Metallography of High Burnup PHWR Fuel

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Abstract

Detailed metallographic examination and β-γ autoradiography have been carried out on the samples taken from the fuel pins of two high burn-up bundles. The observed microstructural features indicate that the main mechanism of fission gas release in the fuel has been formation of interconnected grain boundary fission gas bubbles at the hot central region of the fuel. Quantitative information generated has shown that these bubbles grow, interconnect and finally get released through the grain boundary channels.

Keywords- Fission Gas Bubbles, Grain Boundary Pores, High burn-up PHWR Fuel, Microstructure, β-γ autoradiography

Introduction

Two high burn-up fuel bundles with approximately 15,000 MWd/T burn-up from Kakrapar Atomic Power Station were examined to asses their performance. Visual examination and leak testing did not reveal any evidence of failure of the fuel bundle.

In order to evaluate the microstructural changes evolved during irradiation and to asses their bearing on the performance of fuel specially related to fission gas release, detailed metallographic examination has been carried out on the samples taken from the outer, intermediate and central pin of the bundles. The amount of fission gas released was significantly more in the outer pins. This paper elucidates the fission gas release pattern as revealed from the observed microstructural features.

Experimental

Detailed metallographic examination[1,2,3,4] has been carried out on the samples taken from the outer, intermediate and central pin of the bundles.

The metallographic preparation of the transverse section has been carried out in the hot cells using a remotised grinding-polishing equipment. The samples were ground on 180 grit SiC paper till the cross sections have been completely exposed and then progressively ground on 240, 320 and 600 grit SiC papers. The ground samples were polished using diamond slurry of 3μm and then 1μm size. The samples have been examined in as polished and etched conditions using a remotised metallograph. β-γ autoradiography has been carried out in as polished condition to reveal the fission product redistribution pattern.

Observations

The observations on the two bundles were nearly identical; typical observations of one of the bundles are detailed below.

Microstructural examination revealed two distinct regions in the fuel. At low magnification the central region was dark and the peripheral region was relatively bright. Fig.1 shows the photomicrographs and the corresponding β-γ autoradiographs of the fuel section. When examined at high magnification the central dark region revealed the presence of large number of big size pores (Fig.2&3) and the peripheral region revealed the as fabricated microstructure.

In the outer pin, the central dark porous region had extended up to about the mid radius. The nature of the features changed with radial location in the central dark porous region (Fig.3). At the periphery of the dark region, the pores have been observed in the grain matrix and at the grain boundary. The pores have been predominantly at grain boundaries with very few in the grains at the center of the dark region. The pores observed in the central region have been either isolated or interconnected and big sized. The size of the dark zone decreased in the intermediate pin and central pin in that order when compared to the outer pin.

The β-γ autoradiographs (Fig.1) of the samples revealed depletion of radioactivity in the central region of the fuel pellet. This indicates that volatile fission products such as cesium have migrated from the center to the periphery of the fuel pellet. The size of fission product depleted zone decreased in the intermediate pin and central pin as in the case of the dark zone.

Quantitative microstructural information at different radial locations of various sections of the fuel element has been generated to understand the observed difference in fission gas release in different fuel elements. The radial profiles of grain size and porosity have been generated.

The microstructures revealing the grain structure at different radial locations of a section of the outer pin is shown in Fig.3. The plot of the grain size verses radial locations is given in Fig. 4. In the outer pin the grain size at the peripheral region did not vary significantly but the grain size began to increase abruptly from the mid radial location. The intersecting point of the linear fit of the grain size versus radial location of the peripheral and inner region has been chosen as the starting point of grain growth. Using this as the reference point, the temperature at the center of the section has been estimated.

The percentage porosity versus the estimated temperature is plotted in Fig.5. The percentage porosity has been found to remain unaltered in the two measurements carried out at periphery and central locations of the central element. But in the case of intermediate and outer fuel pins the percentage porosity has been found to increase from the boundary of the dark porous region to the center of the sections. The plot of number of small pores (<4microns), round pores and interconnected pores along the radius of the outer pin is shown in Fig.6.

The area fraction of total crack of the section, including the pellet clad gap area has been measured. The crack area percentage of outer, intermediate and central pins, have been shown with the estimated central temperatures in Fig.7. The total area fraction covered by fuel-cracks and pellet clad gap in the fuel section has increased with the central temperature of the pin.

Discussion

Dark Porous Region:

The fission gas atoms are highly mobile at the central region of the fuel due to the higher temperature. The mobile fission gas atoms migrate to the grain boundaries and coalesce to form bubbles. These fission gas bubbles appear as pores in the fuel section because of which the central region appears dark at low magnification. The bubbles get interconnected and finally the fission gas gets released from the fuel. Hence, the fission gas release in the pin is related to the area of the dark porous region.

Grain Size and Central Temperature:

The measured grain size at the peripheral bright locations of the cross section did not show much variation and increased substantially from mid radius location to the centre in the case of the outer pin. The diameter of the fission gas depleted central region as revealed from the radiograph is more than that of the grain growth region. The radial location[2,3] at the beginning of the grain growth is taken as temperature marker and the central temperatures are estimated. The estimated central temperatures for central, intermediate and outer fuel pins of the bundle are 1150, 1320 and 1600ºC respectively.

Porosity:

In the central pin, no significant change in porosity distribution is observed. The porosity increase occurs at mid-radial location in the case of outer pin and close to the center in the case of the intermediate pin. The observed porosity increase shows correlation to that of the fission gas depleted region in the radiograph and that of the dark porous region observed in the microstructure. The percentage of the number of rounded and interconnected pores is shown in the Fig 6. The number of rounded and interconnected pores shows an increase first and then reduces along the radius towards the center and small pores show a reverse trend. The reduction in number of pores at the center is due to the interconnectivity of the pores as revealed by the microstructure. The interconnected pores at the central region lead to the release of fission gas.

Crack Area:

In PHWR at 15000 MWD/Te burn up the clad creeps to the fuel and leaves no gap at operating temperature. During cooling the area generated in the form of cracks and pellet-clad gap depends mainly to the contraction from the operating temperature to the ambient temperature. The crack area (including pellet-clad gap) depends on the operating temperature of the fuel.

Fission Product Migration:

The β-γ autoradiographs on the samples reveal a depletion of radioactivity at the central region of the pin section. The fuel pins examined have been cooled for long time. Hence cesium, being the predominant and volatile radioactive nuclide in the fuel, migrates from center to the periphery due to the temperature gradient. The extent of the depleted radial central zone gives a direct indication of fission gas release and central temperature.

Fission Gas Release Mechanism:

The area of the dark porous region in the microstructure, central grain growth region and the β-γ depleted region are indicate the amount of fission gas release and the central temperature of the fuel pin. The microstructural observation infers that due to the higher mobility of fission gas atoms in the thermally activated central hot region of the fuel section, the atoms migrate, get entrapped at the grain boundary and subsequently coalesce to form bubbles. These fission gas bubbles grow by further accumulation of the gas atoms to bigger size, get interconnected, form channels and finally the gas gets released from the fuel matrix.

The linear heat rating and estimated central temperature are more in the outer pin than in the intermediate and central pin in that order. The relative difference in the area of central zone of the central, intermediate and outer pins are due to the operating temperature caused by the difference in the linear heat rating of the pins.

Remedial Measures:

In PHWRs fuel pins fission gas release can be reduced either by increasing the diffusion distance or by reducing the diffusion rate[5] of fission gas atoms. Increasing the grain size in the fuel can increase the diffusion distance. The diffusion rate depends on temperature. Increasing the thermal conductivity of the fuel and decreasing linear rating can reduce the fuel central temperature and thus diffusion rate.

The results revealed that the use of current design of 19 element for PHWR fuel bundle cannot be used as such for increasing the discharge burnup without carrying out design change for reducing the fission gas release of the outer fuel elements. Changing from 19 to 22 element bundle can decrease the linear power rating and central temperature in the outer pins. Equalising heat rating by enriching the central pins with Pu can help in reducing the fission gas release of the outer pins.

Conclusions

1. The metallographic examination of fuel sections of high burn up fuel pins revealed two distinctly different zones viz. the central dark and the peripheral bright region. Micro-examination showed the presence of pores at the grain boundaries in the central dark region and as fabricated microstructure at the peripheral region.

2. The sizes of the central dark porous region, grain growth region and the fission product depleted region are related to the heat rating and centre temperature of the fuel pins. The size is largest in outer pin and smallest in the central pin. The estimated temperature at the center of the fuel pins are 1150, 1320 and 1600ºC for central, intermediate and outer pins of the bundle respectively.

3. At the thermally activated central region the fission gas atoms migrate to the grain boundary and coalesce to form gas bubbles. These bubbles grow, interconnect and finally get released through the grain boundary channels.

4. The results revealed that the use of current design of 19 element for PHWR fuel bundle cannot be used as such for increasing the discharge burnup without carrying out design change for reducing the fission gas release of the outer fuel elements. Changing from 19 to 22 element bundle can decrease the linear power rating and central temperature of the outer pins, which is the main concern. Equalising burn up by enriching the central and intermediate pins with Pu can help in reducing the linear heat rating of the outer pins and fission gas release.

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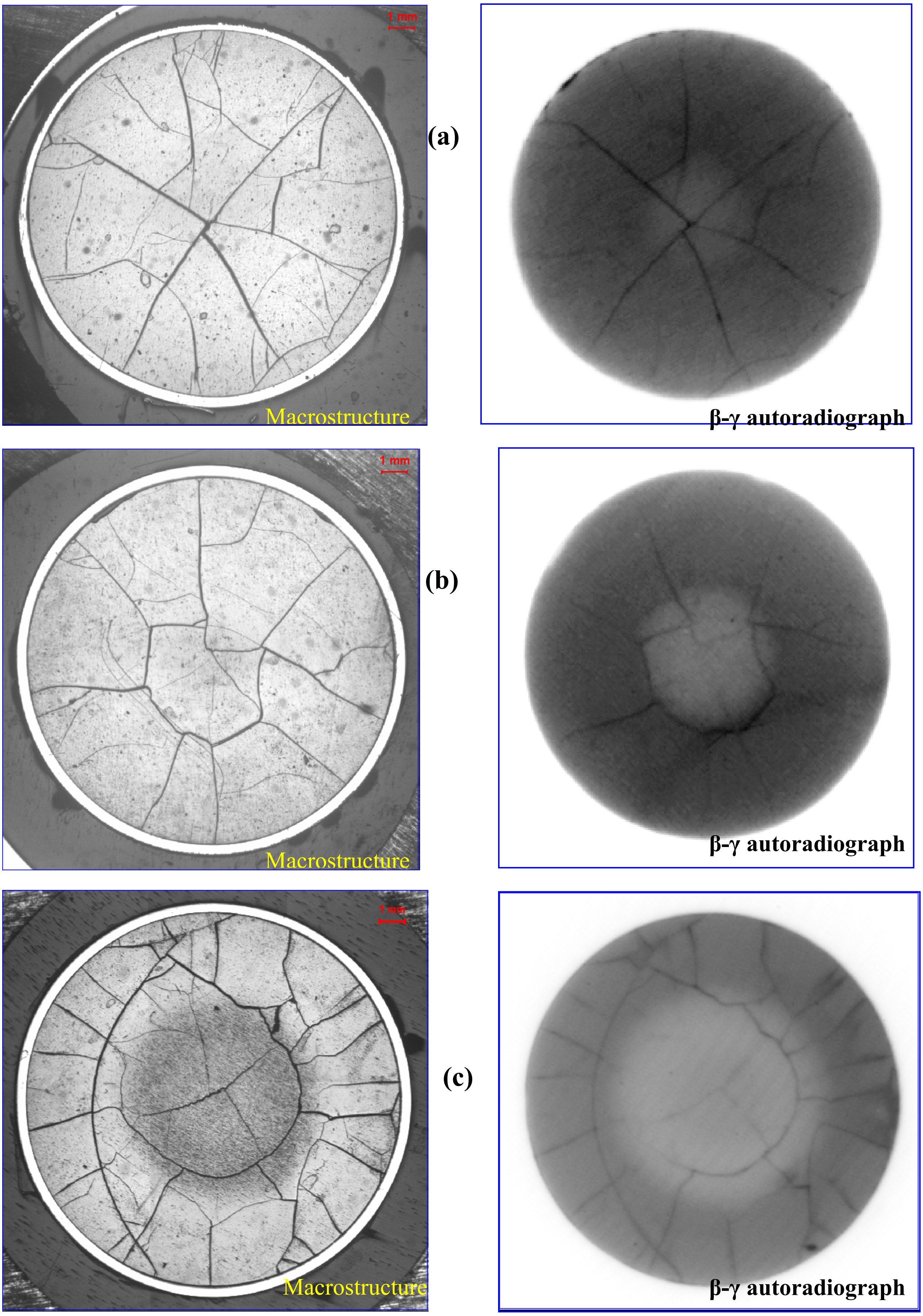

Fig. 1 Fuel cross-section and β-γ autoradiograph of (a) central pin (b) intermediate pin (c) outer pin showing the dark central region of the optical micrographs and corresponding light region in the β-γ autoradiographs.

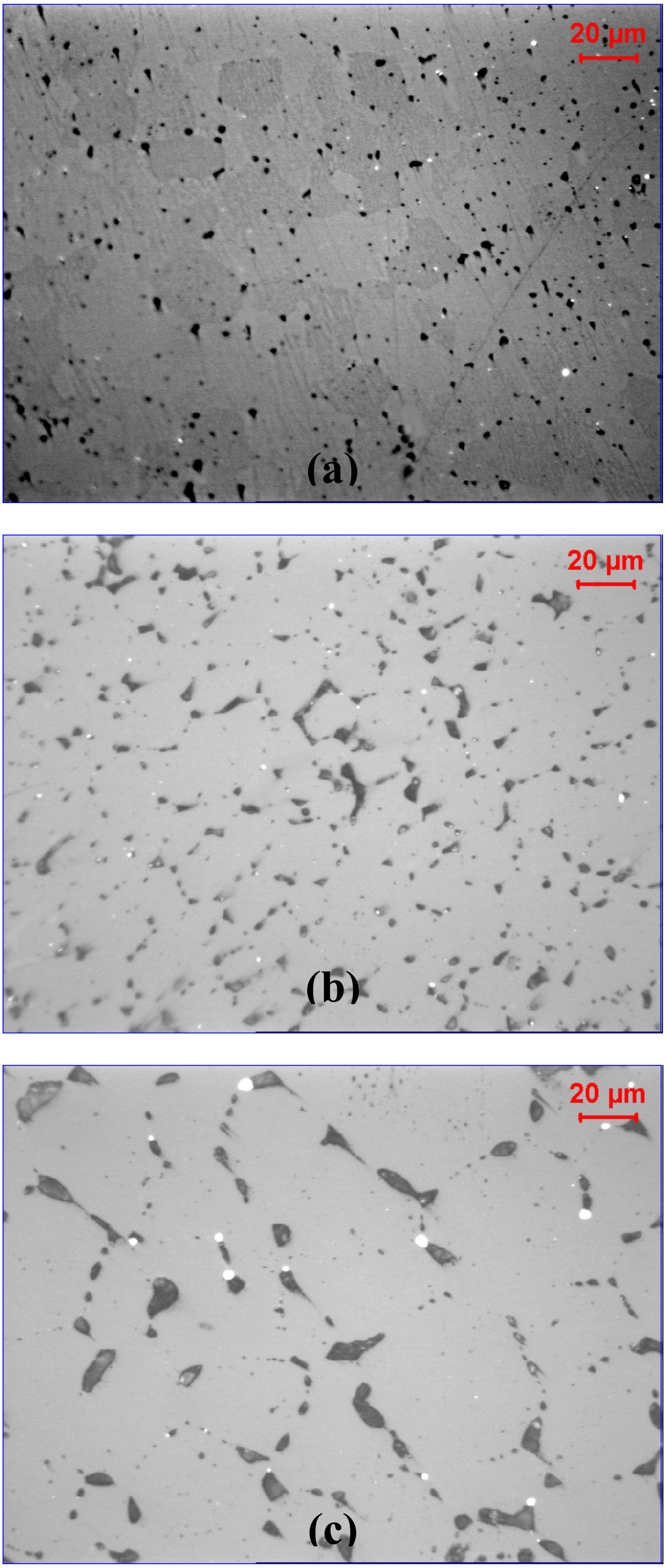

Fig. 2 Microstructure at the center of the fuel of (a) central (b) intermediate and (c) outer Pin

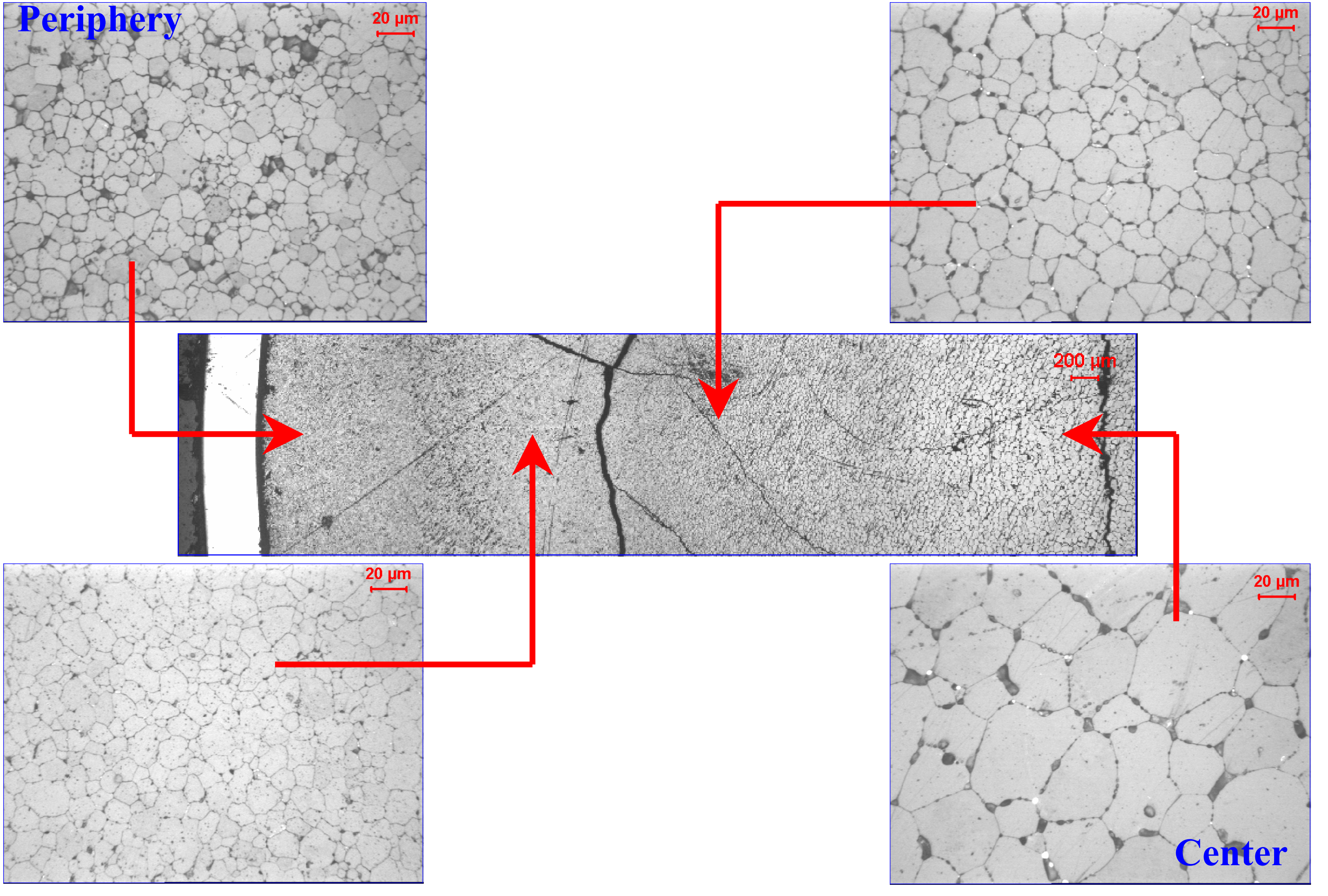

Fig. 3 Different microstructures observed from center to periphery of outer pin

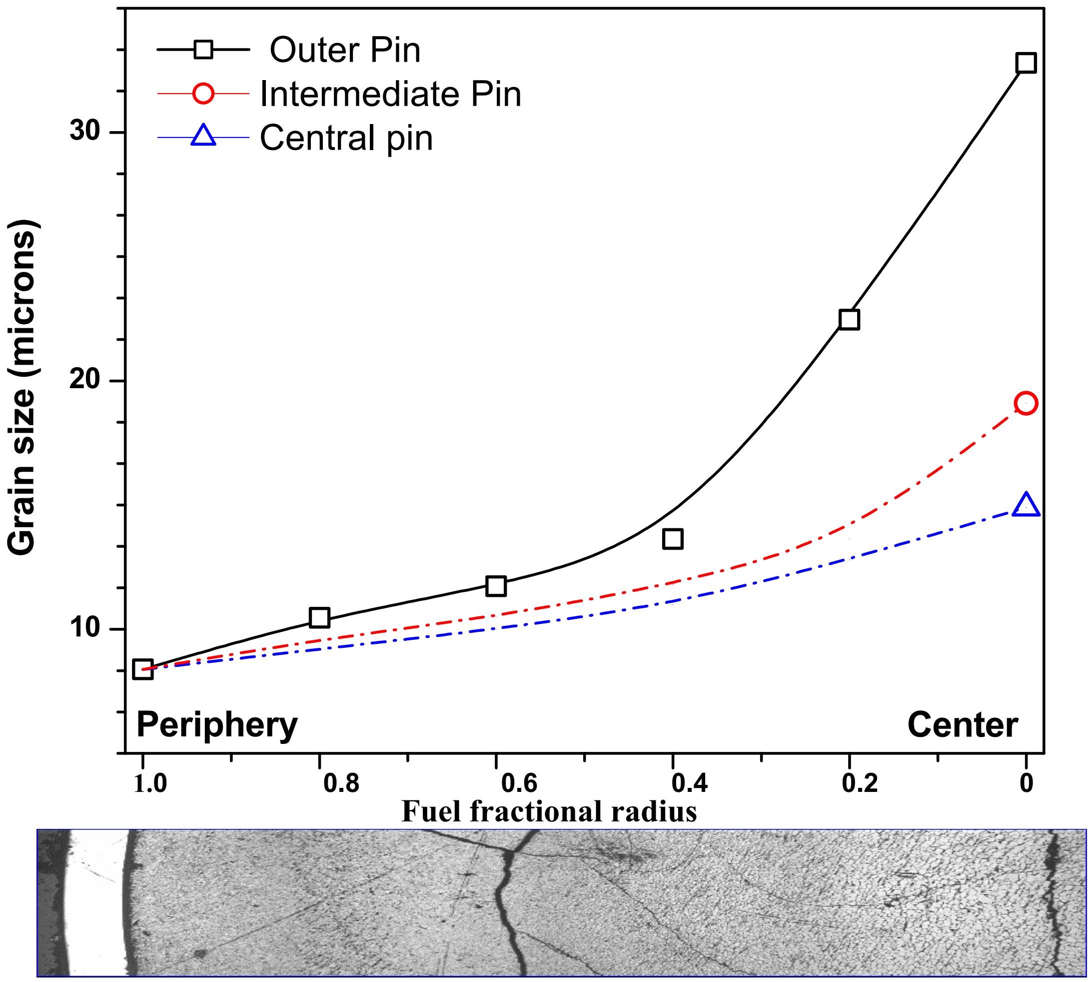

Fig. 4 Variation in grain size from periphery to center of the outer pin

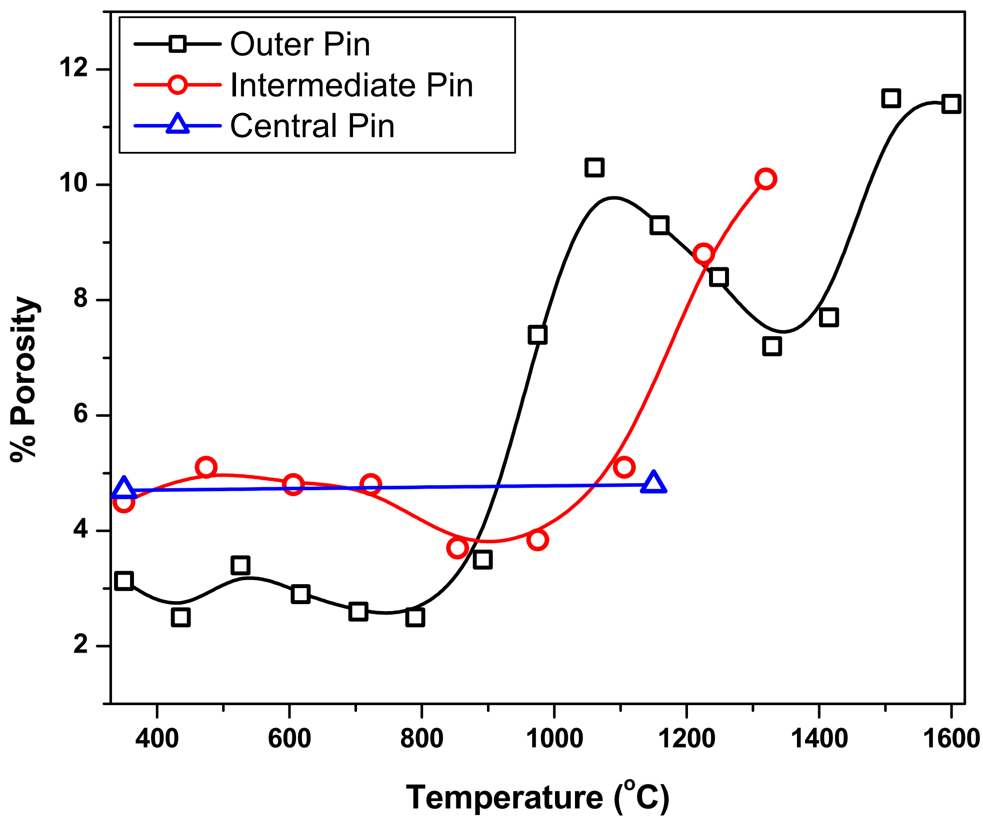

Fig. 5 Variation in porosity with temperature

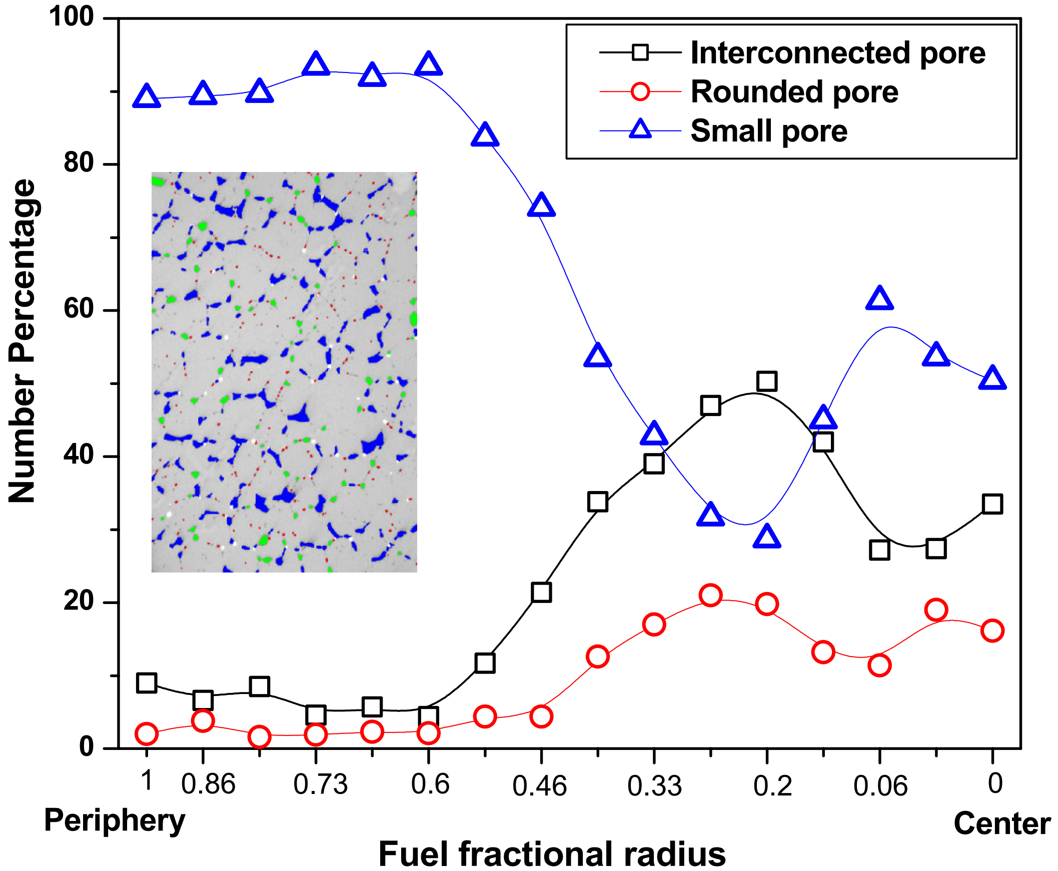

Fig. 6 Variation in different types of pores from periphery to center of the outer pin

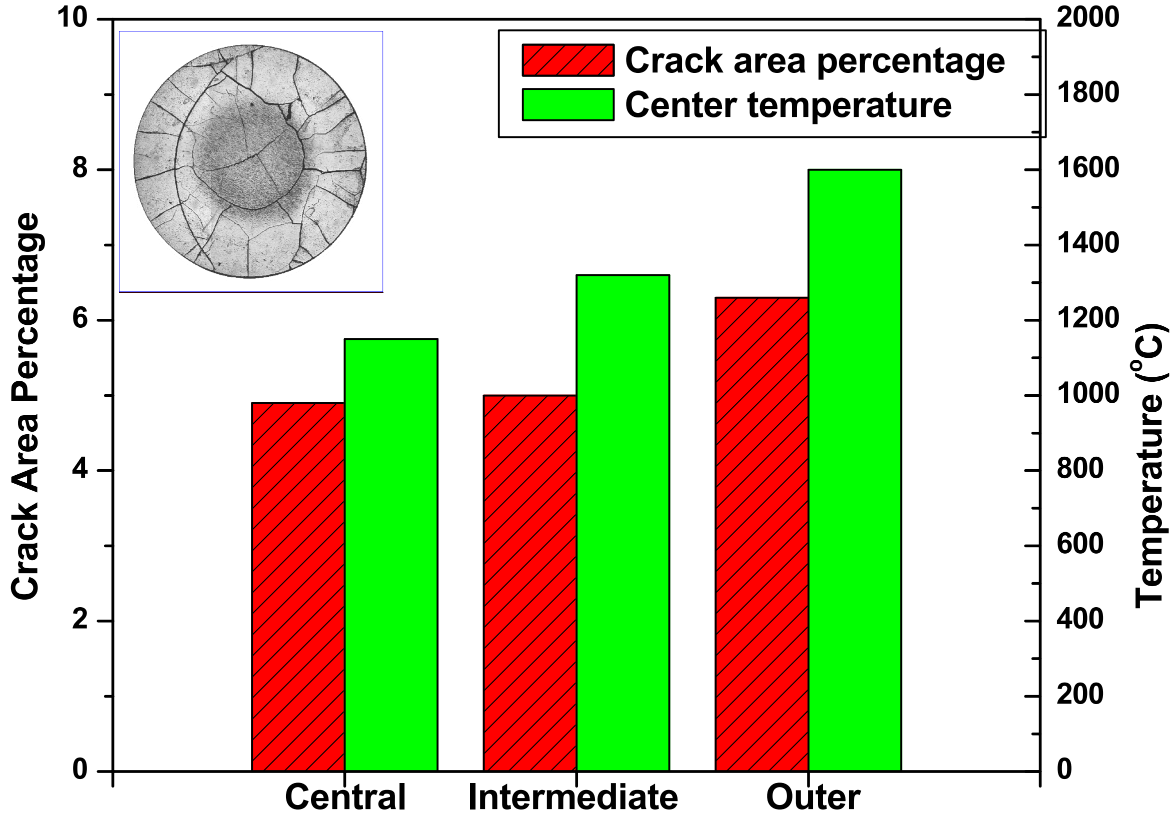

Fig. 7 Crack area percentage and estimated temperature in the outer, Intermediate and central pin