sup>2</sup> GRESPI, Université de Reims Champagne Ardenne, Campus Moulin de la Housse, 51100 Reims, France

<sup>3</sup> CEA Cadarache, DEN/CAD/DER/SPEX/LDCI, F-13108, St-Paul-lez-Durance, France

<sup>4</sup> CEA Cadarache, DEN/CAD/DER/SRJH/LEDI, F-13108, St-Paul-lez-Durance, France

<sup>5</sup> CEA Cadarache, DEN/CAD/DER/SRJH, F-13108, St-Paul-lez-Durance, France

\*benjamin.bouvry@cea.fr

**Abstract.** In order to improve the safety of nuclear power plant, loss-of-coolant accident experiments are implemented in research reactor. In this framework, we develop an optical pyrometry device to measure surface temperature (700-1200°C) of Zircaloy cladding without contact. The whole set-up of the simplified device (under air, without radiation) and the measurement procedure including data treatment based on bichromatic pyrometry are presented, as well as results for various temperature levels. Temperature retrieval based on the hypothesis of emissivity ratio equal to a constant, is scanned over a large wavelength range. A rather constant surface temperature is obtained on the spectral range of measurement, confirming the relevancy of emissivity hypothesis. Differences between this non-contact temperature measurement and a complementary thermocouple temperature measurement are also discussed.

## 1. Introduction

One of the most accidental scenarii considered in nuclear power plant design is the LOCA (Loss Of Coolant Accident) which implies a large break on the primary circuit and a violent pressure decrease (from about 150 bars to few bars). This dewatering makes the fuel clads overheat (up to 1200°C)(1). The oxidation by steam at high temperature weakens claddings, which can burst and release fission products. To simulate the LOCA, the Light-water One Rod Equipment for Loca Experimental Investigations (LORELEI) test device will be implemented in the Jules Horowitz research reactor under construction at the CEA Cadarache center. The aim is mainly to test Zircaloy-4 claddings behavior in hard environment where steam oxidation is enhanced by high temperatures (from 700 up to 1200°C)(2). A monitoring of the surface temperature of cladding is necessary to link burst conditions with temperature but a measurement by thermocouples is not acceptable because it would create a cold point on the cladding that generates a modification of burst conditions. The device for temperature monitoring should be non-invasive as well to minimize the perturbation of the neutron flux coming to the cladding into LORELEI. The only solution for a distant temperature measurement, available in the LORELEI device, is the optical pyrometry focused on the infrared spectral range. According to literature, this



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temperature measurement technic has never been applied on a study device of LOCA. This leads to design a non-contact real time (1Hz) sensor suited to these extreme conditions (high temperature, nuclear radiations and steam environment) and with an uncertainty of about  $\pm 10^\circ\text{C}$ . In a first step of development, an optical pyrometry device is then developed in laboratory to measure the surface temperature of Zircaloy cladding without contact, under air, without steam or radiation (3). The aim is to test the measurement capability of pyrometry method on Zircaloy-4 cladding at high temperature and study the emissivity property of cladding without additional perturbation. Thermocouples will be only used in this experimental device, to give a temperature reference. When it will be consolidated, the emissivity profile will be applied to measurement in more severe conditions.

## 2. Experiment

### 2.1. Zircaloy cladding sample

In current nuclear reactors, pellets of nuclear fuel are inserted in Zircaloy-4 claddings and are integrated in the reactor core. The visual aspect of a new cladding is metallic color. During the reactor operating, a thin layer of black color oxide shortly covers the new cladding, growing up to around a hundred of microns at the end of the life time of the fuel in the reactor core. So, we use an experimental facility named EDGAR 2, located at the CEA Saclay research center, to oxidize claddings in air/steam environment at high temperature. The cladding used in this study was oxidized at a temperature of  $1100^\circ\text{C}$  for 100s.

The Scanning Electron Microscopy (SEM) picture presented in Figure 1 (a) shows a cross-section of an oxidized cladding: the thickness of the oxide layer is about  $15\ \mu\text{m}$ . The Energy Dispersive Spectroscopy (EDS) measurement presented in the Figure 1 (b) and corresponding to the SEM picture shows the relative composition of the cladding with the depth. The discontinuities in oxygen and zirconium rates, red (bottom) and blue (top) lines in Figure 1 b), respectively, between  $15$  and  $16\ \mu\text{m}$ , mark out the transition between the oxide and the metallic base and corroborate the SEM measurement. The EDS analysis shows that the oxide is mainly composed by Zirconium and Oxygen elements, which induces that oxide has a chemical composition close to zirconia material ( $\text{ZrO}_2$ ).

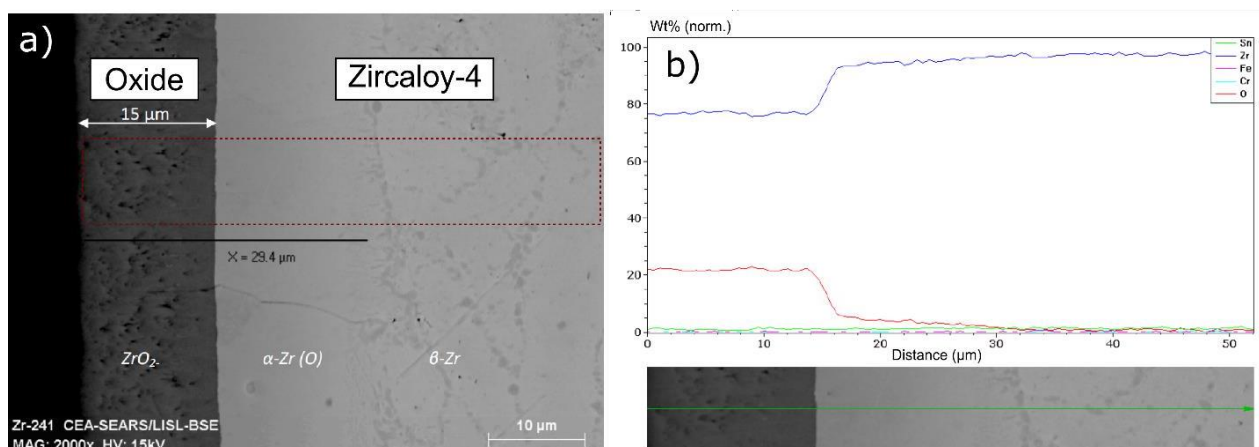


Figure 1: a) SEM picture of oxidized Zircaloy cladding and b), EDS measurement (from outside surface of oxide to the depth of the cladding)(3). The black layer on the left part of Figure 1 a) corresponds to the resin coating used for SEM measurement.

### 2.2. Optical pyrometry set-up(3)

This optical pyrometry method is based on the comparison between the radiance emitted by the sample and by a heat source with known temperature and emissivity ( $\epsilon$ ), generally a black body ( $\epsilon \sim 1$ ), used as reference, at the same wavelength. Our reference thermal source is a cylindrical black body furnace (HGH RCN1200N1) working up to a temperature of 1200°C, with an aperture of 12.5 mm.

An objective (made of two converging lenses), collecting thermal radiation at a distance about 10 cm from heat source or surface sample on an analysis area with a diameter about 2 mm, is located first in front of the heat source. This set of lenses focuses thermal flux emitted by the centre of the aperture, on the end face of an optical fibre. We use an optical fibre in the pyrometry set-up to carry the optical power to the detector, as the future sensor will do it in the research reactor. On the LORELEI device the fibre will be a pure silica core (or slightly doped silica core) fibre in order to limit the attenuation due to nuclear radiations, the optimum spectral range will be then: 0.9-1.6  $\mu\text{m}$  (4). In order to select the most relevant wavelengths for our technical specifications, we used an infrared spectrometer Ocean Optics NIRquest 512. The photodiode detector allows measurement in a spectral range from about 0.9  $\mu\text{m}$  to 1.7  $\mu\text{m}$  with a resolution of about 1.5 nm.

Figure 2.a) presents a picture of the heating holder, developed (by the ThermoEst company) for this study, and suited to the cladding sample. The holder has a cylindrical shape and allows heating up to about 1000°C. Figure 2.b) presents a thermography picture of the Zircaloy cladding equipped with a welded type N thermocouple. The optimum analysis area for pyrometry measurement, as well as the thermocouple location, can be observed on this figure. This preliminary observation made by a bolometric infrared camera (FLIR systems, SC655 LS), working on the spectral range 7-12  $\mu\text{m}$ , allows us to observed the temperature homogeneity on the sample which is mainly located between 5 and 8 cm (centre of the cladding) above the holder basement. The thermocouple is placed 1 cm below the pyrometry measurement area. Locally, oxide is withdrawn by polishing and thermocouple wires are welded on the surface of zirconium alloy.

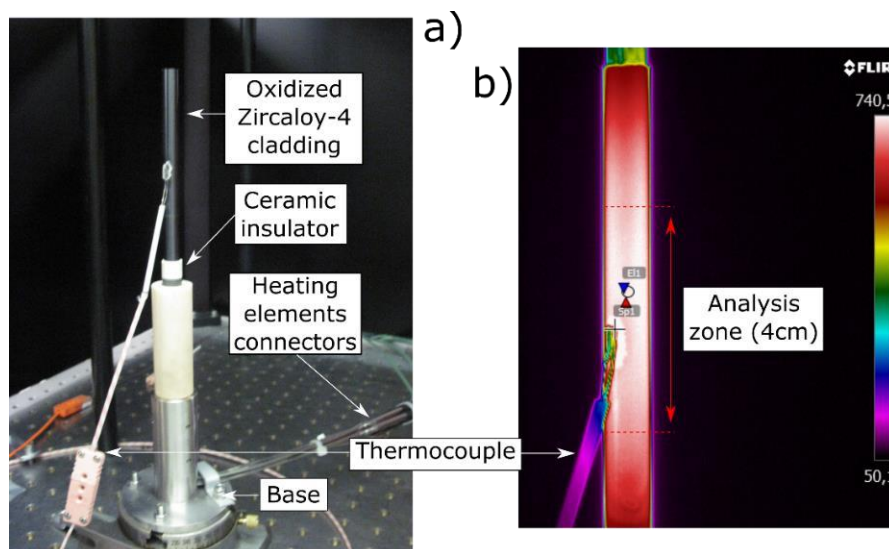


Figure 2: a) Description of sample holder and b) Thermography picture, obtained by an infrared camera, for a temperature setting 805°C and showing the relevant analysis zone for pyrometry measurement.

### 2.3. Experimental procedure

Figure 3 presents a diagram of the laboratory pyrometry set-up where sample holder, black body, objective, infrared spectrometer and optical fibre are shown.

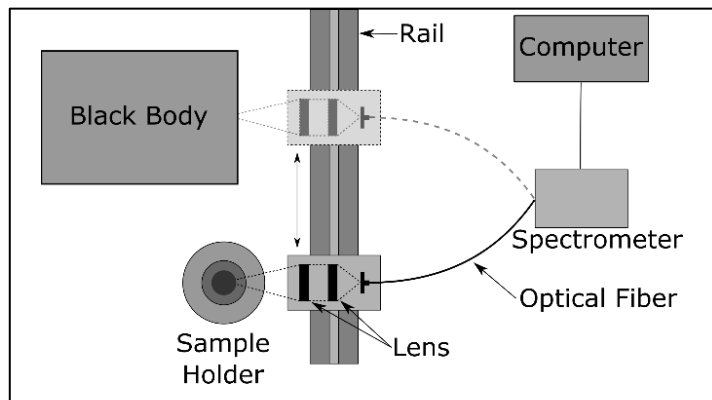


Figure 3: Diagram of pyrometry device for surface temperature measurement of Zircaloy-4 cladding.

In this paper, two setting temperatures of the holder will be studied: 805°C and 905°C, called  $T_1$  and  $T_2$  in the following of the text, respectively, temperatures at the center of the heater and far from the surface. The first experimental step of the measurement procedure, the calibration (3), consists in collecting the power from the black body at different setting temperatures. The temperatures of this source are chosen so that the range of emitted powers covers those emitted by the heated sample. In our procedure, four black body temperatures are used. Therefore, the reference source is heated from 710 to 740°C, with a step of 10°C, and from 820 to 850°C, also with a step of 10°C, for setting temperatures of the holder  $T_1$  and  $T_2$ , respectively. Indeed, the thermal insulated air layer between the heater and the cladding, as well as various thermal conductivities of Zircaloy and oxide, lead to a temperature difference between the setting and the surface temperatures. The (optical) power from the black body is measured for each temperature by the infrared spectrometer (result in “number of counts”). For each wavelength, the result is “plotted” as a function of the radiance, deduced from Planck’s law. The linear regression obtained with the 4 points is the transfer function. Each wavelength has its own transfer function. The integrating times of the spectrometer must be adapted with the aimed sample temperature because of saturation problem of detector but must be the same for sample measurement and calibration. For this study, 102 ms and 52 ms integration times are selected for sample holder temperatures  $T_1$  and  $T_2$ , respectively. The integrating time is set so that it works as much as possible on the linear range of the detector.

In the second step of the procedure, the measuring device (lens, optical fibre and spectrometer) is translated in front of the sample. The distance between lens and sample is identical to those used for calibration. Number of counts from cladding surface is measured by the infrared spectrometer and is converted into radiance spectrum thanks to the transfer function.

### 2.4. Temperature retrieved by bichromatic method

Several pyrometry techniques may be applied to retrieve the temperature (5). We focus here on the bichromatic method based on the hypothesis of known emissivity ratio between two wavelengths. Using Wien approximation, acceptable on the spectral range of measurement, the temperature can be retrieved as shown in (1).

$$\frac{1}{T} = \frac{\lambda_1 * \lambda_2}{\lambda_1 - \lambda_2} * \frac{1}{C_2} * \ln \left( \frac{f^{-1}(S_1)}{f^{-1}(S_2)} * \frac{\varepsilon_2}{\varepsilon_1} * \left( \frac{\lambda_1}{\lambda_2} \right)^5 \right) \quad (1)$$

With  $f$ , the transfer function of spectrometer, calculated from black body calibration, and,  $f^{-1}(S_1)$  and  $f^{-1}(S_2)$ , the radiance calculated from signal  $S_1$  received at wavelength  $\lambda_1$  and signal  $S_2$  received at wavelength  $\lambda_2$ , respectively. The values  $\varepsilon_1$  and  $\varepsilon_2$  are the emissivity values at  $\lambda_1$  at  $\lambda_2$ , respectively. Based on the hypothesis of the emissivity ratio  $\varepsilon_2/\varepsilon_1 = 1$ , temperature is retrieved for each couple ( $\lambda_1, \lambda_2$ ) selected in the spectral range considered, resulting in an extended bichromatic method. Different interval values  $\lambda_2 - \lambda_1$  (50, 100, 150, 200 and 250 nm) are tested and mean temperature values are calculated as well as the standard deviations.

### 3. Results and discussion

The experimental calibration procedure, described in the previous part, is used to measure the power from black body, in order to calculate the transfer function. Figure 4 presents intensity spectra measured on black body, for different setting temperatures (710, 720, 730 and 740°C). We can observe on Figure 4 a) a local depletion around 1.38  $\mu\text{m}$ . It is due to absorption bands of water vapour, present in atmosphere(6, 7). As the vapour concentration may fluctuate between calibration and sample measurements, the spectral range 1.3-1.45  $\mu\text{m}$  is removed from calculation. The spectral range beyond it (from 1.45 to 1.6  $\mu\text{m}$ ) is not selected either for calculation because of the few number of wavelengths usable for bichromatic calculation. Figure 4 b) presents also transfer functions calculated for two wavelengths (1  $\mu\text{m}$  and 1.25  $\mu\text{m}$ ).

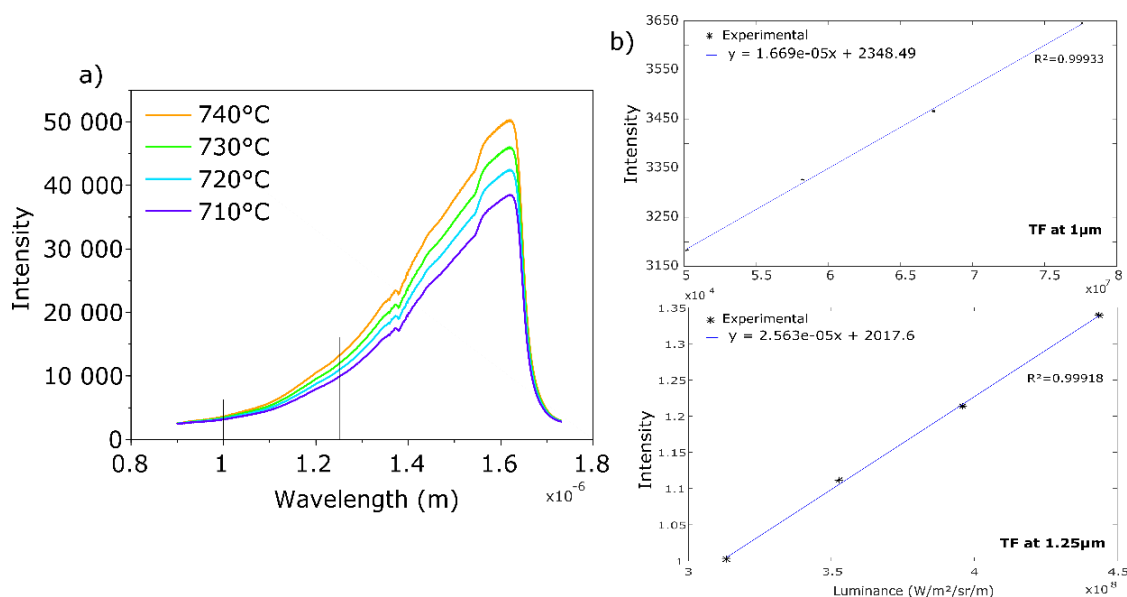


Figure 4: a) Intensity spectra from black body for temperatures from 710 to 740°C and b), Transfer functions calculated at 1 $\mu\text{m}$  and 1.25  $\mu\text{m}$ .

Figure 5 shows a scanned bichromatic temperature calculation, in the spectral range from  $\lambda_1 = 1.0 \mu\text{m}$  to  $\lambda_2 = 1.3 \mu\text{m}$ , for holder setting temperatures  $T_1$  and  $T_2$  as well as the temperature measured by thermocouples ( $T_{TC}$ ), welded on the Zircaloy cladding surface, for each temperature level. Each point is the result of a bichromatic calculation for a wavelength couple. For example, the last point of the curve for a gap of 50 nm, corresponds to a calculation with  $\lambda_1 = 1.25 \mu\text{m}$  and  $\lambda_2 = 1.3 \mu\text{m}$ .

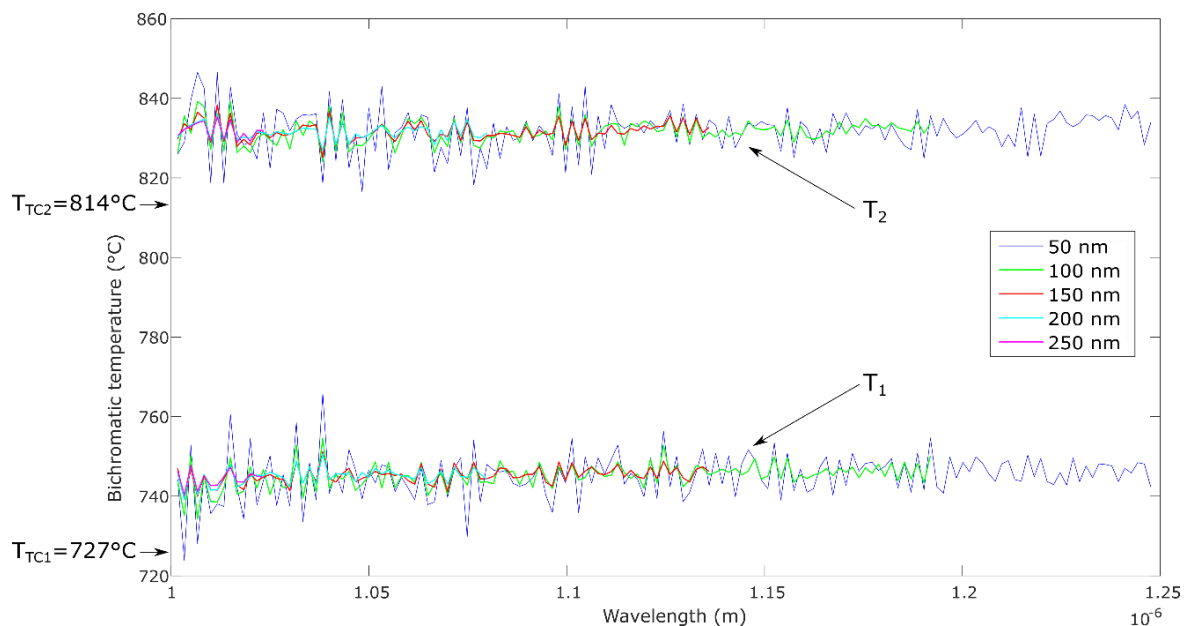


Figure 5: Scanned bichromatic temperature as function of the wavelength  $\lambda_1$ , with calculation made for several gaps (from 50 to 250 nm) and both  $\lambda_1$  and  $\lambda_2$  included in the wavelength range: 1-1.3  $\mu\text{m}$ , for two sample holder temperatures  $T_1$  and  $T_2$ , with an emissivity ratio hypothesis  $\varepsilon_2/\varepsilon_1 = 1$ .

The rather constant value of the temperature, for each experiment and wavelength gap presented on Figure 5, makes the calculation of a mean temperature, as well as the associated standard deviation, relevant. Table 1 gathers the thermocouple temperatures and the means of scanned bichromatic temperatures. All gaps of wavelengths lead to similar mean bichromatic temperature values for the two temperatures  $T_1$  and  $T_2$ . As shown in Figure 5, there are strong fluctuations of the bichromatic temperature for the interval of 50 nm, confirmed by the highest value of the standard deviation of about 6°C (cf. Table 1). These standard deviations decrease with the increase of the wavelength interval until 150 nm, as expected with non-correlated fluctuation of the radiance (5). The deviations obtained, due mainly to the fluctuation of the response of each pixel of the CCD array, corresponds to variations in radiance measured of about 1%.

Table 1: Thermocouple temperatures ( $T_{TC}$ ) and bichromatic temperatures (hypothesis:  $\varepsilon_2/\varepsilon_1 = 1$ ) for wavelength intervals from 50 to 250 nm and a step of 50 nm, obtained for two holder setting temperatures  $T_1$  and  $T_2$ , and the standard deviation.

$T_{SH}(\text{°C})$	$T_{TC}(\text{°C})$	Bichromatic temperatures (°C)				
		$T_{50\text{nm}}$	$T_{100\text{nm}}$	$T_{150\text{nm}}$	$T_{200\text{nm}}$	$T_{250\text{nm}}$
$T_1$	727	$743 \pm 6$	$743 \pm 3$	$743 \pm 2$	$743 \pm 2$	$744 \pm 2$
$T_2$	814	$832 \pm 6$	$832 \pm 3$	$832 \pm 2$	$832 \pm 2$	$833 \pm 2$

Actually, these standard deviations are comparable to the uncertainties associated to the bichromatic temperature measurement resulting from complete calculation taking into account uncertainties on radiance measurement for each pixel of detector, correlation of fluctuation on various pixels and uncertainties on transfer function parameters and (3, 8). Uncertainty about the real surface temperature value, when the emissivity profile is unknown, is the reason why a reference measurement by thermocouple was used. The type N thermocouple has a specified measurement uncertainty of about  $\pm 6^\circ\text{C}$  between 700 and 900°C. We can observe a temperature difference of about 17°C between pyrometry temperatures and thermocouple values (cf. Table 1). This temperature difference is constant

for the two holder setting temperatures  $T_1$  and  $T_2$ . This discrepancy is probably not entirely due to the error of optical pyrometry measurement, but may be induced by thermal losses due to a temperature gradient inside thermocouple wires, creating a cold point on the cladding surface (3). Gradient issues prevent accurate measurement of the surface temperature by thermocouples, and make difficult the comparison with optical pyrometry results. Nevertheless, considering the thermocouple temperature as reference, the accuracy of pyrometry measurement error is  $<20^\circ\text{C}$ .

By the knowledge of temperature and radiance, one can deduce emissivity values of cladding of 0.9 and 0.89 for the holder-setting temperatures  $T_1$  and  $T_2$ , respectively. Calculated emissivity values of oxidized Zircaloy-4 cladding are quite similar to the values found in literature in the same spectral and temperature ranges (9). An uncertainty of about  $\pm 6^\circ\text{C}$  on temperature gives an uncertainty on emissivity of about  $\pm 6\%$  in the spectral range 1-1.3 $\mu\text{m}$ . No modification of the surface cladding and oxide layer thickness has been noted after the experiment.

However, are these temperature and emissivity, obtained by pyrometry measurement with the hypothesis  $\varepsilon_2/\varepsilon_1 = 1$ , the true ones? Indeed, others available profiles of  $\varepsilon(\lambda)$  can also lead to a quite constant temperature profile. For example, bichromatic temperatures, for a gap of 100 nm and calculated for an emissivity ratio  $\varepsilon_2/\varepsilon_1 = 0.995$ , corresponding to exponential emissivity profile, are presented in Figure 6 for a temperature setting  $T_2$ . It leads to a mean temperature of  $826^\circ\text{C}$ , not so far from the mean temperature calculated with emissivity ratio  $\varepsilon_2/\varepsilon_1 = 1$ . The difference between pyrometry and thermocouple temperatures decreases from  $17^\circ\text{C}$  to about  $12^\circ\text{C}$ , which is a temperature difference not so far from the output of preliminary modelling results of the bias of thermocouple measurement [3]. The emissivity profile should be determined once this bias will be consolidated.

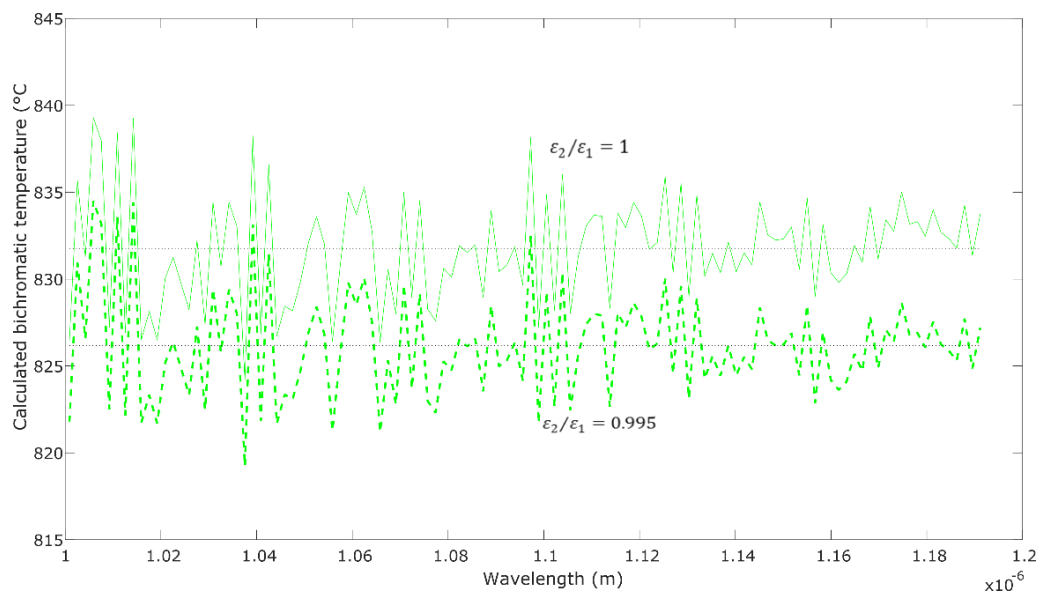


Figure 6: Scanned bichromatic temperatures as function of the wavelength  $\lambda_1$ , calculation made for a gap of 100 nm for a sample holder temperature of  $T_2$ , with emissivity ratios of 1 (solid line) and 0.995 (dashed line). Horizontal lines are mean temperatures associated.



#### 4. Conclusion

A non-contact temperature measurement is needed rather than thermocouple for the LORELEI test device because it will not disturb the burst conditions of the cladding. In this condition, a simplified optical pyrometry set-up has been developed to test the temperature surface measurement capability of optical pyrometry on Zircaloy-4 cladding and study emissivity property of oxidized cladding under air without radiation. The whole set-up and the experimental procedure have been presented as well as the retrieval of temperature based on an extended bichromatic method. A difference of about 17°C was noted between pyrometry and thermocouple measurement. A rather constant emissivity of 0.89 was calculated for the two holder setting temperatures. The hypothesis of an emissivity ratio equal to 1 appears to be relevant for both setting temperatures of holder  $T_1$  and  $T_2$ . However, other emissivity ratio, close to 1 and corresponding to a little variation of  $\varepsilon(\lambda)$  also lead to relevant temperature values with this method. One perspective of this study is a comparison of bichromatic results with those calculated by others pyrometry methods like monochromatic or polychromatic technics. This last method may allow us approaching the true temperature and emissivity profile by a study of “residues” after minimization process. Another way to consolidate the true temperature is a complete study of the bias of thermocouple measurement. A miniaturization of optical pyrometry set-up is also expected for an introduction into LORELEI device.

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