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Laser pulse heating of nuclear fuels for simulation of reactor power transients

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Abstract. It is important to study the behaviour of nuclear fuels under transient heating conditions from the point of view of nuclear safety. To simulate the transient heating conditions occurring in the known reactor accidents like loss of coolant accident (LOCA) and reactivity initiated accident (RIA), a laser pulse heating system is under development at BARC, Mumbai. As a prelude to work on irradiated nuclear fuel specimens, pilot studies on unirradiated UO_2 fuel specimens were carried out. A laser pulse was used to heat specimens of UO_2 held inside a chamber with an optically transparent glass window. Later, these specimens were analysed by metallography and X-ray diffraction. This paper describes the results of these studies.

Keywords. Laser applications; nuclear fuel elements; nuclear safety.

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1. Introduction

The safety of nuclear power reactors requires a thorough understanding of phenomena like loss of coolant accident (LOCA) and reactivity initiated accident (RIA). At high burnup, these phenomena become especially hazardous, as the probability of cladding failure goes up with the increased corrosion and hydriding [1]. The common feature of these accidents is the transient heating of the fuel to high temperatures. This can lead to fuel cracking, pellet-clad mechanical interaction (PCMI) and/or sudden fission gas release, all of which can accelerate cladding failure. It is important to study these phenomena, especially as we are about to embark on new reactor concepts about which no previous operating experience exists.

Instrumented fuel pins undergoing ramp tests can give valuable information about these phenomena. But facilities for making such fuel pins, and carrying out test irradiations for the power levels required are still not available in India, and we can only aim to participate in international cooperation towards such experiments in the near future. Meanwhile, to begin understanding these phenomena, attention may be given to simpler instruments which can simulate the transient heating very well.

Laser beams offer both high intensities required for such heating and excellent control of spatial and temporal profiles of temperatures achieved. The cooling part of the thermal history of a laser-heated specimen can be manipulated by an external cooling arrangement. A laser beam-based heating system, wherein the laser beam can heat both the surfaces of a wafer sectioned out of a fuel pin, can help us to study what happens to the fuel and its contents during such heating. Such a system is being developed elsewhere in the world. About this system, called POLARIS, a single tangential reference [2] exists. No other information is available in the open literature. Therefore, our approach had to start with pilot studies and graduate towards a full version for hot cell use. To design such a system for irradiated fuels, pilot studies on unirradiated fuels were carried out. This paper describes the experiments and the results of metallographic examination and X-ray diffraction studies of the laser-treated specimens.

2. Experimental procedure

Pellets of UO_2 were prepared metallographically and subjected to laser pulses of 700 ms duration with an average power of 1 kW. The pulse shape in time was rectangular. All studies were done with single pulses. The laser beam was focussed to a diameter of 8 mm on the specimen surface. These laser pulses were programmed pulses from a 1 kW CW Nd:YAG laser. The power density incident on the specimens was about 2000 W/cm^2. The specimens were enclosed in a stainless steel chamber with a glass window which was transparent to the laser radiation at 1064 nm. The chamber had a provision to pass an inert gas or steam through it, enabling experiments to be conducted in air, steam or inert atmosphere. Figure 1 illustrates the experimental set-up schematically.

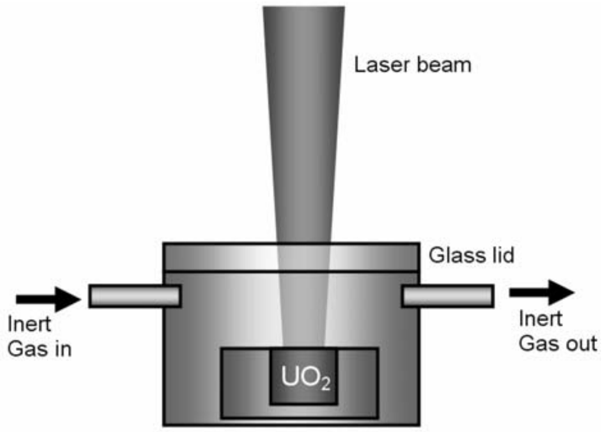

Figure 1. The experimental set-up used.

Using the analytical solution for heating by a continuous Gaussian laser source [3] and the above experimental parameters, the temperature attained by the central point was estimated to be 11,922 K. The whole area under the beam would then have reached the melting point, i.e., 3120 K. A heating rate of about 8000 K/s was estimated in the molten zone. For simulating the reactor accidents mentioned above, these heating rates were adequate. In the first phase of the experiments, one accident situation was simulated: a UO_2 pellet exposed to a single laser pulse under air, simulating a severe accident condition wherein molten fuel met air after breaching the containment. The objective was to study the behaviour of the fuel under the transient heating induced by the laser pulses.

After laser heating, an instant adhesive was poured over the invariably cracked specimen to retain the relative positions of the fragments. This made the pellet stable enough to be handled for the subsequent metallographic preparation. The pellets were vertically sectioned into two equal parts, and then prepared metallographically, to enable examination of the cross-section of the laser-treated region. During the laser heating studies conducted in air, it was observed that the glass window was coated with some material resulting possibly from evaporation and/or a chemical reaction between the evaporated material and air. These glass windows were removed after every experiment and were analysed by X-ray diffractometry to study the phases formed.

3. Results and discussion

UO_2 pellets heated in air were found to undergo melting and cracking. The laser-treated zone was characterized by metallography. Figure 2 shows the original microstructure of the UO_2 specimens. Figure 3 shows the appearance of the specimen after laser treatment, showing severe cracking. Figure 4 shows the microstructure of the cross-section of the laser-treated specimen. The presence of columnar grains in the laser-treated zone confirm the melting and solidification undergone by the specimen. Figure 5 shows the agglomeration of porosity near the interface between the molten and unmolten regions. The molten region was found to have very large grains compared to the original material. The original grains were 10 μm in

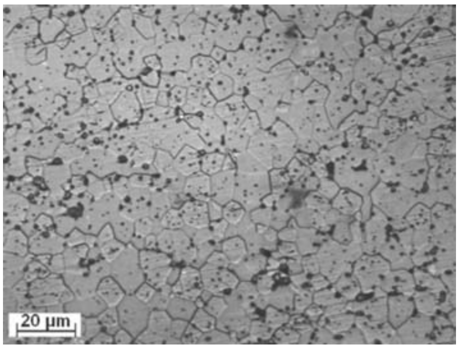

Figure 2. The original microstructure of UO_2 specimen.

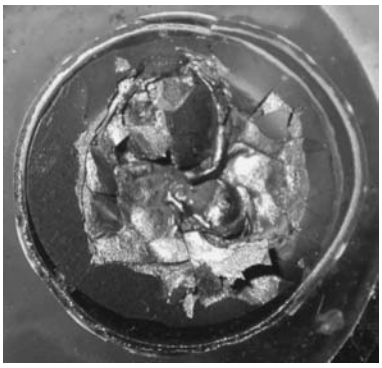

Figure 3. Appearance of the specimen after laser treatment in air.

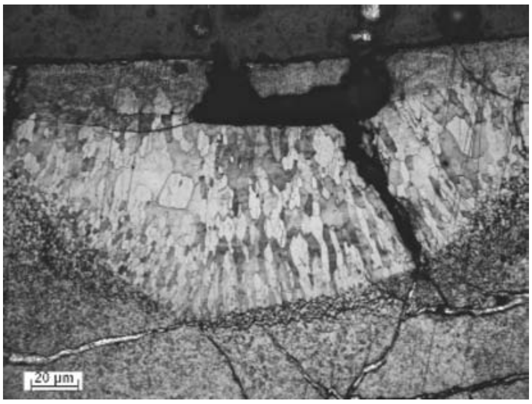

Figure 4. The cross-section of laser-melted zone showing columnar grains.

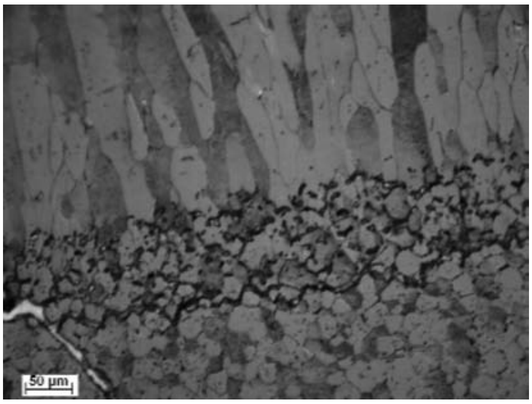

Figure 5. Agglomeration of porosity near the interface between columnar grains and the unaffected region.

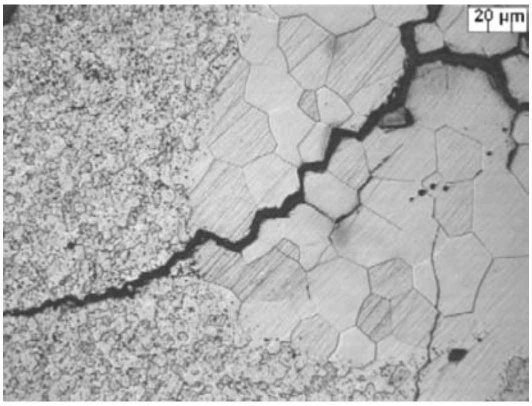

Figure 6. Abnormal grain growth observed.

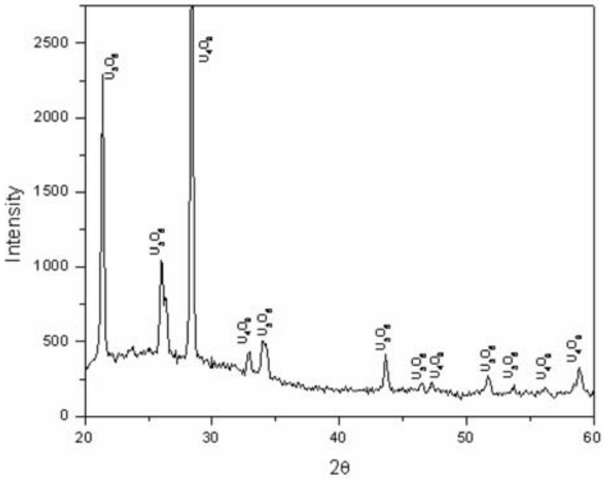

Figure 7. XRD of UO_2 pellet laser-treated in air showing U_4O_9 and U_3O_8.

size on the average, and these grew into sizes ranging up to 300 μm during the laser heating. Also the porosity which was present throughout the grains in the pellet vanished from the larger grains, and was present only at the grain boundaries. These features of the laser-treated specimen can be seen in figure 6. An X-ray diffraction pattern recorded from the laser treated pellet surface is shown in figure 7.

X-ray diffraction studies revealed that the higher oxides of uranium, namely U_3O_8 and U_4O_9, constituted most of the surface of the pellet. These are known to form at the temperatures reached by the pellet during laser heating [4]. The considerable plume produced during the laser heating led to deposition of a film on the glass window of the chamber. The film was found by XRD to consist of mainly U_3O_7.

The presence of higher oxides in the surface and in the plume was expected from the temperature (believed to have gone above 3000°C) and the ambient [4,5].

Extraordinary grain growth, associated with a large increase in the diffusion coefficients of all species with x in UO_{2+x}, was reported in [6]. This fact was recently exploited in a technique to produce extra large grains and even single crystals of UO_2 by sintering in air [7].

These pilot studies were successful in demonstrating the feasibility of simulating fuel melting and allied phenomena using laser pulse heating. Based on these studies, further experiments on fuel with or without cladding in air and steam are planned. To graduate to a complete simulation set-up, the following are needed: (a) A means of modifying the incident laser beam intensity distribution from the usual Gaussian to the shape of a typical temperature distribution in the nuclear fuel will be needed. This can be a suitably designed lens system. As the specimen is a thin wafer, the temperature distribution will closely resemble the power distribution. The steady state temperature distribution in a nuclear fuel pellet is like a cosine curve. Using two laser beams, one for simulating the steady state temperature distribution, and the other for superimposing a thermal transient, may be necessary. (b) A high-speed optical pyrometer which can measure the temperature distribution in the fuel specimen as a function of time during the laser pulse will be needed. The specimen chamber windows should be transparent to both the laser radiation and the infrared radiation used for pyrometry. (c) A high frame rate camera to record the specimen morphology during the laser simulation will be necessary to enable studies on fuel fragmentation kinetics. (d) For studies on transient fission gas release, a gas extraction system connected to a mass spectrometer would be ideal. Such a complete system will function in a hot cell as highly radioactive irradiated fuel specimens need to be studied. An effort is under way to fabricate such a system at Post Irradiation Examination Division, BARC, Mumbai, India.

4. Conclusions

The feasibility of using laser beams for simulating reactor transients was demonstrated. During the simulation of a postulated severe accident scenario, of molten UO_2 fuel coming into contact with air, the formation of higher oxides of uranium and extraordinary grain growth assisted by the high diffusion rates were observed.

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