Fission product and swelling behaviour in FBTR mixed carbide fuel

C.N. Venkiteswaran, V.V. Jayaraj, B.K. Ojha, M. Padalakshmi, C. Padmaprabu, V. Anandaraj, T. Saravanan, Ran Vijay Kumar, V. Karthik, A. Vijayaraghavan, T. Johny, Divakar R., B.P.C. Rao, Jojo Joseph

[1]Indira Gandhi Centre for Atomic Research, Kalpakkam, India

E-mail contact of main author: cnv@igcar.gov.in

Abstract. The advantage of a fast reactor, especially one that uses Uranium-Plutonium Carbide as its fuel, is well documented. Irradiation performance assessment of carbide fuels began with experimental irradiations in EBR-II, FFTF, HFR, Rapsodie and Phenix etc. India has the extensive experience with this type of fuel at the Fast Breeder Test Reactor at Kalpakkam that has been operating for over 25 years. The fuel has attained a peak burn-up of 155 GWd/t at linear heat rating of 400 W/cm, in a large number of fuel pins. Comprehensive post-irradiation examinations (PIE) at various stages up to this high burn-up have yielded a wealth of information on behaviour of mixed-carbide fuel under steady state operations. In this paper, selected recent results on fission product migration, gas release, fuel swelling behavior and micro-structural evolution of the mixed-carbide fuel will be presented. Results of the PIE towards analysing the cause of failure in a fuel pin are also discussed.

Key Words: Carbide fuel, fuel swelling, fission product distribution, microstructure

1. Introduction

Fast Breeder Test Reactor at Kalpakkam achieved criticality in the year 1985 with 22 Mark I fuel subassemblies containing plutonium rich carbide fuel $(U_{0.3}\ Pu_{0.7})C$. The core has been progressively expanded with additional subassemblies containing Mark I fuel, Mark II fuel having a composition of $(U_{0.45}\ Pu_{0.55})C$ and mixed oxide fuel $(U_{0.56}\ Pu_{0.44})C$ to increase the reactor power. Table 1 gives the salient specifications of the Mark I carbide fuel. FBTR is also being used for test irradiation of future candidate fuel and structural materials including sphere-pac fuel and metallic fuel.

Irradiation experience with the carbide fuel in the reactor amounts to over 1500 fuel pins achieving target burn-up of 155GWd/t. The fuel mixture used was relatively unknown at that time, necessitating a systematic and periodical examination of the irradiated fuel for its performance characteristics as well as for detecting any early signs of life-limiting phenomena [1]. The burnup of this fuel was progressively enhanced from the initial design burnup limit of 25 GWd/t based on the performance feedback from post irradiation examinations at different burnup levels. The feedback from PIE has resulted in the enhancement of the burnup of this unique fuel to 155 GWd/t [2]. The fuel has operated at a peak linear heat rating of 400 W/cm. As a part of performance assessment of the mixed carbide fuel, swelling, fission product behaviour and microstructural evolution have been evaluated.

2. Post Irradiation Examination of FBTR mixed carbide fuel

PIE on the mixed carbide fuel subassemblies have been carried out at the hot cells of Radiometallurgy Laboratory co-located with FBTR. The hot cells are equipped with facilities and equipments for non-destructive and destructive examinations on the FBTR in-core materials [3]. PIE has been carried out on the fuel pins operating at different linear heat ratings on account of its position in the core and resulting flux level. The subassemblies discharged after 25 GWd/t was irradiated at the core centre, whereas the subassemblies after burnups of 50, 100 & 155 GWd/t were irradiated in the first ring which have seen peak LHR of 400 W/cm. Recently PIE has been carried out on two fuel subassemblies irradiated in third ring which have operated at lower LHR of 285 W/cm. One of the subassemblies in the third ring had undergone fuel pin failure at a burnup of 148.3 GWd/t. The fuel pins of this subassembly have undergone comprehensive PIE to investigate the cause of failure. Table I gives the specifications of FBTR Mark I fuel. Fig. 1 shows the irradiation location of the subassemblies in the core which have been subjected to PIE. Table II gives the irradiation data of the subassemblies examined.

The examination of the subassemblies operated at different LHR has provided valuable information on the behaviour of carbide fuel with respect to the swelling, fission product redistribution, gas release and microstructural evolution. The salient results of the examinations are given below.

Table I Specifications of FBTR Mark I fuel

No. of fuel pins per subassembly	61
Fuel	Mark I:($U_{0.3}$ $Pu_{0.7}$)C
Fuel stack length	320 mm
Type of bond	Helium
Linear mass g/cm	1.67 g/cc
Fuel Density (% TD)	90 % of T.D.
Smear Density (% TD)	83 % of T.D.
Pellet diameter	4.18 mm
Outer diameter of fuel pin	5.1 mm
Inner diameter of fuel pin	4.36 mm
Clad & Wrapper material	20% CW AISI 316

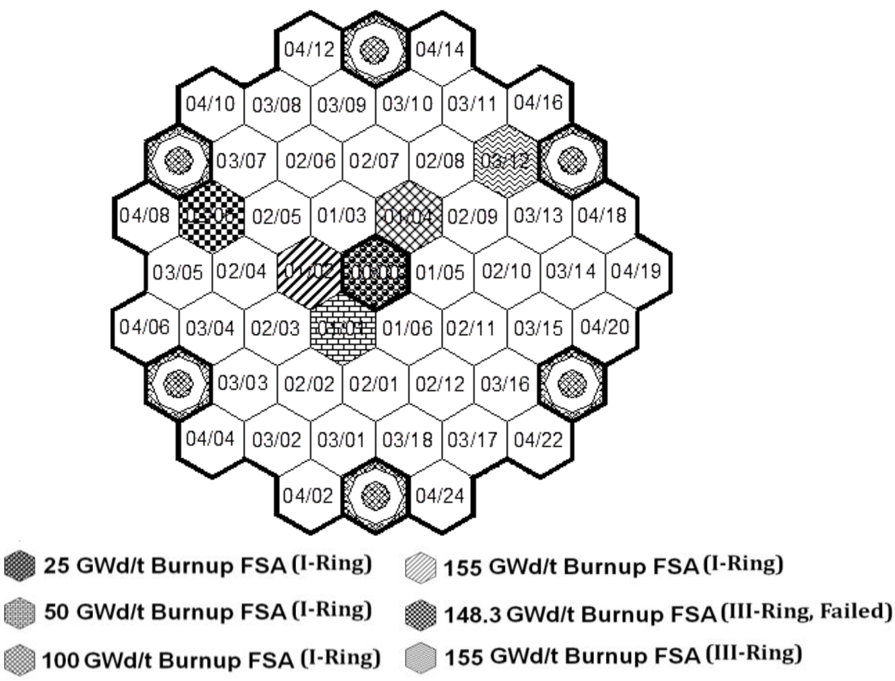

Fig. 1 Irradiation locations of fuel subassemblies subjected to PIE

Table II Irradiation data of Mark I fuel subassemblies

Irradiation parameter	Fuel subassemblies subjected to PIE					
	I ring subassemblies				III ring subassemblies	
Peak Burn-up (GWd/t)	*25*	49.2	101.5	155	148.3	155
Peak Linear power (W/cm)	320	320	400	400	285	285
Peak 'dpa'	14	28	56	83	87	87

2.1 Fuel swelling

Swelling of the Mark I carbide fuel was measured through diverse non-destructive examination techniques such as X-radiography, Neutron Radiography and Gamma scanning. Fuel stack length of selected pins from each subassembly was measured using the above techniques.

Table III gives the fuel stack length measured from radiographs in the fuel pins of different burnups. Progressive increase in stack length was observed with increasing burnups. Increase in stack length was found to be significantly higher in the fuel pins of III ring subassemblies as compared to pins irradiated in I ring having similar burn-ups. Fig. 2 shows the histogram of the average stack length increase observed in the fuel pins irradiated in I ring and III ring. Localised bending was observed in the failed pins and the surrounding pins as shown in the radiographs (Fig.3). Such localised bending were not observed in the fuel pins examined at different burnup levels which were irradiated in the I ring.

Table III Stack length increase in 320 mm fuel column

	Stack length measurement in fuel pins					
	I ring subassemblies				III ring subassemblies	
Burnup (GWd/t)	25	49.2	101.5	155	148.3	155
No. of fuel pins examined	09	09	09	13	08	04
Fuel stack length increase (mm)	2.17- 5.35	4.1 -5.32	3.23 - 8.35	8.47-11.71	13.6-15.6	14.68 - 18.55

2.2 Fission product redistribution

Fission product redistribution in the irradiated carbide fuel pins have been evaluated at different burnup levels through axial gamma scanning. The details of the gamma scanning system for irradiated fuel pins are given in [4].

Fig. 4 shows the axial distribution of ^{137}Cs in one of the fuel pins from a subassembly irradiated in the I ring after attaining a peak burnup of 155 GWd/t. The axial distribution of fission products such as ^{106}Ru & ^{95}Zr was commensurate with axial flux profile. Peak of ^{137}Cs was observed near the fuel pellet-insulation pellet interface indicating its axial migration.

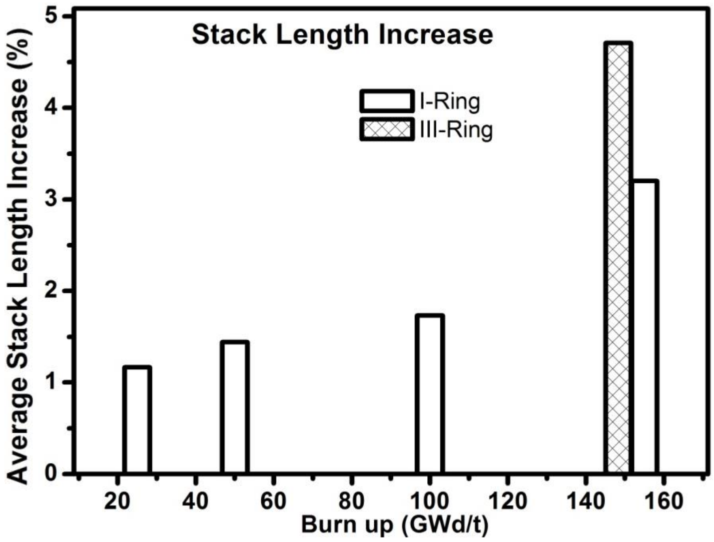

Fig. 2 Histogram showing stack length increase in fuel pins irradiated at different core locations

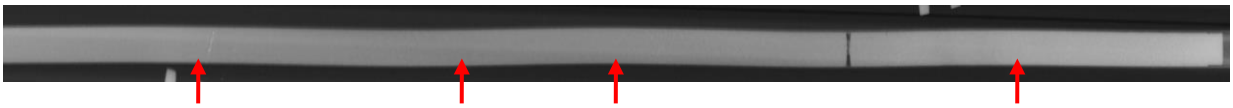

Fig. 3 Localised bend observed in the radiograph of a fuel pin irradiated in III ring

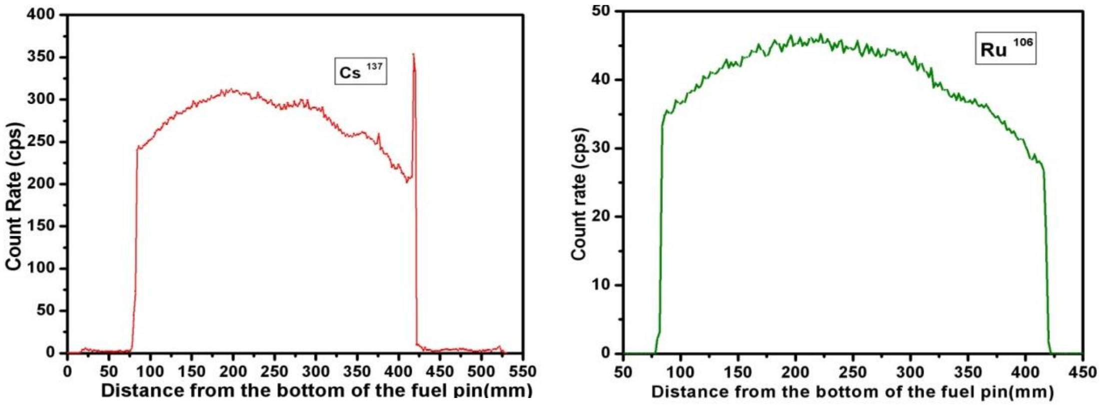

Fig. 4 Axial distribution of ^{137}Cs and ^{106}Ru in the fuel pins of subassemblies discharged from I ring after a burnup of 155 GWd/t

Fig. 5 shows the axial distribution of ^{137}Cs and ^{106}Ru in two fuel pins of a subassembly discharged from III ring. This particular subassembly which had attained a peak burnup of 148.3 GWd/t had a clad rupture in one of the fuel pins. Fig. 6 shows the arrangement of fuel pins in the failed subassembly indicating the location of the failed pin with respect to the core centre.

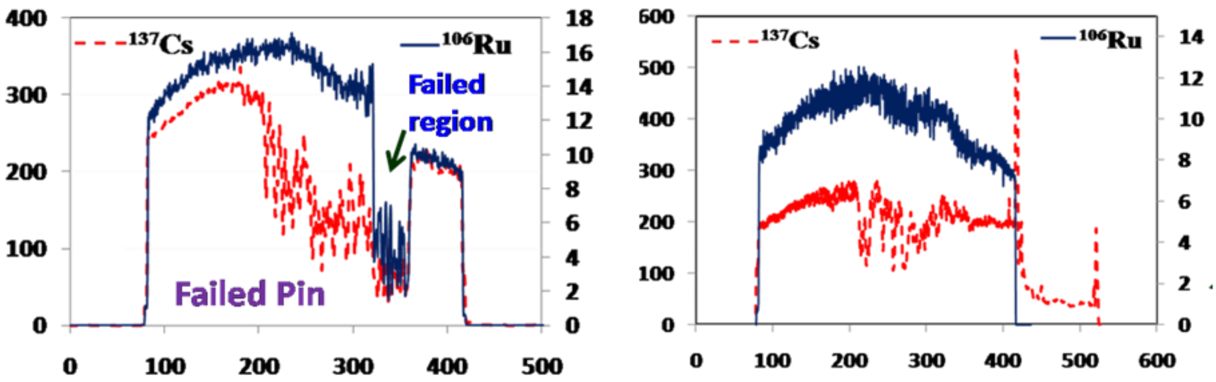

Fig. 5 Axial distribution of ^{137}Cs and ^{106}Ru in the fuel pins of a failed subassembly irradiated in III ring

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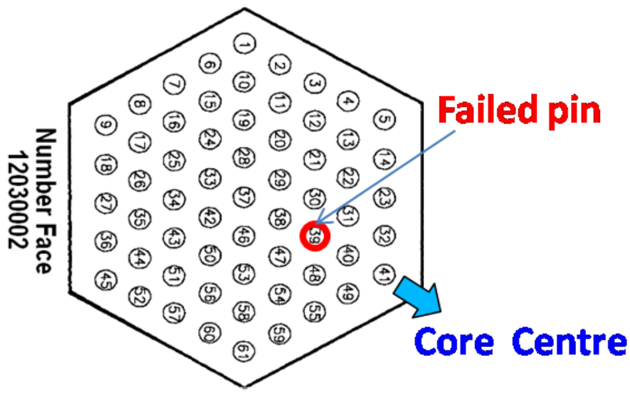


Fig. 6 Location of the failed fuel pin in the pin cluster

It is evident from the axial profile of ^{137}Cs in the failed pin that it has undergone severe redistribution at axial locations starting near the peak power location extending upto the top of the fuel column. Similar behaviour was also observed in the pins adjacent to the failed pin. However, no such migration was noticed in the pins located far away from the failed pin within the subassembly. Axial distribution of ^{137}Cs obtained from a fuel pin of another subassembly irradiated in III ring also exhibited similar behaviour (Fig. 7).

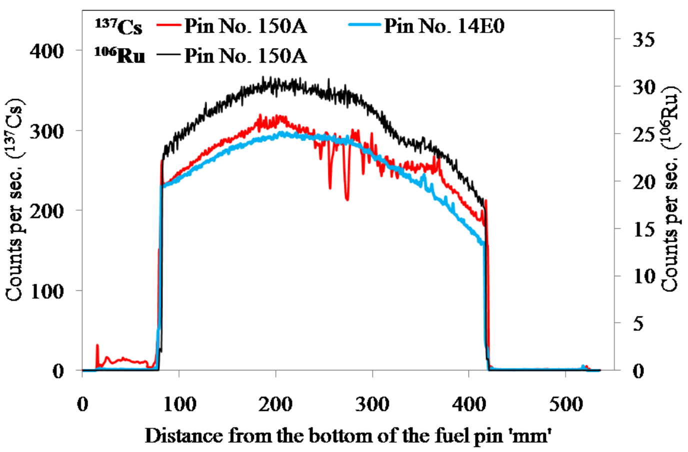


Fig. 7 Axial distribution of ^{137}Cs and ^{106}Ru in the fuel pins from an intact subassembly irradiated to a burnup of155 GWd/t in III ring

2.3 Fission gas release

Fission gas release measurements were carried out on selected fuel pins by puncture tests and and analyzed by gas chromatograph. Percentage of gas release, internal pressure and Xe/Kr ratio was measured. Table 4 gives the gas release and fission gas pressure measured in the fuel pins.

Fission gas release was found to be around 1 % in 25 GWd/t burn-up fuel pins. Fission gas release values were varying between 7% - 18 % in the case of 50 GWd/t burnup and the corresponding values for 100 GWd/t burn-up was in the range of 4 – 14 % [5]. Considering that yield of fission gas release in 100 GWd/t burnup is two times as that of 50 GWd/t burnup, percentage gas release values were marginally lower in 100 GWd/t burnup fuel pins. This could be attributed to the lower fuel temperature due to significant reduction in the fuel-clad gap by fuel swelling which was indeed confirmed from the micrographs of the fuel pin cross-sections. The fission gas pressure showed a monotonic increase with increasing burnups. Maximum fission gas release measured in fuel pins irradiated in I ring after 155 GWd/t burn-up was 16 %. The corresponding internal pressure in the fuel pin was 2.1 MPa. The ratio of Xenon to Krypton was measured to be around 13 which is expected for fissions in plutonium.

The fission gas release in the fuel pins of the failed subassembly irradiated in III ring showed two different trend as observed in the redistribution ^{137}Cs during gamma scanning. The fuel pins surrounding the failed pin showed higher gas release as compared to the fuel pins irradiated in I ring having similar burnup. However, the fuel pins located farther from the failed pin in the failed subassembly indicated significantly lower gas release as compared to the I ring pins as shown in Table III. Fig. 8 shows the histogram of maximum fission gas pressure in the fuel pins at different burnup levels.

Table III Fission gas release and pressure in FBTR fuel pins

<table>
<tr><th rowspan="2"></th><th colspan="6">Fission gas release and pressure</th></tr>
<tr><th colspan="4">I ring subassemblies</th><th colspan="2">III ring subassemblies</th></tr>
<tr><td>Burnup (GWd/t)</td><td>25</td><td>49.2</td><td>101.5</td><td>155</td><td>148.3</td><td>155</td></tr>
<tr><td>No. of fuel pins examined</td><td>05</td><td>05</td><td>05</td><td>04</td><td>09</td><td>02</td></tr>
<tr><td rowspan="2">Fission gas release %</td><td rowspan="2">0.5 – 1.2</td><td rowspan="2">7 - 18</td><td rowspan="2">4 - 14</td><td rowspan="2">8.5 - 15.7</td><td>17 - 19.2 *</td><td rowspan="2">--</td></tr>
<tr><td>4.0 - 8.4 #</td></tr>
<tr><td rowspan="2">Pressure at ambient temp. (MPa)</td><td rowspan="2">0.12</td><td rowspan="2">0.22-0.77</td><td rowspan="2">0.5 – 1.2</td><td rowspan="2">1.15 – 2.0</td><td>2.0 -2.25 *</td><td rowspan="2">0.54– 1.07</td></tr>
<tr><td>0.53 – 1.1</td></tr>
</table>

* Gas release and pressure in pins adjacent to failed pin #Gas release& pressure in farther pins

2.4 Fuel microstructure

Micrographs of the fuel pin cross-sections of 25 & 50 GWd/t burnup fuel pins indicated radial cracks and progressive reduction in the fuel-clad gap due to fuel swelling. The free swelling rate of FBTR carbide fuel was estimated from the image analysis of micrographs [6]. It was found to be in the range of 1 – 1.2% per atom percent burnup which is lower than anticipated during fuel design [1].

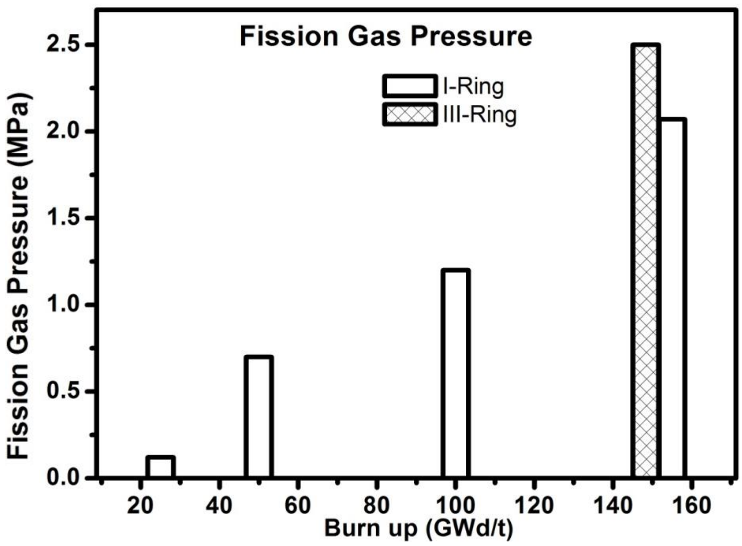
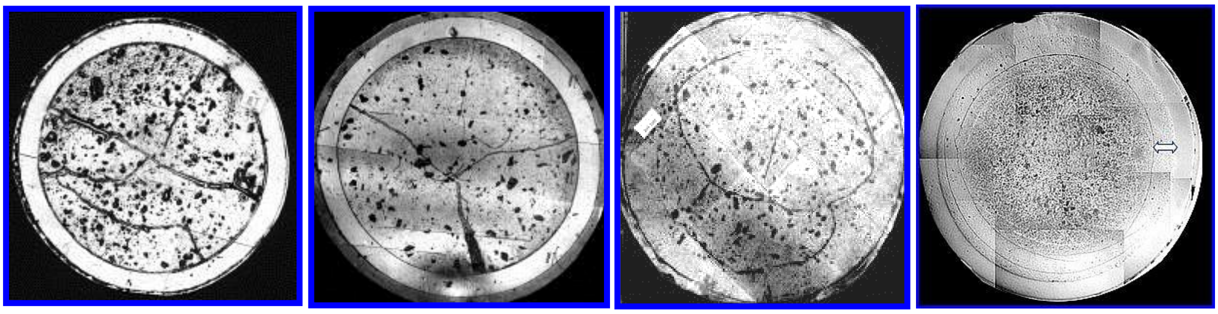


Fig. 8 Histogram showing the fission gas pressure in the fuel pins at different burnups

In 100 & 155 GWd/t burn-up fuel pins, the fuel-clad gap had closed completely along the entire length of the fuel column. Fuel pin cross-sections showed circumferential cracks indicating restrained swelling of the fuel. A porosity free outer rim was observed near the outer diameter of the fuel in 155 GWd/t fuel pin cross-section at the centre of the fuel column. The porosity exhaustion is attributed to the creep of the fuel due to FCMI. Fig. 9 shows the photomosaics of fuel cross-sections at the centre of the fuel column after 25, 50, 100 & 155 GWd/t burn-up.



25 GWd/t **50 GWd/t** **100 GWd/t** **155 GWd/t**

Fig. 9 Photomosaics of fuel pin cross section at the centre of fuel column after burn-up of 25, 50, 100 & 155 GWd/t

The fuel pin cross-section of the failed pin irradiated in III ring showed asymmetric circumferential cracking pattern in some of the axial locations which is indicative of non uniform temperature distribution around the pins. Fig. 8 shows the micrographs of fuel pin cross-sections in a pin adjacent to the failed pin.

While the axial locations at the bottom of the fuel column indicated symmetric cracks similar to those observed in the fuel pins irradiated in I ring, those above the peak power location towards the top of the fuel pin showed asymmetric circumferential cracks. This asymmetry is attributed to the constriction of coolant flow in localised regions around the pins, where the pins are in near-contact with the adjacent pins due to diameter increase and localised bending.

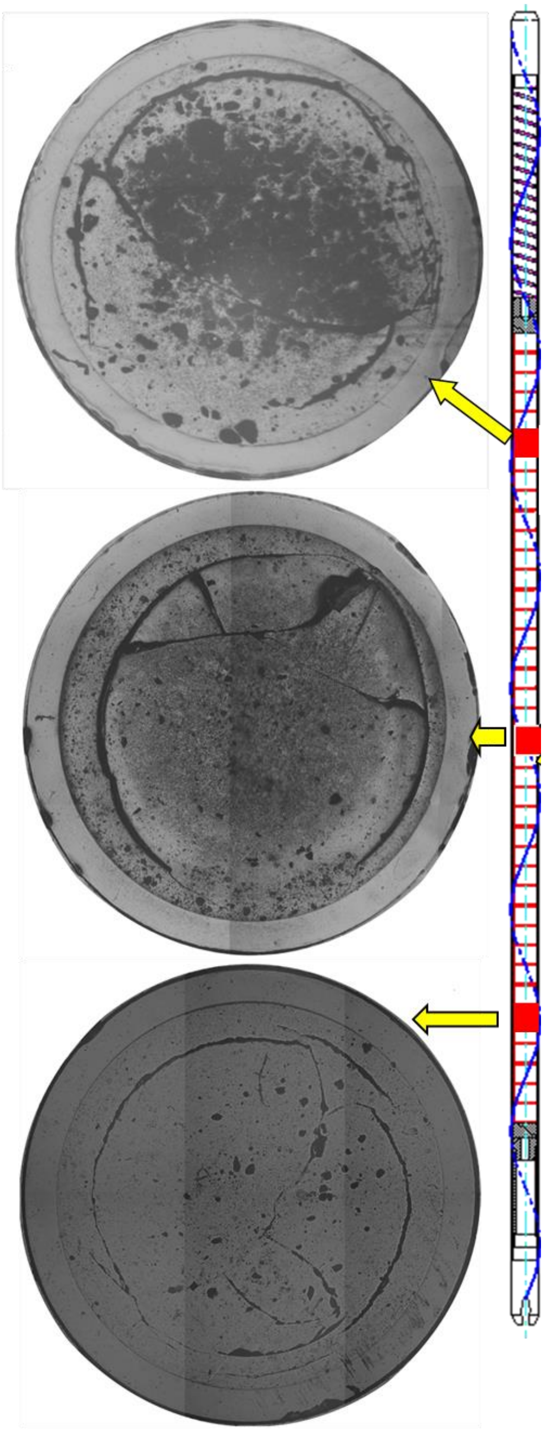


Fig. 10 Photomicrographs of the cross-sections of a fuel pin irradiated in III ring

3. Summary & Discussions

PIE of mixed carbide fuel pins operated at different linear heat ratings exhibited considerable variation in the irradiation behaviour with respect to swelling, axial migration of volatile fission products, fission gas release and microstructure.

While, the axial re-distribution of ^{137}Cs was very subtle in the fuel pins irradiated in I ring, its redistribution was found to be more severe in the fuel pins of a failed subassembly from the

III ring. The fact that such redistribution was observed even in the fuel pins of another intact subassembly irradiated in III ring reveals that the factors leading to such behaviour is generic due to different operating conditions of the fuel pin in the I ring and the III ring. The fuel pins of III ring also exhibited higher diameter and length increase, fuel stack length increase as well as localised bends. The localised bends and higher diameter increase could result in localized reduction in coolant flow and non-uniform temperature distribution around the pins affecting the migration of volatile fission products.

Fission gas release measurements indicated that the gas release in general is lower as expected for carbide fuels. However, different trends in the gas release were observed in the fuel pins irradiated in I ring and the fuel pins of failed subassembly from the III ring. The gas release were higher in the fuel pins adjacent to the failed pin as compared to the peripheral fuel pins which showed very low gas release. The higher gas release around the failed pin also could be attributed to highly localized temperature increase at the failure region within the hexcan.

Metallographic sections of the fuel pins showed asymmetric circumferential cracking pattern in some of the axial locations. Such asymmetric crack pattern can occur due to the non-uniform coolant flow around the pins at the locations where the fuel pins have undergone localized bending and near contact with the adjacent pin.

From the comprehensive examinations carried out on the fuel pins irradiated at different core positions and their irradiation behaviour, the following can be concluded. The linear heat rating of the fuel pins in the third ring was significantly lower than the corresponding value for the pins of the subassemblies in the central location or in the I ring, Typical peak LHR of the fuel pins in the failed FSA is around 280 W/cm as compared to the peak LHR of 400 W/cm in the I ring. The lower heat rating results in lower fuel temperatures and temperature gradients leading to lower fission gas release.

It is well known that carbide fuel tends to have high fission gas retention due to its high thermal conductivity and lower fuel temperatures. The irradiation experience in USA and European countries on carbide fuel pins shows that the fission gas release (FGR) is very modest, in the range of 10 - 30 % at 5 at % burnup even at high heat rating of 700 - 1200W/cm and smear densities of 75 - 80% of theoretical density [7]. Apart from lower gas release, the lower fuel temperature results in lower creep of fuel and lesser accommodation of the fuel swelling in sinter porosities.

Higher fuel swelling in III ring pins as indicated above has led to severe Fuel Clad Mechanical Interaction (FCMI) stresses on the cladding which could have caused localised bending the fuel pins and temperature increase affecting the fission product behaviour and microstructure. The failure of the clad is attributed to the high FCMI stresses generated in the fuel pin.

4. Conclusion

The performance assessment through PIE has provided valuable insights into the behavior of the mixed carbide fuel at different burnups. Feedback from PIE was crucial to extend the burnup of the fuel beyond the conservative design limits. Fuel behaviour in the failed subassembly provided important inputs to analyse the cause of the failure. The authors acknowledge the contributions from the fuel fabricators at BARC, Mumbai and several colleagues from the reactor facilities group, design group and chemistry group at IGCAR for many useful inputs and discussions.

5. References

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