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Measurement of fission gas release, internal pressure and cladding creep rate in the fuel pins of PHWR bundle of normal discharge burnup

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ABSTRACT

Fuel pins of a Pressurised Heavy Water Reactor (PHWR) fuel bundle discharged from Narora Atomic Power Station unit #1 after attaining a fuel burnup of 7528 MWd/tU have been subjected to two types of studies, namely (i) puncture test to estimate extent of fission gas release and internal pressure in the fuel pin and (ii) localized heating of the irradiated fuel pin to measure the creep rate of the cladding in temperature range 800 °C–900 °C. The fission gas release in the fuel pins from the outer ring of the bundle was found to be about 8%. However, only marginal release was found in fuel pins from the middle ring and the central fuel pin. The internal gas pressure in the outer fuel pin was measured to be 0.55 ± 0.05 MPa at room temperature. In-cell isothermal heating of a small portion of the outer fuel pins was carried out at 800 °C, 850 °C and 900 °C for 10 min and the increase in diameter of the fuel pin was measured after heat treatment. Creep rates of the cladding obtained from the measurement of the diameter change of the cladding due to heating at 800 °C, 850 °C and 900 °C were found respectively to be $2.4 \times 10^{-5}\ s^{-1}$, $24.6 \times 10^{-5}\ s^{-1}$ and $45.6 \times 10^{-5}\ s^{-1}$.

1. Introduction

Fission gas release is one of the important performance parameters for Zircaloy clad UO_2 fuel pins used in water cooled nuclear reactors [1–4]. Release of fission gases (Xe and Kr) has deleterious effect on the fuel–clad gap conductance because their thermal conductivity is much lower than the filler gas helium. Deterioration of fuel–clad gap conductance leads to increase in the fuel centre temperature causing more gas release. Fission gas release is also important with regard to behaviour of fuel pins during postulated loss of coolant accident (LOCA) conditions. The internal pressure in the fuel pin due to fission gas release is responsible for creep deformation of the cladding at high temperature during LOCA. The measurement of fission gas release and its possible effect on the cladding deformation at high temperature, anticipated during a postulated accident condition, is therefore of paramount importance. Such data is required for the development of models and for the validation of fuel performance codes. We have earlier measured fission gas release in irradiated fuel pins of Tarapur Atomic Power Stations and in experimental mixed oxide (MOX) fuel pins of boiling water reactor (BWR) design, irradiated in pressurized water loop of CIRUS [5,6].

The aims of the present study are (i) to measure the fission gas release and internal gas pressure in the fuel pins of PHWR fuel bundles discharged from the reactor after its design burnup is achieved and (ii) to generate data on high temperature thermal creep of irradiated cladding by heating the fuel pins inside the hot-cells. The design and operating conditions of PHWR fuel pin differ from light water reactor (LWR) fuels [7], the former being short in length having no separate plenum. The PHWR fuel pins operate at significantly higher linear heat rating compared to LWR fuel pins. Higher heat rating in PHWR fuel pins causes significant fuel restructuring which promotes fission gas release. In the present study, puncture tests have been carried out on fuel pins of a PHWR bundle which was irradiated up to its design life for the measurement of fission gas release and pin internal pressure. A few intact fuel pins from the bundle have been subjected to in-cell heating at high temperatures in an inert atmosphere to study the cladding deformation as a function of time and temperature of heating. This paper presents the details and results of the investigation.

2. Design and irradiation of fuel bundles

The fuel bundles for 220 MWe Indian PHWRs are short cylindrical assemblies. There are 12 fuel bundles in each coolant channel assembly of the reactor. Each fuel bundle used in the 220 MWe reactors contains nineteen fuel pins. A typical fuel pin consists of a stack of sintered natural uranium dioxide fuel pellets, cylindrical in form, placed inside a Zircaloy fuel tube and sealed at both ends by end plugs. Nineteen such fuel pins arranged in concentric rings are assembled by welding them to an end plate on each side to form a bundle. The bundle consists of a central pin surrounded

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by six pins in the middle ring and 12 pins in the outer ring. The bundle length is 495 mm and the weight is 16 kg. The different components of the fuel bundle are shown in Fig. 1.

On-power bi-directional fuelling is carried out with the aid of two fuelling machines, one each at either end of the coolant channel. The fuel bundles are cooled by heavy water at a pressure of about 10 MPa.

In the present work, fuel pins of a PHWR fuel bundle No 54505 discharged from Narora Atomic Power Station unit #1 (NAPS-1) were studied. The bundle power during irradiation was in the range of 384–389 kW. The fuel bundle was discharged from the reactor after achieving an average burnup of 7528 MWd/tU which is close to the normal discharge burnup of PHWR fuel bundle in a 220 MWe reactor. The peak burnup achieved in the outer fuel pins in this bundle was 8205 MWd/tU. The important design data of fuel, cladding and fuel pins of PHWR fuel bundle are given in Table 1.

Table 1
Data on fuel, cladding and fuel pin of bundle No. 54505.

Fuel pellet data		
1	Chemical composition	Natural UO_2
2	*O/U* ratio	2.005
3	Pellet inner diameter (cm)	0.0
4	Pellet outer diameter (cm)	1.432
5	*L/D* ratio	1.2
6	Pellet end geometry	Spherical dish at one end
7	Fuel density, %TD	96.3 (10.55 g/cm^3)
8	Grain diameter (μm)	10
Cladding data		
9	Cladding material	Zircaloy-2
10	Metallurgical condition of the clad	Stress relieved
11	Cladding ID (cm)	1.440
12	Cladding OD (cm)	1.520
13	Clad thickness (cm)	0.04
14	Cladding surface roughness (μm)	0.8
Fuel pin data		
15	Active stack length (cm)	48.10
16	Axial gap in fuel pin (cm)	0.17
17	Fuel–clad diametrical gap (μm)	80
18	Total void volume (cm^3)	3
19	Fill gas composition	He
20	Fill gas pressure, absolute (kg/cm^2)	1.2

3. Experimental

3.1. Bundle dismantling and leak testing

After preliminary survey of the fuel bundle for its overall integrity, the bundle was dismantled using a 150 W pulsed Nd–YAG laser, installed in the hot-cells and controlled by a CNC based system. After dismantling the bundle, all the fuel pins were subjected to detailed visual examination using a wall-mounted periscope. Individual fuel pins were leak tested to check their integrity before taking them for puncturing to measure fission gas release. The leak testing of the fuel pins was carried out using liquid nitrogen–alcohol test inside the hot-cells.

3.2. Fission gas release measurement and analysis

3.2.1. Fuel pin puncturing and measurement of gas volume and pressure

Fission gas release measurement was carried out using a puncturing set up developed for PHWR fuel pins. The details of this set up and method of measurement are given in Ref. [8]. The set up essentially consists of a puncture chamber fixed inside the hot-cell which is connected to the gas collection and measuring part (located in the hot-cell operating area) by means of stainless steel tubes. The whole system can be evacuated to a vacuum level of less than 0.1 mbar by a mechanical pump.

Estimation of parameters such as system volume, system pressure, void volume of the fuel pin and the pressure and volume of released gases was carried out by connecting calibration flasks to the system and by applying gas laws. Capacitance diaphragm gauges were used for pressure measurements.

The percentage of fission gas released (FGR) was calculated using the following formula:

$$\%\ \mathrm{FGR} = \left(\frac{V_R}{V_G}\right) \times 100, \tag{1}$$

where V_R is the volume of the fission gas released as estimated by the experiment and V_G is volume of fission gas generated in the fuel. The quantity of fission gas generated in the fuel was estimated by the equation:

$$V_G = 30 * W * B, \tag{2}$$

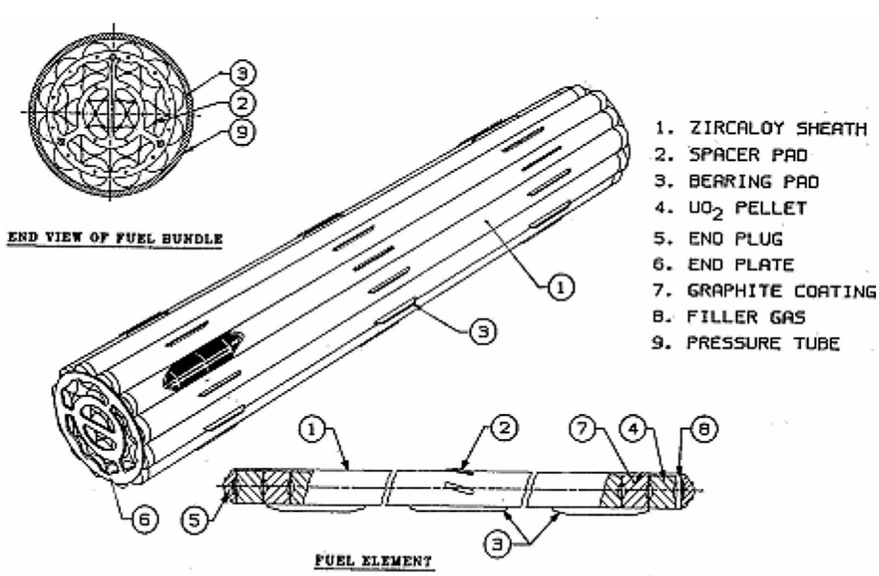

Fig. 1. Design details of PHWR fuel.

where W is the weight of the fuel in kg, B is fuel element burnup in GWd/tU, and V_G is Volume of fission gas generated in the fuel in cubic centimeter (cm^3) at STP.

3.2.2. Chemical analysis of fission gases

Chemical composition of the released gases was carried out using a dual column gas chromatograph with helium as the carrier gas. Thermal conductivity detector was used for the detection of individual gases.

3.3. In-cell heating of fuel pins

A closed cylindrical type horizontal electrical furnace developed for heating a small part of the irradiated PHWR fuel pin inside the hot-cells was used. The details of this furnace are given elsewhere [9]. The overall length of the furnace was 750 mm which allowed a full length fuel pin to be inserted in the furnace. The furnace was designed to provide heating of 100 mm length of fuel pin towards one end under argon atmosphere. An Inconel tube with 125 mm diameter and 3 mm wall thickness is used as the heating chamber of the furnace. The fuel pin is charged into a ceramic tube of 20 mm outer diameter and 2 mm wall thickness and passes concentrically through a reflector–insulator assembly. 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 side 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. A platinum–rhodium (R-type) thermocouple with compensation leads was used for temperature measurements. The temperature of the portion of the fuel pin in the heating zone was raised slowly at a heating rate of 8–12 °C/min to the desired temperature (800 °C, 850 °C and 900 °C) and held there for specified period to allow cladding to undergo thermal creep under the influence of internal pressure in the pin. The fuel pin was furnace cooled after the heating experiment.

3.3.1. Leak testing of heated pins

After the in-cell heating experiment the fuel pins were subjected to leak testing to check whether the fuel pin cladding had failed due to deformation during heating. Leak testing was carried out using liquid nitrogen–alcohol method inside the hot-cells.

3.3.2. Diameter measurement along the length of heated pins

The outer diameter of the fuel pin was measured at close distances along the length of the heated fuel pins, by using a remotely operated stage fitted with a micrometer.

4. Modeling calculations

4.1. Fuel centre temperature, fission gas release and internal pressure

The fuel centre temperature and fission gas release behaviour were analyzed using computer code PROFESS, which is an axisymmetric one dimensional code applicable to Zircaloy clad UO_2 fuel pins of water cooled reactors [10,11]. In this code fuel pellet cross section is divided into 50 concentric rings of equal width for calculation. The code considers neutron flux depression, in-pile fuel densification, solid and gaseous fission product swelling, fission gas release, fuel restructuring, fuel relocation, rim effect, and in-pile cladding creep for analyzing the behaviour of fuel pins during irradiation. A modified Ross and Stout gap conductance model is used in the code which takes into account the effect of gap size, gap–gas composition, gas pressure and pellet eccentricity. The code contains a temperature dependent fuel thermal conductivity model with factors incorporated in the model to account for the effects of fuel porosity and fuel burnup. The fission gas release in the fuel is calculated using a thermal diffusion model. The basic diffusion equation considered in the model is as follows:

$$\frac{\partial c}{\partial t} = D_{gas}\nabla^2 - gc + bm + \beta, \qquad (3)$$

where D_{gas} is diffusion coefficient of gas in fuel matrix, c is concentration of gas in the grain, g is probability of capture of gas by defects, b is resolution probability, m is concentration of gas in traps and β is rate of generation of fission gas atoms. The correlations for thermal diffusivity of xenon gas atoms in solid UO_2, used in the fission gas release model of the code, are taken from Ref. [12]. The details of the model for calculating the gas atom diffusion inside the grain and the number density of gas atoms at the grain boundary are described in Ref. [10].

For calculating the fuel temperature and fission gas release in the fuel pins of the PHWR fuel bundle, the design data of the fuel pellet, cladding and fuel pin along with irradiation data of the fuel pins were used as input. Though the fuel bundle power during reactor operation was in the range of 384–389 kW, the operation was assumed to be at a constant power of 386.5 kW. Taking into account the radial power factors of 1.09, 0.84 and 0.77, the linear power rating of the outer, middle and central fuel pin are estimated to be 460 W/cm, 355 W/cm and 326 W/cm, respectively. Fuel pin was considered as a single segment for calculation.

4.2. High temperature creep rate of cladding

The steady state creep rate of Zircaloy cladding in α phase region was estimated using correlations of Clay and Redding [13,14], and that of Rose and Hindle [15]. As the correlation of Clay and Redding was for uniaxial creep rate, the constant in the correlation was modified using method described in Ref. [16] for calculating steady state creep in circumferential direction in the tube under biaxial stress condition. Correlation for α phase region was used up to 850 °C following Ref. [14]. Creep rate at 900 °C was calculated using a correlation for $\alpha + \beta$ phase region from Ref. [17]. As the cladding hoop stress expected in the temperature range of 800 °C–900 °C in a fuel pin with internal pressure of 0.55 MPa is around 36 MPa, the calculations for the creep rates were made assuming a hoop stress of 36 MPa at the inner surface of the cladding tube.

5. Results

5.1. Visual examination and leak testing

The visual examination and leak testing of fuel pins of the bundle indicated that all the fuel pins were in good condition and there was no leak in any of them.

5.2. Fission gas release in the fuel pins

Fission gas release and internal pressure were measured in two outer fuel pins, two middle fuel pins and the central pin of the fuel bundle. The results of fission gas measurement on these fuel pins are shown in Table 2. It is observed that percentage of fission gas release in the central fuel pin and in the intermediate fuel pins is marginal. However, outer fuel pins showed an appreciable fission gas release of 8%. The measured internal gas pressure inside the central fuel pin and the middle fuel pins was in the range 0.12–0.15 MPa at room temperature. The internal pressure in the two outer fuel pins was found to be 0.51 MPa and 0.59 MPa.

Table 2
Fission gas release in fuel pins of fuel bundle No. 54505.

Fuel bundle no.	Average bundle burnup (MWd/tU)	Fuel pin location in the bundle	Internal pin pressure at room temperature (MPa)	Volume of fission gas at STP (cm^3)	Fission gas release (%)
54505	7528	Outer ring	0.59	15.6	8.8
		Outer ring	0.51	14	7.8
		Middle ring	0.15	0.19	0.1
		Middle ring	0.14	0.24	0.2
		Central	0.12	0.15	0.1

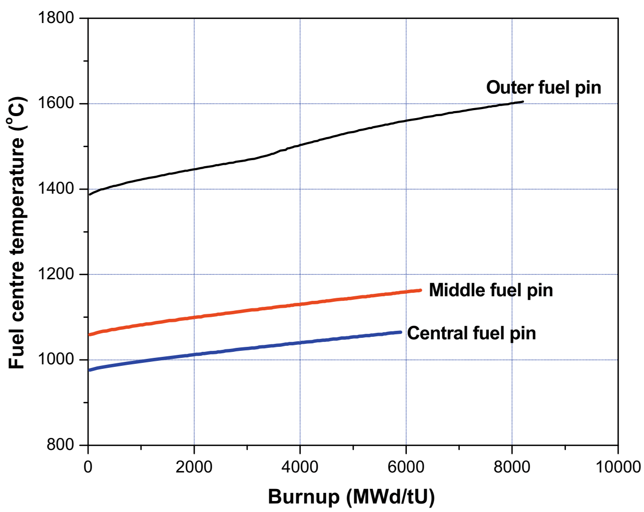

Fig. 2. Predicted fuel centre temperatures in the three types of fuel pins during irradiation.

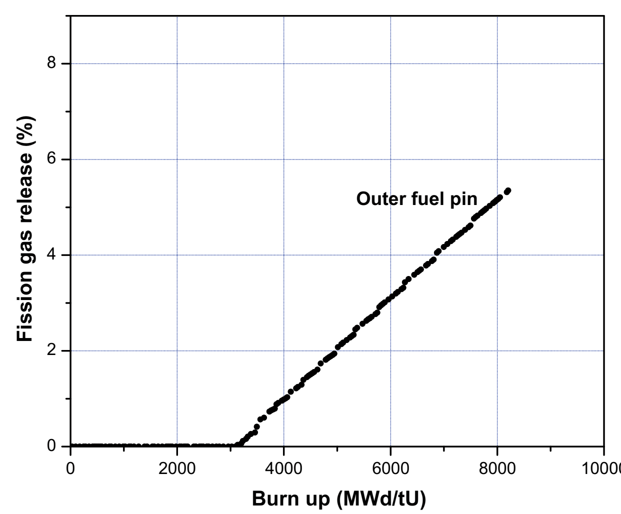

Fig. 3. Predicted fission gas release in the outer fuel pin during irradiation.

Table 3
Fuel centre temperature predicted by computer code PROFESS.

Fuel pin	Predicted fuel centre temperature (°C)	
	BOL	EOL
Outer fuel pin	1387	1605
Intermediate fuel pin	1059	1163
Central fuel pin	976	1065

5.3. PROFESS calculation of fuel temperature and fission gas release

The fuel centre temperatures in the outer, middle and central fuel pins during irradiation predicted by computer code PROFESS are shown in Fig. 2. The fuel centre temperatures at the beginning of life (BOL) and at the end of life (EOL) are presented in Table 3. The plot of predicted fission gas release vs. burnup in the outer fuel pin during its irradiation is given in Fig. 3. It is observed that fission gas release in the fuel pin starts only at a burnup of about 3000 MWd/tU, which seems to be the threshold or incubation burnup for the gas release. Thereafter, the gas release steadily increases with burnup. It is observed from Fig. 2 that the temperature of the fuel increases with burnup in all three types of fuel pins. A change in the slope of the temperature curve is observed in the case of outer fuel pin soon after exceeding a burnup of about 3000 MWd/tU, which corresponds to the start of fission gas release in the fuel pin (Fig. 3).

5.4. Cladding deformation during high temperature heating

Isothermal heating test was performed on four outer fuel pins which had the highest internal pressure. Three fuel pins which were heated at 800 °C, 850 °C and 900 °C for 10 min showed diametral deformation but no failure. The fourth fuel pin was heated at 900 °C for 15 min. Leak testing showed that this fuel pin had failed. Appearance of the two fuel pins, one heated at 850 °C for 10 min and other at 900 °C for 10 min is shown in Fig. 4. An increase in the diameter of the fuel pins in the heated portion is clearly visible in Fig. 4(a). The axial diametral profiles of the heated fuel pins are shown in Fig. 5. This figure shows that the maximum diametral deformation in the fuel pin increases with increasing temperature for constant heating time of 10 min. It is also observed that the maximum diametral strain also increases by increasing the heating time at the same temperature. This clearly demonstrates that the deformation is occurring by creep of the cladding. The measured maximum diametral strain and the average strain rate in the fuel pins are given in Table 4.

6. Discussion

6.1. Fission gas release behaviour

The measurement of fission gas release has shown that the outer fuel pin showed significant release but release in the other fuel pins was marginal. The linear power rating and the fuel centre temperature in the outer fuel pins are higher than those in the other fuel pins. This could be the reason for the appreciable fission gas release observed in the outer fuel pins. The difference in the fission gas release behaviour of the outer, middle and central fuel pins is explained below in terms of fission gas release threshold.

The fission gas release in UO_2 fuel depends on fuel temperature, burnup, and the grain size of the fuel. Based on the fission gas release data of fuel pins irradiated in Halden BWR, a threshold for 1% fission gas release in terms of the fuel centre temperature and burnup has been defined by Vitanza et al. [18,19]. This threshold is defined empirically as follows:

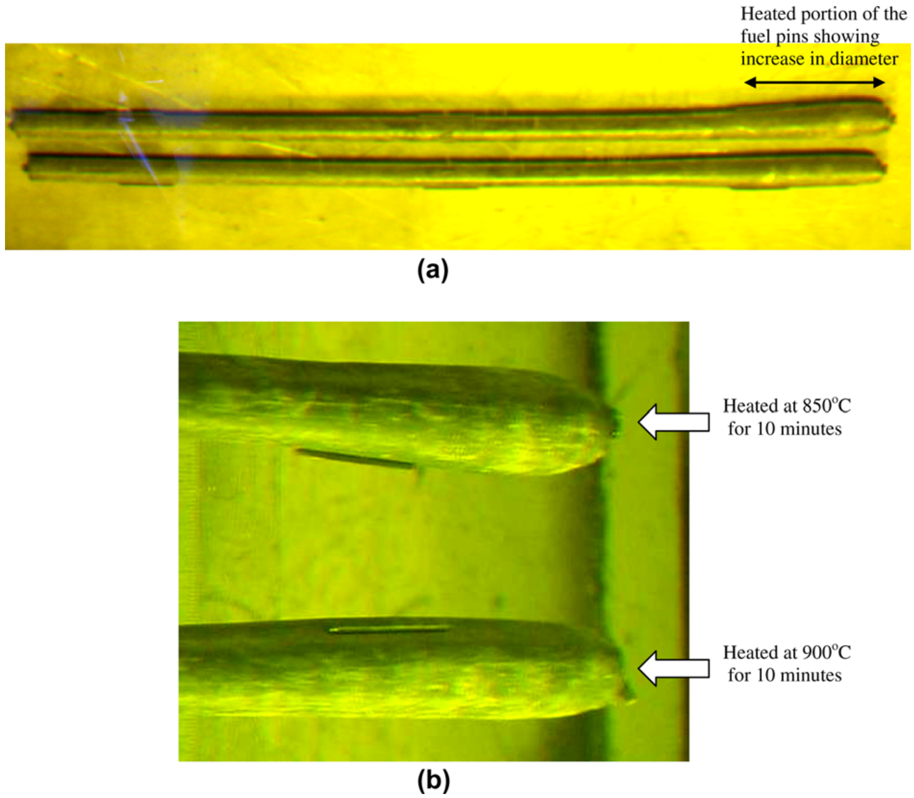

Fig. 4. The appearance of fuel pins after heating: (a) full view of two heated fuel pins showing diameter increase in the heated portion and (b) close-up view of the heated end of the fuel pins.

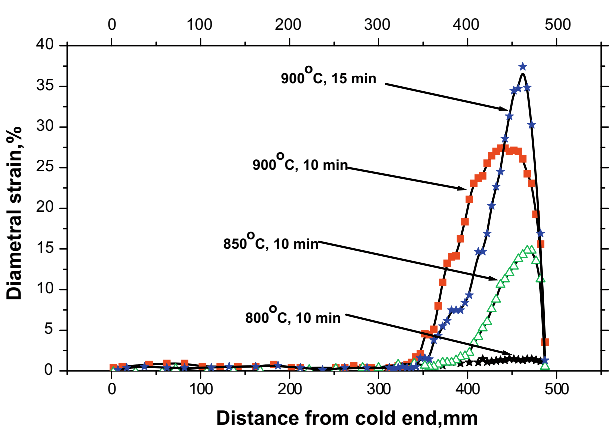

Fig. 5. Axial diametral profile of fuel pins subjected to heating.

$$Bu\ (\mathrm{MWd/kgUO_2}) = 5 \times 10^{-3} \exp(9800/T_{\mathrm{centre}}), \tag{4}$$

where Bu is the burnup of the fuel in $\mathrm{MWd/kgUO_2}$ and T_{centre} is the fuel centre temperature in °C.

According to this empirical relation, if the burnup in the fuel pin is less than the threshold burnup, then for a given fuel centre temperature the fission gas release will be insignificant (<1%).

While the abovementioned Vitanza threshold (also referred to as Halden threshold) is derived from the Halden experimental data, the theoretical consideration of gas release by solid state diffusion of fission gases in the fuel also indicates the existence of an incubation burnup or threshold burnup below which the fission gas is not released from the fuel. This threshold is related to the grain boundary saturation by fission gas bubbles, which is an essential condition for fission gases to get released from the fuel.

This threshold burnup for grain boundary saturation [20] is defined by the following correlation:

$$\text{Threshold Bu} = \frac{b\lambda N_{max}}{2D_{\phi}}, \tag{5}$$

where b is resolution probability from grain boundary (s^{-1}), λ is resolution layer depth (cm), D is Diffusion coefficient (cm^2/s), N_{max} is Number of gas atoms per cm^2 area of grain boundary at saturation and ϕ is number of gas atoms generated per cc per unit burnup.

Threshold for 1% fission gas release has also been estimated using above criterion in computer code PROFESS [10] and it is found that at low burnup (less than 10 000 MWd/tU) this threshold is slightly lower but close to Vitanza threshold.

Fig. 6 shows the Halden threshold and PROFESS threshold along with plot of variation of fuel centre temperature in the outer, middle and central fuel pins during their irradiation. It is observed that the fuel centre temperatures in the intermediate fuel pin and the

Table 4
Cladding diametral strain in the fuel pins during heating.

Sr No	Temperature of heating (°C)	Time of heating (min)	Max. cladding strain	Average strain rate (s^{-1})
1	800	10	0.0144	2.4×10^{-5}
2	850	10	0.148	24.6×10^{-5}
3	900	10	0.274	45.6×10^{-5}
4	900	15	0.37	Leak testing showed failure

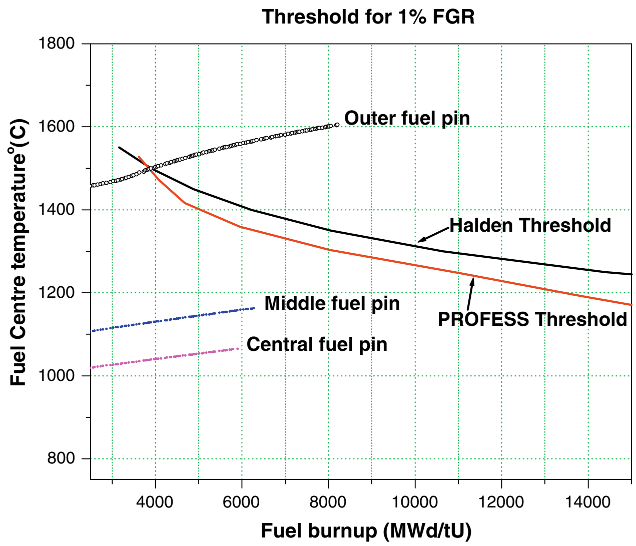

Fig. 6. Fission gas release threshold and fuel centre temperatures in the PHWR fuel pins examined.

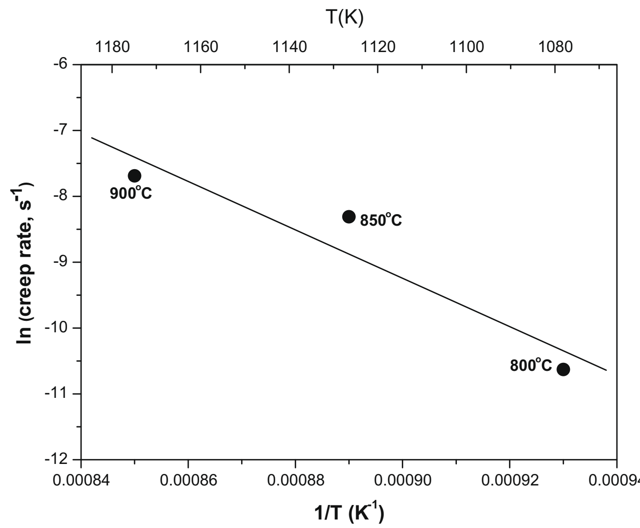

Fig. 7. Plot of creep rate vs. inverse of temperature.

central fuel pin were below the threshold lines, throughout their irradiation. Therefore, little release was expected in these fuel pins as observed from the experimental results. However, the fuel centre temperature in the outer fuel pin is observed to have exceeded the threshold during its irradiation, causing substantial fission gas release in this fuel pin.

6.2. Comparison of PROFESS results with actual measurements

A comparison of predicted and measured values of fission gas release and internal pressure in the fuel pins is given in Table 5. Good agreement is observed between the predicted values and the measured values of internal pressure in the fuel pins. However, there is an under-prediction of fission gas release in the outer fuel pin. The reason for this under-prediction is that the present model of fission gas release used in the code has not considered the release contribution by grain boundary sweeping of gases which is likely to be operating in this fuel pin. It may be noted that the fuel centre temperature in the outer fuel pin during its irradiation was calculated to be in the range 1400 °C–1600 °C which is sufficient to cause equiaxed grain growth in the central region of the fuel promoting fission gas release by grain boundary sweeping from that region. Incorporating contribution of grain boundary sweeping mode in the FGR model of the code could result in better agreement between the prediction and the measurement in this fuel pin.

6.3. Creep rate of cladding

6.3.1. Temperature dependence

In order to derive a creep rate correlation from this data, the strain rate of the cladding in these fuel pins at the location of maximum deformation is plotted as a function of temperature in the form of Arrhenius plot (natural logarithm of strain rate vs. inverse of temperature) in Fig. 7. The data from the failed pin is not included while plotting. The activation energy estimated from the slope of the line is found to be 305.5 kJ/mol which is found to be close to the mean of values of activation energy reported in literature [13–17,21,22]. Arrhenius equation, giving temperature dependence of average diametral creep rate at near-constant stress of 36 MPa, is represented as follows:

$$\text{Creep rate } (s^{-1}) = 2.23 \times 10^{10} \times \exp(-305500/RT), \tag{6}$$

where R is gas constant, 8.314 J/mol K and T is temperature in K.

6.3.2. Comparison with literature

The creep rates derived from the diametral strain data of the fuel pins heated at 800 °C and 850 °C are compared with the creep rates calculated using correlations available in literature and are given in Table 6. It is observed that the creep rates in the fuel pins at 800 °C and 850 °C match well with the rates calculated using correlations of Clay and Redding [13,14]. However, the values were lower than the creep rates calculated using Rose and Hindle correlation [15]. It may be noted that correlations available in literature are derived from tests performed on unirradiated empty cladding tubes or on samples from unirradiated cladding material. In the present case the measurements were carried out on actual irradiated fuel pins filled with fuel pellets. The cladding in these fuel pins is in irradiated condition and also has oxide layers on the outer and inner surfaces. The cladding tube in the fuel pins also has constraints on the cladding surface in the form of appendages like bearing pads and spacer pads. Any difference between the observed creep rates and rates calculated using correlations given in literature could be attributed to the differences related to metallurgical conditions and geometry of the samples.

Table 5
Comparison of predicted and measured FGR and internal pressure.

Fuel pin	Fission gas release (%)		Internal pin pressure at room temperature (MPa)	
	Predicted	Measured	Predicted	Measured
Outer	5.3	7.8–8.8	0.53	0.51–0.59
Intermediate	0	0.1–0.2	0.12	0.14–0.15
Central	0	0.1	0.12	0.12

Table 6
Comparison of calculated and measured creep rates.

Heating temperature (°C)	Calculated diametral creep rate (s^{-1})		Measured creep rate (s^{-1})
	Clay and Redding [13,14]	Rose and Hindle [15]	
800	5.8×10^{-5}	16.2×10^{-5}	2.4×10^{-5}
850	25.9×10^{-5}	67.8×10^{-5}	24.6×10^{-5}

7. Summary

The main findings of studies carried out on the fuel pins of PHWR bundle discharged after an average burnup of 7528 MWd/tU can be summarized as follows:

1. The fission gas release from the fuel pins from outer ring of the fuel bundle was about 8%. Fuel pins from the intermediate ring and the central pin released <1% of fission gas. It is believed that grain boundary sweeping of gases contributed to the fission gas release in the outer fuel pin.
2. The internal gas pressure in the outer fuel pin was found to be 0.55 ± 0.05 MPa at room temperature. The internal pressure in the central and intermediate fuel pins was in the range 0.12–0.15 MPa. Internal pressure predicted by computer code PROFESS in the outer fuel pin was 0.53 MPa which is close to the measured value.
3. The maximum calculated fuel centre temperatures in the outer pin, the intermediate pin and the central fuel pin were 1605 °C, 1163 °C and 1065 °C, respectively. While the temperatures for the middle ring fuel pins and the central pin had remained below the threshold required for significant fission gas release, during their irradiation, the centre temperature for the outer pin had exceeded the threshold level.
4. The creep rate of the cladding of the outer pin with an internal pressure of 0.55 ± 0.05 MPa on heating at 800 °C, 850 °C and 900 °C was found to follow the following correlation:

   $$\text{Creep rate } (s^{-1}) = 2.23 \times 10^{10} \times \exp(-305500/RT)$$

   where, R is the gas constant and T is the temperature in K.
5. The measured value of steady state creep rate of Zircaloy cladding in α phase region matched well with rates calculated using the available correlations in literature.

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