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Letter to the Editors

# Observation of a high burnup rim-type structure in an advanced plutonium–uranium carbide fuel

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**Abstract**

The observation is reported of a 'rim-type' structure with small subgrains in an advanced plutonium–uranium carbide $(U_{0.8}Pu_{0.2})C$ fuel pin, which had been irradiated in the Dounreay Fast Reactor to a burnup of 8.3% FIMA. © 1997 Elsevier Science B.V.

It has been known for some time [1–3] that the rim regions of high burnup ($\geq 45$ GWd/tM cross section average) LWR $UO_2$ fuel pellets, where the local burnup can be very high due to neutron resonance capture of U-238 causing locally increased concentrations of fissile Pu-239, show a structure quite different from that observed in the as-fabricated state, or at pellet radial positions with lower burnup values. This structure is characterised by transformation of the initial large-grained (typically 10 μm) $UO_2$ sintered structure into a 'cauliflower-type' microstructure resulting from grain subdivision, to produce subgrains with typical grain sizes of 0.1 to 0.3 μm. This transformation is usually referred to as the 'rim effect', and is normally only seen to a marked extent in the first 150 to 200 μm depth from the pellet surface, though it can extend much deeper into the $UO_2$ pellets at high burnup (e.g. 1.6 mm depth at 74 GWd/tM cross sectional burn up [4]). The threshold burnup for relatively cold fuel (outer region of the fuel pellets) for this polygonization to occur is around 70 GWd/tM local burnup.

The grain subdivision (polygonization) can be observed by transmission electron microscopy of small fuel fragments extracted from the fuel rim, and by scanning electron microscopy of fractured fuel surfaces [4]. In the latter case the subgrain formation is often particularly clearly visible on the inner surfaces of pores or large fission gas bubbles, leading to micrographs such as that in Fig. 1. This is a scanning electron micrograph of the structure inside a pore from the first 200 μm depth of an LWR $UO_2$ fuel pellet with an average burnup of 74 GWd/tM, but a local burnup at the position examined estimated to be in the range of 150 to 200 GWd/tM. The main peak of the grain size distribution in this fuel rim region was found to be at 0.15 μm from such micrographs [4].

It was originally thought that this type of microstructure was unique to the rim region of high burnup LWR $UO_2$ fuels. This, however, is not the case because similar pore surface structures have been found in samples of advanced fast reactor fuel $(U_{0.8}Pu_{0.2})C$. Fig. 2 shows an example of such a structure observed on the inner surface of a large bubble and similar structures were imaged on large irregularly-shaped pores [9]. Energy dispersive X-ray analysis was performed locally on these regions, and excluded the possibility that the structure resulted from a reaction with fission products, since only the fuel components could be detected. Fig. 2 was recorded at an approximately mid-radial position of the fuel pin DFR 330-X3, e.g. [5], irradiated in the Dounreay Fast Reactor, to a constant radial burnup value of 8.3% FIMA. Since this pin used sodium bonding the fuel temperature was relatively low, estimated at $900 \pm 100$°C at the radial position corresponding to Fig. 2, and was constant during the irradiation. Similar structures were also observed in nitride fuels.

Fig. 3 shows the subgrain size histogram measured on subgrains revealed on pore faces at a mid-radial position on the (U, Pu)C sample. The average subgrain size and the

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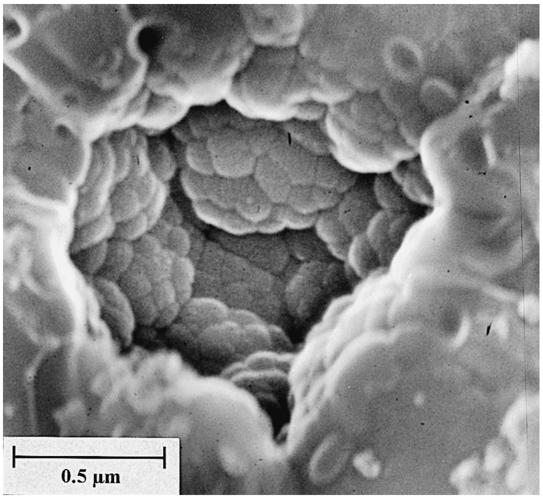


Fig. 1. An example of the sub-grain structure observed by SEM in the rim region of a high burnup $UO_2$ fuel pellet, revealed particularly clearly on the inner surface of a pore.

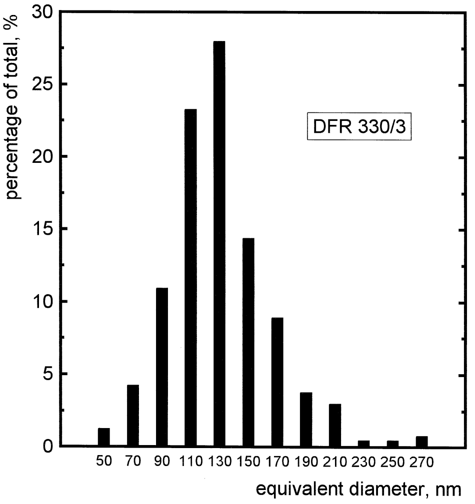


Fig. 3. A histogram showing the subgrain sizes, expressed in terms of equivalent diameters, based on the SEM images taken on the advanced fuel pin DFR 330-X3.

form of the distribution are very closely similar to those measured at the rim of high burnup LWR $UO_2$ fuel [4].

The relevance of this observation on advanced fuels to the study of the high burnup structure in LWR oxide fuels is that the processes responsible for grain subdivision in the fluorite structure of $UO_2$ must also be compatible with the sodium-chloride structure of the carbides and nitrides, or that they are structure independent. This is also indicated from ion-implantation experiments on different materials where polygonization was observed. These materials include MgO [6], $U_3Si$ [7] and spinel ($MgAl_2O_4$) [8]. A summary of the present state of understanding of the processes causing grain subdivision is given in Ref. [9].

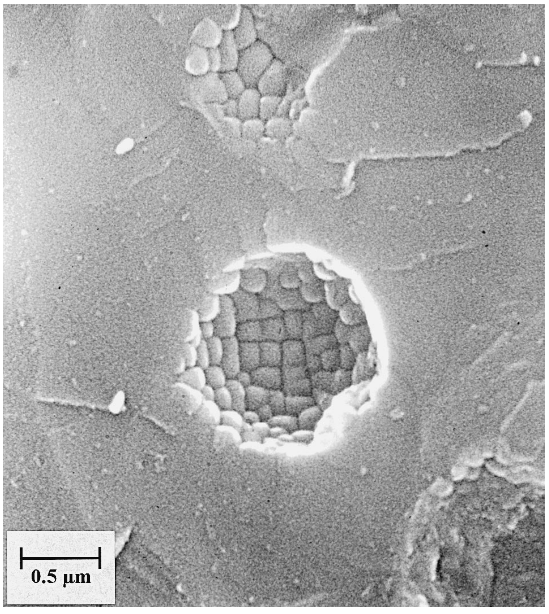


Fig. 2. An analagous structure to that shown in Fig. 1, but observed by SEM on the inner surface of a large fission gas bubble at a mid-radial position in the advanced $(Pu_{0.2}U_{0.8})C$ FBR fuel pin DFR 330-X3 irradiated to 8.3% FIMA burnup in the Dounreay Fast Reactor.

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