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Behaviour of irradiated PHWR fuel pins during high temperature heating

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ABSTRACT

Fuel pins removed from an irradiated pressurised heavy water reactor (PHWR) fuel bundle discharged after an extended burn up of 15,000 MWd/tU have been subjected to isothermal heating tests in temperature range 700–1300 °C inside hot-cells. The heating of the fuel pins was carried out using a specially designed remotely operable furnace, which allowed localized heating of about 100 mm length of the fuel pin at one end under flowing argon gas or in air atmosphere. Post-test examination performed in the hot-cells included visual examination, leak testing, dimension measurement and optical and scanning electron microscopy. Fuel pins having internal pressure of 2.1–2.7 MPa due to fission gas release underwent ballooning and micro cracking during heating for 10 min at 800 °C and 900 °C but not at 700 °C. Fuel pin heated at 1300 °C showed complete disruption of cladding in heating zone, due to the embrittlement of the cladding. The examination of fuel from the pin tested at 1300 °C showed presence of large number of bubbles; both intragranular as well as intergranular bubbles. Details of the experiments and the results are presented in this paper.

1. Introduction

Water reactor fuel pins consist of a stack of sintered ceramic UO_2 pellets contained in a Zircaloy cladding tube which is hermetically sealed at both ends. The void space in the fuel pin is filled with helium at atmospheric pressure. The internal pressure inside the fuel pin increases with burnup due to release of fission gases xenon (Xe) and krypton (Kr) from the fuel. The behaviour of fuel pin during a loss of coolant accident (LOCA) is governed by temperature of the cladding, the internal gas pressure and the state of the cladding such as extent of oxidation, mechanical strength etc. The issue becomes more relevant for the fuel with high discharge burn up due to the accumulation of gaseous and volatile fission products. During the course of LOCA, one of the major safety requirements is the feasibility of cooling down a distorted fuel assembly. This requirement puts some limits on the extent of allowable deformation like ballooning of individual fuel elements [1].

Ballooning behaviour is usually studied in out-of-pile tests using internally pressurised cladding tubes or using simulator rods [1]. Ballooning tests on actual irradiated fuel pins are more meaningful but difficult to perform because it is required to be performed inside hot-cells. In the present work, actual fuel pins irradiated to an extended burn up have been used for the experiment.

The specific aspects investigated in these experiments include clad deformation and ballooning-failure as function of cladding temperature, internal gas pressure and burn up of the fuel pin. Chemical interaction between high burn up fuel and cladding at temperatures relevant to postulated accident condition has also been examined.

2. Experimental

2.1. In-cell heating facility

A remotely operable electrical furnace capable of heating up to 1350 °C under air or argon atmosphere (Fig. 1(a)) has been used for heating the fuel pins inside the hot-cells. The furnace is essentially of closed cylindrical type with an overall length of 750 mm and capable of providing a constant temperature zone of 100 mm. An Inconel tube with 125 mm diameter and 3 mm wall thickness is the heating chamber of the furnace. The fuel pin is charged into a ceramic tube of 20 mm OD and 2 mm wall thickness and passes concentrically through a reflector–insulator assembly (Fig. 1(b)). The whole assembly is loaded into the heating chamber in such a way that only 100 mm of the fuel pin is in the constant temperature zone. Heat resistant pads are provided on either sides of the constant temperature zone, so that temperatures on the remaining length of the sample are kept as low as possible. The temperature control of the furnace was achieved by proportional integral derivative (PID) controllers and platinum–rhodium (R-type) thermocouple with compensation leads was used for temperature measurements. The fuel pins for ballooning experiments were heated at temperatures from 700 to 900 °C under argon atmosphere and held for 10 min at those temperatures, followed by

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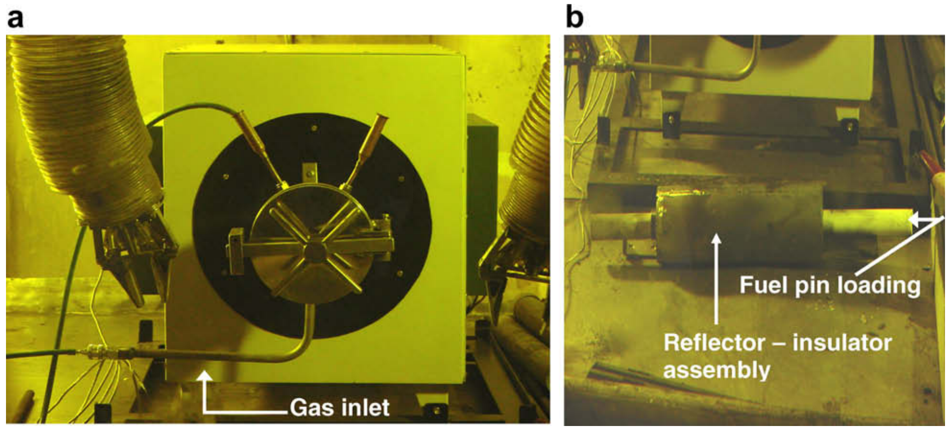

Fig. 1. (a) Remotely operable electrical furnace capable of heating fuel pins up to 1350 °C under air or argon atmosphere and (b) reflector–insulator assembly for charging the fuel pin in the furnace, as viewed through the viewing window of the hot-cell.

furnace cooling to room temperature. Severe test fuel pin was heated at 1300 °C in air and held at that temperature for 3 h before furnace cooling to room temperatures. Heating rates used in all heating experiments were 12 °C/min up to the temperature of 800 °C and 8 °C/min, thereafter, up to 1300 °C.

It is established from initial calibration runs on unirradiated fuel pins that corresponding to a temperature of 1150 °C at the constant temperature zone, temperatures measured on the pin at 100 mm, 200 mm and 250 mm away from the constant temperature zone were 790 °C, 474 °C and 300 °C, respectively.

2.2. Fuel pins used for the in-cell experiments

Fuel pins used in the experiment were taken from outer ring of a PHWR fuel bundle which had been discharged from Kakrapar Atomic Power Station, after achieving an average fuel burn up of 15,000 MWd/tU. A PHWR fuel bundle consists of 19 fuel pins, each of 493 mm in length and 15.2 mm in diameter, arranged in a circular array, as shown in Fig. 2. There are 12 fuel pins in the outer ring, six fuel pins in the middle ring and one pin at the centre. The linear power rating is highest in the outer pins and the lowest in the central pin. Detailed post irradiation examination (PIE) of the fuel pins of this fuel bundle was carried out earlier and findings are reported elsewhere [2]. During PIE, five fuel pins of the outer ring in the fuel bundle were punctured to measure the internal pressure in the pin due to fission gas release which was in the range of 2.1–2.7 MPa [3]. The remaining pins were kept aside for in-cell heating experiments. Some of these fuel pins were utilized in the present experiment.

Fig. 2. A typical 19-element PHWR fuel bundle.

2.3. Leak testing

Leak tests were carried out inside the hot-cells, by using liquid nitrogen–alcohol method, on all fuel pins before and after the heating experiment.

2.3.1. Optical and scanning electron microscopy (SEM)

Microstructural examination of fuel and cladding and fractography of the cladding of the ruptured pin were carried out using remotised shielded optical microscope and a shielded SEM. The samples were prepared remotely in the metallographic cell using remote controlled grinding and polishing machines. Longitudinal section from a broken piece of the fragmented cladding was mounted in cold setting resin. The broken piece of the fuel pin after the severe test was impregnated with cold setting resin before extracting a cross-section by using slow speed cut-off wheel. The samples were ground on 180 grit silicon carbide (SiC) paper till the section was completely exposed and then progressively ground on 240, 320 and 600 grit SiC papers. The ground samples were successively polished using diamond slurry with particle size of 3 μm and 1 μm. The fracture surface of the cladding was cleaned ultrasonically in acetone and then sputter coated with copper, before SEM examination.

3. Results and discussion

3.1. Ballooning experiments

Fig. 3(a)–(c) shows, respectively, the photographs of the UO_2 fuel pins from the outer ring of the fuel bundle, heated at 700 °C, 800 °C and 900 °C for 10 min under argon cover. No perceivable deformation was observed on the fuel pin heated at 700 °C. However, at 800 °C and 900 °C well defined ballooning of the cladding was noticed. The average diametral deformation was in the range of 35–45%. The ballooning had occurred over a length of about 50 mm, between the end cap and the bearing pad region of the pin which was in the constant temperature zone in the furnace. It was also noted that heating had not caused any extra oxidation to the surface of the fuel pins. However, large numbers of fine crack- like features were noticed on the ballooned regions of both fuel pins. Leak

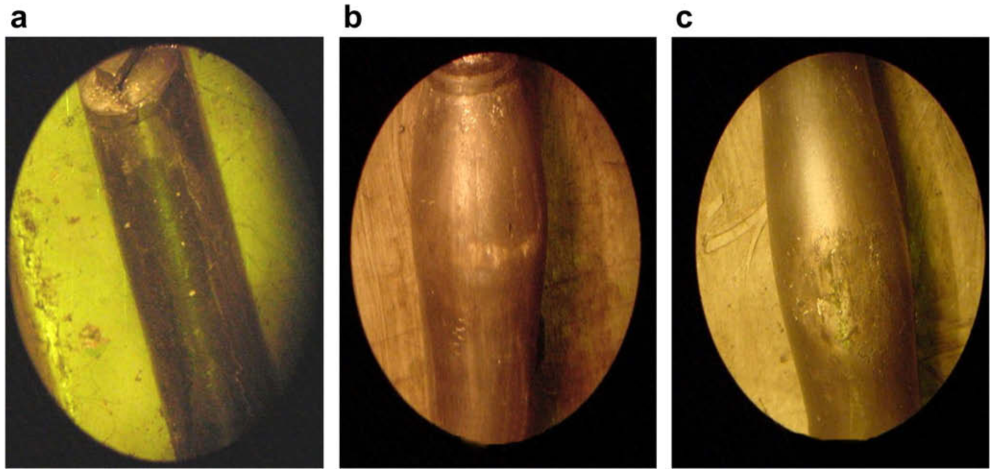

Fig. 3. Fuel pins from the outer ring of the fuel bundle heated (a) at 700 °C, (b) at 800 °C and (c) at 900 °C for 10 min under argon cover.

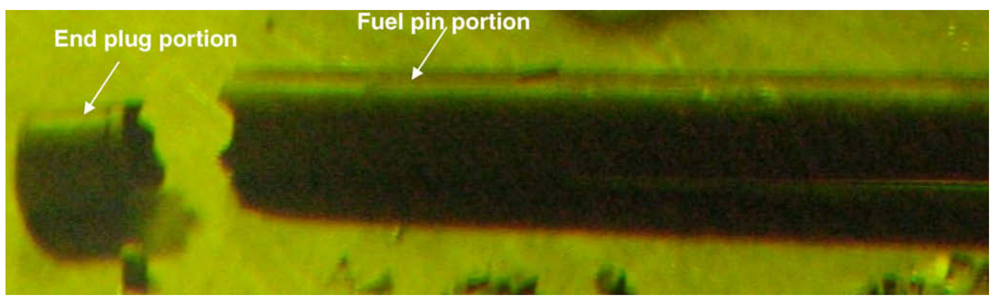

Fig. 4. Fragmented fuel pin after severe test at 1300 °C in air.

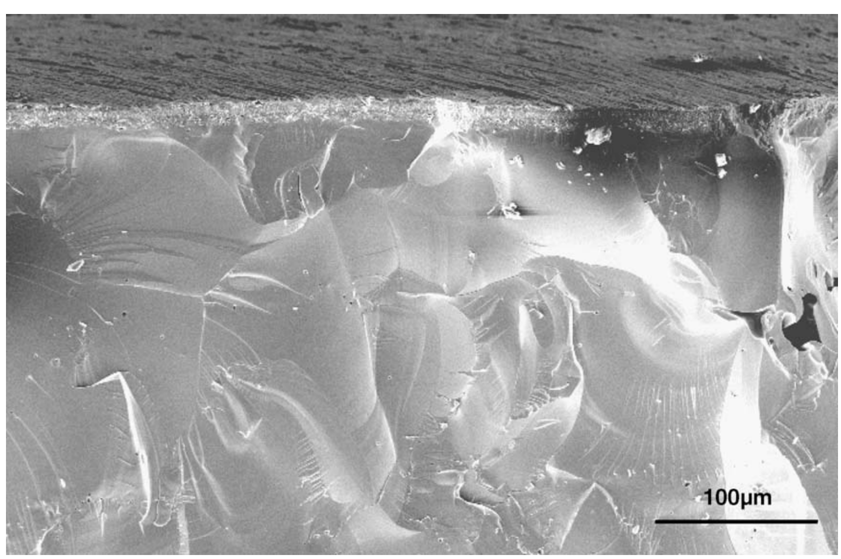

Fig. 5. Appearance of the fracture surface of the fuel pin subjected to severe test at 1300 °C in air.

testing showed profuse bubbling from the middle of the ballooned portion of the pins heated at 800 °C and 900 °C. The fuel pin heated at 700 °C, however, was intact and did not show any leak.

Internal cold gas pressure due to the release of fission gases was of the order of 2.1–2.7 MPa. Ignoring the internal void volume changes of the fuel pins due to thermal expansion of the fuel and the cladding, the internal gas pressure at 800 °C and 900 °C will be in the range of 7.5–9.7 MPa and 8.2–10.6 MPa, respectively. Assuming that the initial internal diameter of 14.4 mm and the wall thickness of 0.4 mm of the cladding remain unchanged during heating, these internal gas pressures could cause hoop stresses in the range of 135–175 MPa and 148–190 MPa, at 800 °C and 900 °C, respectively. These stresses are much above the ultimate tensile strength (UTS) of Zircaloy-2 at these temperatures [4].

During heating, ballooning of the cladding occurred due to an increase in the internal pressure of the fuel pin and also due to the decrease in cladding strength [4]. Ballooning might have progressed till the cladding failed by way of cracks and the entire gas pressure is released. This is confirmed by the presence of leaky cracks on the ballooned region of the fuel pins.

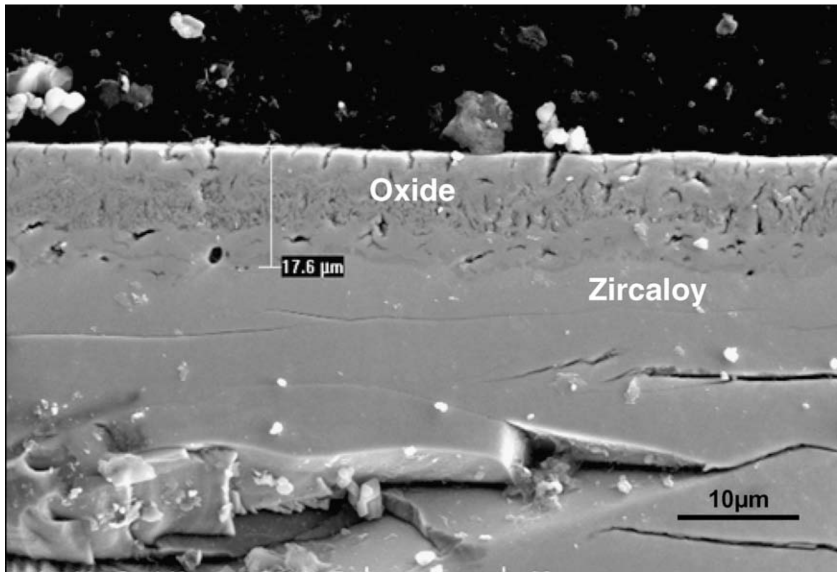

Fig. 6. Section of the cladding of the fuel pin subjected to severe test at 1300 °C in air, showing oxide layer on the outer surface of the tube and cracks in the cladding.

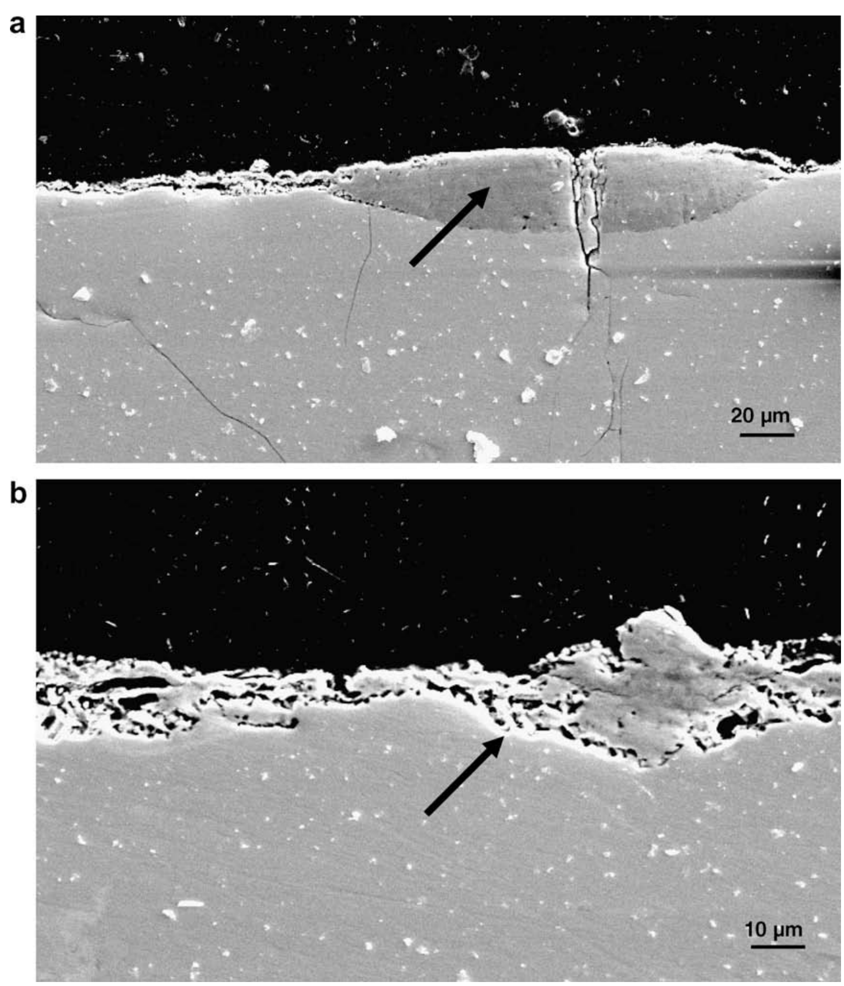

Fig. 7. Microstructure of the clad-fuel interface of ruptured cladding showing pellet-clad interaction products of (a) nodular type and (b) porous type.

3.2. Severe test experiment

It was observed that approximately 165 mm of the fuel, 35 mm away from the end cap, in the hot zone of the furnace was in a shattered condition, when the furnace was opened after cooling to room temperature. The remnants of the fuel pin are shown in Fig. 4. The fragments of the cladding had become so fragile that handling them with master slave manipulators was very difficult. The inner surface of the fragmented piece of the cladding showed white oxide layer and the outer surface was dark in appearance.

The fracture surface of the ruptured cladding showed brittle appearance, as shown in Fig. 5, similar to those observed by Suparna et al. [5] in cladding containing high oxygen content. Microstructure from the section of the cladding showing oxide layer on the outer surface of the ruptured cladding is shown in Fig. 6. The oxide layer appeared to be porous, containing a large number of radial and axial micro cracks. Microstructure of the fuel clad interface from the inner surface of the clad is shown in Fig. 7(a) and (b). Two types of interaction products namely (i) localized nodular type, as shown by arrow in Fig. 7(a) and (ii) interaction layer of porous and non uniform thickness, as shown by arrow in Fig. 7(b), could be noticed.

The cross-section of the fuel pin subjected to severe test is shown in Fig. 8. Microstructure of the fuel in the pellet centre is shown in Fig. 9(a). Microstructure of the fuel from an identical location of an unheated companion fuel pin is shown in Fig. 9(b) for comparison. There is a distinct difference between the microstructure of the unheated fuel pin and the fuel pin heated to 1300 °C for 3 h. The fuel microstructure before the heating test showed mostly intergranular gas bubbles decorating the fuel grain boundaries. The microstructure of the fuel after subjecting to heating showed an increase in the number density of bubbles and most of the bubbles are of intragranular type. Some degree of coalescence of the bubbles is also observed. These observations indicate growth of fission gas bubbles within the grain due to gas atom migration at this temperature.

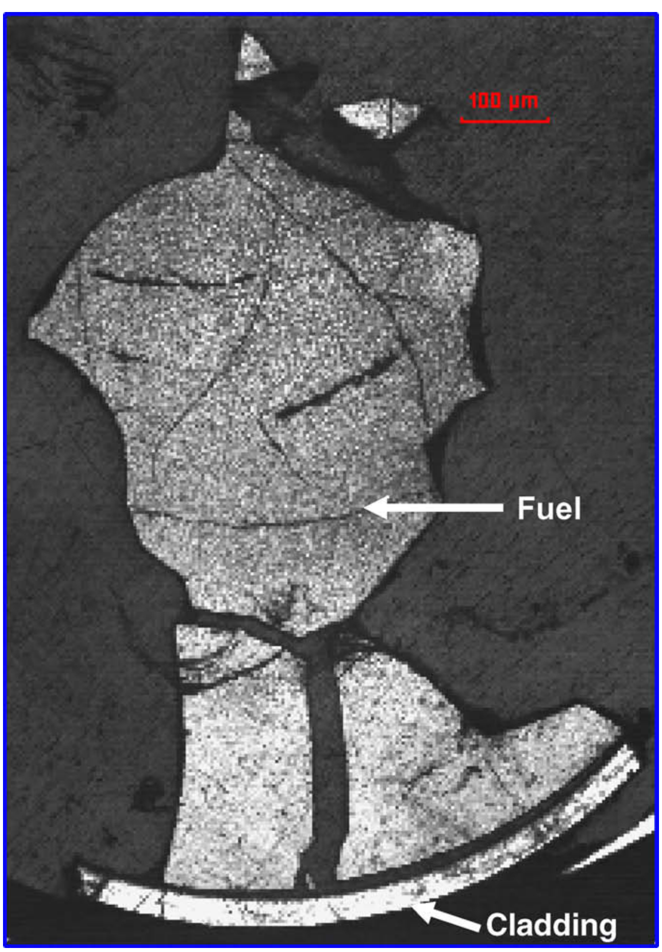

Fig. 8. Part of the cross-section of the fuel pin fragmented during severe test.

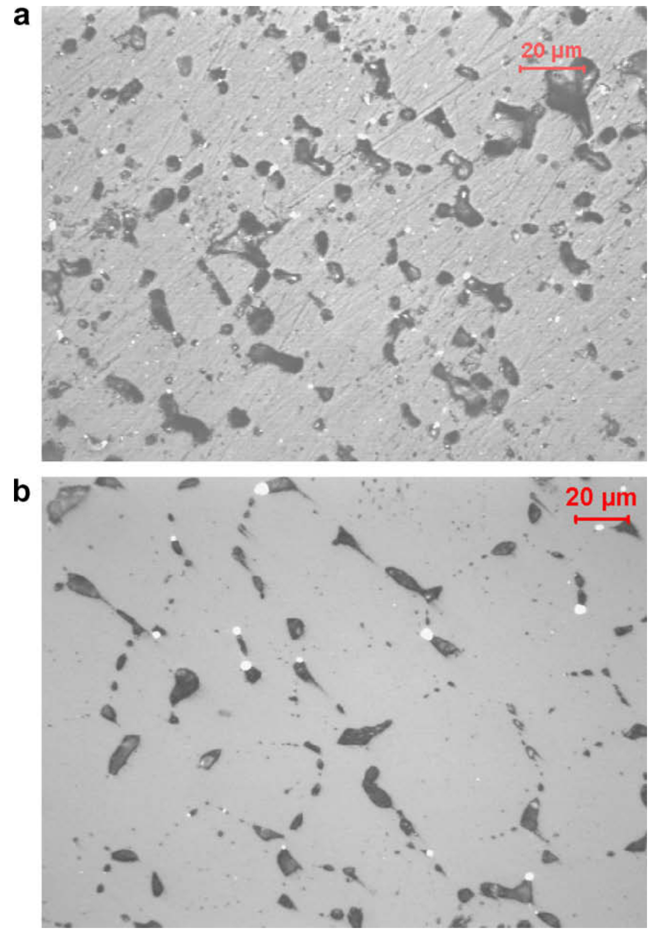

Fig. 9. Microstructures of the fuel (a) in the pellet centre from the severe-tested pin and (b) in the pellet centre from an untested companion pin.

4. Conclusions

Based on the isothermal heating experiments carried out in the temperature range of 700–1300 °C on irradiated fuel pins discharged after an average burn up of 15,000 MWd/tU and having an internal fission gas pressure of 2.1–2.7 MPa, the following conclusions are drawn:

I. No ballooning of the cladding occurred on heating at 700 °C for 10 min. However, fuel pins heated at 800 °C and 900 °C for 10 min showed well defined ballooning.
II. Ballooned fuel pins failed due to the formation of micro cracks at the ballooned region.
III. The maximum diametral deformation in the ballooned portion of the pin was in the range of 35–45%.
IV. Fuel pin heated at 1300 °C for 3 h in air failed by complete disruption of cladding in heating zone. Failure was due to oxygen embrittlement of Zircaloy cladding.
V. Microstructure of the fuel heated at 1300 °C showed an increase in number density of fission gas bubbles and most of the bubbles were of intragranular type.

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