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Simulation of Reactivity Initiated Accident thermal transients on nuclear fuels with laser remote heating

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Abstract

The investigation on the behaviour of nuclear fuels under thermal loads representative of RIA (Reactivity Initiated Accident) conditions in nuclear reactors is a significant challenge. In this work we introduce a new concept of a laboratory experiment relying on high power lasers to reproduce the temperature gradients experienced by nuclear fuel pellets during such events. The concept is experimentally demonstrated on UO_2 samples, supported with numerical simulations to estimate the temperature gradients within the samples. This is a first step in the perspective of applying the technique on irradiated fuels.

Keywords: Nuclear Fuels, Uranium Oxyde, Annealing

The understanding of the behaviour of nuclear materials in relation to the different thermal loads to which they can be subjected, i.e. during nominal operation or accidental conditions, is of paramount importance in the nuclear field. By improving the knowledge, it is possible to upgrade nuclear reactor safety and to precisely define specific on-site emergency plans in the case of an extreme event. Predicting correctly the fission gas (FG), fission product (FP) releases (i.e. the source term) and the corresponding fuel fragmentation 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 the specific case of the so-called RIA[1], a fast rise of nuclear power and of temperature occurs in the fuel. The power excursion may lead to failure of the nuclear fuel rods and release of radioactive material into the primary reactor coolant. In the most severe cases, the fuel rods may be shattered and a large amount of the fuel pellets can be dispersed into the primary coolant. To prevent those potential consequences, safety

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- ¹⁵ criteria are usually set up to limit the enthalpy rise of the fuel. During a RIA event, the temperature distribution in the cylindrical pellet is almost flat top with a maximum localized in an outer ring closed to the pellet periphery[2]. Typical temperature increase above 2000 °C occurs in a few tens of millisecond on the edges of the fuel as illustrated by the simulation results reported in Fig. 1.
- Industry interests, for instance, in increasingly higher burnup fuels drove additional experimental emphasis. In order to study the impact of this particular accident, in the early 1990s, experimental programs, in the form of pulse irradiation tests performed in Material Testing Reactor (MTR), were launched in France, Japan and Russia in order to check high burn-up fuel behaviour under RIA. The pulse rector tests are done at

²⁵ condition that approximate, as closely as possible, those expected in PWR under RIA. Consequently, they provide crucial information on the fuel rod during the transient. These test programs were primarily intended to check the adequacy of regulatory acceptance criteria for RIA, which were mainly based on experiments on low (performed in MTR such as PBF [3], IGR [4], BIGR [5], NSRR [6] or CABRI [7]) or un-irradiated ³⁰ fuel [8].

Therefore, up to now, integral experiments (i.e. MTR type) are most often the only solution for this category of R&D. This means that the potential number of experiments that can be performed is very limited both in terms of their cost and their complexity. Additionally it is also more or less impossible to study the impact of specific parameters

- to the integral fuel rod behavior. To overcome these difficulties, the present reported work aims at exploring a concept to reproduce the so-called RIA (Reactivity Initiated Accident) thermal conditions in order to study the behaviour of nuclear fuel in such situations. For instance, it could be possible to investigate very important parameters such as correlation between FGR and fuel fragmentation, radial expansion in a fuel
- ⁴⁰ pellet (PCMI behavior) as a function of the thermal profile applied, fuel fragmentation during the test, etc... The corresponding general goal is to provide complementary results to those obtained under integral experiments which study the capacity of Pressurized Water Reactor (PWR) fuel based on uranium dioxide (UO₂) and mixed oxide fuel (MOX) to withstand sudden power transients that may occur during an accident scenario[9].

In order to produce such RIA thermal transients on nuclear fuel samples at the laboratory scale, we have explored the possibility to use a laser heating approach. Laser remote heating has indeed many very interesting characteristics for such an application: it is a contact-less technique which can easily drive materials to extreme temperatures

- with very high spatial precision and temporal control. In the field of nuclear fission research, laser heating devices together with analysis diagnostics and temperature monitoring systems have been successfully applied for the measurement of thermal and thermodynamic properties of nuclear fuels. [10, 11]. Nuclear fuels being highly absorbing materials to visible/IR wavelengths, the laser flux can be efficiently absorbed by the
- material: the reflectivity has an order of magnitude of 0.15 in the visible / near infrared range[12]. Based on the optical properties of UO₂ [12, 13], laser absorption is localized at the surface of the material and the laser heat source can only be used as an incoming heat flux on the sample surface. Experiments has then to be conceived accordingly. On the thermal point of view, considering the one-dimensional thermal diffusion length of
- $_{60}$ $2\sqrt{Dt}$, with D the diffusivity of the material and t the time of applied laser heating,

it occurs that with a diffusivity of $2 \times 10^{-6} m^2 s^{-1}$ [14], homogeneous heating in the depth of the material at the time scale of few tens of millisecond, corresponding to RIA temporal dynamics, is only obtained if the width of the sample is sub-millimetric. Therefore in order to conduct relevant laser heating experiments on fuel materials, the

- ⁶⁵ samples should be thin disks of less than 1 mm thickness cut from a fuel pellet if one want to reach homogeneous heating in the depth. In such conditions, the proposed scenario to reproduce the RIA spatio-temporal thermal transient is described schematically in Fig. 2. Based on a section of a fuel cladding assembly, the sample is maintained in nominal temperature conditions which can be done through laser heating with flat-top
- or Gaussian intensity distribution and adapted insulation/cooling. Then annular laser beams, coming from both sides, can be applied to reproduce the temperature gradients encountered in RIA conditions as illustrated in Figure 2.

Numerical simulations have been performed for the dimensioning of this experiment, and analysis of results that will be shown later, with Finite Element Method (COMSOL Multiphysics Software). The simulations were conducted on a UO₂ sample considering axial symmetry (2D simulations). The model can include Zircaloy cladding around the UO2 sample, and possible sapphire windows corresponding to the experiments that will be described below. The laser heat source was simulated as an incoming heat flux on the sample surface with the following intensity distribution:

$$I(r) = I_{max} e^{-2\frac{(r-R)^2}{\omega^2}}$$
(1)

⁸⁰ R being the radius of the annular beam, ω the beam waist. I_{max} is related to the power P with:

$$I_{max} = \frac{P}{\pi \left(\omega^2 2e^{\frac{-2R^2}{\omega^2}} + R\frac{\omega\sqrt{\pi}}{\sqrt{2}}erfc\left(-2\frac{\sqrt{2}R}{\omega}\right)\right)}$$
(2)

erfc(x) being the complementary error function. We have considered radiative losses at the boundaries but neglected convection and possible losses related to the sample holder. The minimum mesh size is $10\mu m$ in the laser heated area, and maximum $100\mu m$ in the sample center.

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With a typical thickness of the disk sample in the range of 0.5-1 mm, we found that few hundreds Watts of laser power for the transient annular laser beam heating are required to reach temperature close to $2000 \ ^{o}C$ in few tens of milliseconds.

- A pilot experiment has been designed and developed in order to test this concept (Fig. 3). It is based on a high power Ytterbium fibre laser (wavelength of 1080 nm) with 1500 W of available power. This laser has a typical rising time of few microseconds which is suitable to generate well-controlled millisecond pulses, and high optical quality (monomode Gaussian beam) that enables beam shaping generation with simple optical systems, as it will be described below. The laser is coupled in a dedicated
- experimental chamber equipped with laser windows and view-ports for optical monitoring, that can operate in vacuum (10^{-2} mbar) or controlled atmosphere (Argon). The thin disk sample is placed at the centre of the chamber in a mechanical assembly that has been designed to hold the sample vertically, with insulating materials, zirconia and alumina, that can withstand high temperatures (up to 2000 °C). Sapphire optical windows are used to maintain the sample and potentially retain the fragments in place in

case of loss of integrity, while simultaneously allowing optical access from both sides (sample assembly is visible in Fig. 3). The optical instrumentation around the sample is a key-point to monitor and control the temperature, therefore optical diagnostics such as thermal imaging and optical pyrometry can be implemented through the available view-ports of the experimental chamber.

In order to generate the annular beam for controlled energy deposition on the edges of the sample, a classical lens and positive axicon combination has been used[15]. A 8 mm beam diameter with a 150 μm width is obtained with a 13 mm incoming Gaussian beam (diameter at $1/e^2$), a 500 mm focal length and and a 1 degree axicon. The image

of the obtained ring in such conditions is shown in Fig 3, as measured with a laser beam profiler placed in the sample plane. By defocusing the lens it is possible to adjust the width of the annular beam and accordingly modify the temperature gradients. A second Ytterbium fibre laser, not shown in Fig. 3 is used to heat the sample up to the initial conditions of interest. This second laser is controlled by a Proportional, Integral, Derivative (PID) feedback loop, based on the temperature monitoring of the sample with a pyrometer, in order to ensure stable temperature conditions.

As a preliminary work before studying UO_2 samples, we have firstly worked on graphite samples. The objective was to validate the experimental concept on a stable material and compare experimental results and numerical simulations on a material

with well-known properties. We have worked with EDM-3 ultra-fine graphite type (POCO Industries), machined with similar dimensions as the UO₂ samples of interest. The samples were studied without the sapphire windows in order to perform spatially resolved measurements of the temperature with a thermal camera operating in the Long Infrared Wavelength Range (8-13 μm), where sapphire is opaque. An example of measurement is given in Fig.4 for the case of a 1500 W/100 ms annular laser pulse incident on the sample, initially at 300 °C. This is the highest power experiment up to

date on our system.

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In this example the peak temperature has reached 1800 o C with a heat affected area that extends up to the centre of the sample due to the high thermal conductivity of this material (100 W m⁻¹K⁻¹). Numerical simulations have been conducted taking into account the EDM-3 material parameters and their temperature dependencies extracted

from Ref.[16].

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Results of the simulations are displayed in Fig. 4 for the particular case under study. On this case corresponding to 1500 W of input power during 100 ms and many other series of experiments not shown, we have obtained a good correspondence between experimental data and simulations on the whole heating and cooling sequence. This

indicates that the developed numerical model has the ability to describe properly the thermal response of a sample submitted to the specific heat loads under consideration. For demonstration of the concept, we have used UO₂ samples with 8 mm diame-

ter and 0.7 mm thickness. They were synthesized in the UO₂ laboratory at the CEA Cadarache Centre using a standard process: the samples were synthesized with depleted UO₂ from a batch produced through wet process, followed by conventional pre-compaction (50 MPa), pressing (450 MPa) and sintering at 1650 °C for 2h under flowing H₂.

The samples have been inserted between two sapphire windows and mounted in the system described previously. The base pressure during experiments was 2×10^{-2} mbar. In order to reach a controlled initial temperature, a first laser beam with a Gaussian profile was used to heat the sample at 100 o C/s, with the PID regulation loop. The diameter of this laser beam was set to 1 cm (at $1/e^2$) in order to cover the whole

- ¹⁵⁰ sample surface and ensure an homogeneous heating. The temperature was controlled with an optical pyrometer operating at 2.3 μm , considering an emissivity of 0.85 for the sample and a transitivity of 0.88 for the sapphire window. The 2 laser beams with annular spatial profiles were then launched with the first laser beam still active, and the temperature profiles measured with a fast and high definition thermal camera operating in the mid infrared range (2-5 μm). We report in Fig. 5 the temperature profiles
- ing in the mid infrared range $(2-5\mu m)$. We report in Fig. 5 the temperature profiles corresponding to one experiment with a 20 ms pulse transient and a 1000W incident power.

The temperature distribution during the transient has the expected radial symmetry, as illustrated by the good correspondence of the extracted temperature profiles on each side of the sample. A localized temperature up to 2000 °C can be reached in few tens of milliseconds as requested by the RIA thermal conditions with spatial control, while keeping the sample integrity. The reported temperatures correspond to the typical objective of our experiment, as it was shown on Fig. 1. It can however be noticed that if the peak temperature can be reached, the thermal profile displayed on Fig. 1 is not dynamically reproduced: particularly the flat radial temperature profile in the inner part of the pellet. It could however be reproduced on our experimental system by combining

- a second high power laser with a flat top intensity distribution to the annular laser beam. Such a laser will be integrated in future experiments to have all the required flexibility to reproduce the thermal gradients of an RIA event.
- For the comparison of our experimental results to simulations we have adapted the model previously described to UO₂ samples by using the thermophysical material properties described in Ref. [17]. For the optical properties we have used reflectivity values calculated from refractive index measured by Bober et al in the visible range[12]. The different power losses in the system, for instance by reflection on the sapphire windows, were taken into account in the simulations, as the initial steady state temperature
- gradient and the measured laser beam profiles. Despite the simplifications used in the simulations, we have obtained a good correspondence between simulation and experiments, as illustrated in Figure 6. These results give confidence in using such simple numerical models as a guide to design experiments to reach the required RIA thermal transients. In order to improve the in-depth temperature homogeneity in these exper-
- iments (as shown in th insert of Figure 6) thinner samples have to be used, ideally sub- $100\mu m$. However, this brings many technical issues, particularly maintaining sample integrity, which are currently being tackled. Extension to the case of high burnup UO₂ should be also very challenging.
- As a conclusion we have presented an original method based on high power lasers to reach the thermal conditions of an RIA event on a nuclear fuel sample. The concept was tested and demonstrated on natural UO_2 samples, after a calibration phase conducted on graphite samples to validate the numerical simulations that are used to estimate the temperatures. Transient localized temperatures up to 2000 o C can be
- reached, with a spatial distribution on the edges on the sample that approaches the ones encountered in an RIA event. This work will now be continued to optimize the temperature profile and integrate such an experiment in a hot cell environment in order to

work on irradiated fuels.

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Figure 1: Simulations of a RIA event with ALCYONE[2], a fuel performance code codeveloped by the CEA, EDF and FRAMATOME. Fuel pellet temperature radial profile evolution during a pulse irradiation is plotted before the beginning of the pulse (t1), during the power rise (t2), at peak power (t3), during the power descent (t4), at the end of the power descent after the pulse (t5), and just before return to cold state (t6). 0 corresponds to the center of the sample.

Figure 2: Illustration of the proposed concept to reach RIA thermal transient conditions on a fuel sample: annular laser beams are used to deposit the energy on the periphery of a pellet section.

Figure 3: Sketch of the experimental chamber designed for the experiments. Bottom left: picture of the sample holder assembly at the centre of the vacuum chamber. A UO_2 disk, 8 mm diameter and 1 mm thick is visible in the sample holder. Bottom right: image of the beam profile obtained in the sample place with a laser beam profiler.

Figure 4: Measured (points) and calculated (lines) temperature at the surface of a graphite sample submitted to 1500 W during 100 ms. 0 corresponds to the centre of the sample and results are plotted as a function of the radius.

Figure 5: Measured temperature at the surface of a UO_2 sample submitted to a 470 W annular beam irradiation during 20 ms on both sides. Image: thermal camera snapshot at the end of the laser pulse ; Data plot: extraction of the temperature along vertical profile of the thermal images at different times.

Figure 6: Comparison between measurements of the radial temperature profile at the surface of UO_2 (dots) and simulated ones (lines) for the case of a 20 ms, 700 W laser pulse. The insert reports on 2D temperature distribution at the end of the laser pulse (t=20 ms).



Fig. 1



Fig. 2







Fig. 4



265 Fig. 5



Fig. 6