ARTICLE IN PRESS

Nuclear Engineering and Technology xxx (2018) 1–9

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Focused ion beam–scanning electron microscope examination of high burn-up UO_2 in the center of a pellet

J. Noirot*, I. Zacharie-Aubrun, T. Blay

CEA, DEN, DEC, SA3E, F-13108 Saint-Paul-lez-Durance, France

A R T I C L E I N F O

Article history:
Received 20 October 2017
Received in revised form
5 December 2017
Accepted 13 December 2017
Available online xxx

Keywords:
Electron BackScattered Diffraction
Fission Gas Bubbles
Fission Gas Release
Focused Ion Beam–Scanning Electron Microscope
Grain
High Burn-Up
UO_2

A B S T R A C T

Focused ion beam–scanning electron microscope and electron backscattered diffraction examinations were conducted in the center of a 73 GWd/t_U UO_2 fuel. They showed the formation of subdomains within the initial grains. The local crystal orientations in these domains were close to that of the original grain. Most of the fission gas bubbles were located on the boundaries. Their shapes were far from spherical and far from lenticular. No interlinked bubble network was found. These observations shed light on previous unexplained observations. They plead for a revision of the classical description of fission gas release mechanisms for the center of high burn-up UO_2. Yet, complementary detailed observations are needed to better understand the mechanisms involved.

1. Introduction

In general, intragranular and intergranular fission gas bubbles gradually build up in the center of UO_2 water reactor nuclear fuels. Examples can be found in the study by Stehle, Guedeney et al., and Itagaki et al.[1–3]. We will herein call this area the "central precipitation area", the porous area in the center of high burn-up fuels. Experiencing the highest temperatures during irradiation, this central precipitation area is the main contributor to fission gas release in free volumes of rods during base irradiation. Therefore, mechanistic modeling of fission gas behavior particularly involves a modeling of intragranular and intergranular fission gas bubbles in this central precipitation area [4–6].

In a previous article by Noirot et al. [7], in 2004, our study group showed how post irradiation examinations of high burn-up light water reactor UO_2 were used to provide detailed validation data for fuel behavior codes [6,8,9]. The examinations presented in this article focused on fission gas behavior. They included electron probe micro-analyzer, secondary ion mass spectrometer, and scanning electron microscope (SEM) measurements as well as annealing tests providing intergranular gas retention measurements. In particular, high burn-up polished UO_2 samples were examined using a PHILIPS XL30 SEM with a W filament electron gun and a Centaurus KE Developments (Bury St. Edmunds, Suffolk, United Kingdom) back scattered electron detector. The purpose of these SEM examinations was to deduce, from the same images, the bubbles at the surface of the examined fields and the grain boundary network. Using contrasts between the UO_2 grains, this grain boundary network was extracted from the images. These contrasts were due to differences in the electron channeling in the UO_2 crystal lattice, a function of lattice local orientation. The result of this was that it was then possible to obtain information on intergranular bubbles and on intragranular bubbles, in the same fields, using the same images.

However, article [7] also showed that this technique could not be applied in the central precipitation area, where this kind of information was most needed for fuel behavior code validation. Fig. 1 taken from [7] shows an SEM image taken at the external limit of the central precipitation area of a 61 GWd/t_U sample. This image shows, for this sample, a sharp transition around 0.54R (where R is the radius of the pellet, 0R corresponding to the center and 1R to the rim of the pellet). Beyond this limit, grain contrasts are visible; however, on the central side, where a high density of quasi micrometric bubbles had formed, the situation is quite unclear. Grains are partly visible, but there is no way to discern the difference between intergranular and intragranular bubbles. To produce

* Corresponding author.
E-mail address: jean.noirot@cea.fr (J. Noirot).

https://doi.org/10.1016/j.net.2017.12.002

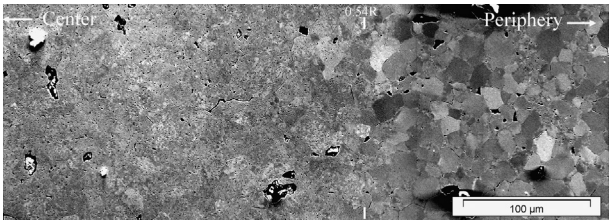

Fig. 1. Crystallographic contrast image at the limit of the central precipitation zone on a 61 GWd/t_U–irradiated UO_2, from the study by Noirot et al. [7].

the data needed by the modelers, a two-step process was then adopted, with image acquisitions for the bubble and pore image analyses, followed by chemical etching to reveal the positions of the grain boundaries and new image acquisitions in the same fields. About the change in the electron channeling, our study group wrote in this previous article [7] that "*This demonstrates the influence of the gas precipitation on the SEM crystallographic contrast and is to be precisely analyzed in a subsequent publication*", but we were not able to go much further than thinking of good reasons for this change, with no experimental evidence to support these ideas.

In the studies by Noirot et al. [10,11], in 2009 and 2008, respectively, among the results presented, there were SEM fractography images of a UO_2 sample irradiated at 73 GWd/t_U. In the center of this fuel, large bubbles were observed (Fig. 2). These bubbles were neither spherical (as typical intragranular bubbles) nor clearly lenticular (as typical intergranular bubbles before interconnection), and they did not seem to be widely interconnected. We then mentioned that it was not so easy to see the difference between intergranular and intragranular surfaces in these fractographs or to identify the grain boundaries.

In the classical representation of the fission gas release process, during normal fuel operation, in the hot center,

- intragranular gas diffusion leads to a buildup of fission gases accumulating in intergranular position.
- these fission gases form intergranular bubbles.
- these intergranular bubbles interconnect, eventually forming tunnel networks that are paths for the release of the fission gases to the free volumes of the rods. [13] (p.318), [14,15].

The absence of such clearly visible tunnels in the fuels presented in the studies by Noirot et al. [7,10,11], in spite of significant fission gas release, was a disturbing point in these observations.

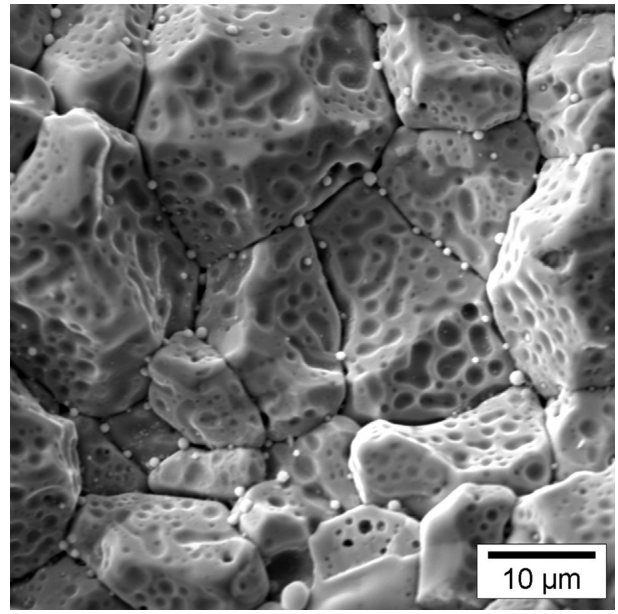

Fig. 3. SEM fractograph in the center of a 38.8 GWd/t_U pressurized water reactor UO_2 fuel after an unfailed ramp test. The maximum linear power, 520 W cm^{-1}, was held for 90 s, from the study by Noirot et al. [12]. SEM, scanning electron microscope.

In fact, in our own experience, such tunnel networks were clearly observed after ramp tests (Fig. 3) and after out-of-pile annealing tests [12,16,17]. In the literature, in addition to ramp tests and annealing tests [18–21], such tunnel networks were observed in experimental irradiations for which the temperature levels in the fuel were set to high levels [22].

In the fuels presented in the studies by Noirot et al. [7,10,11], the burn-ups were high, but the rod average linear powers never reached values higher than 203 W cm^{-1}, and the fuel behavior code calculations showed that the fuel centerline temperatures were in the range of 800–900°C during the last three cycles for the rods examined at 83 GWd/t_U, after seven cycles of irradiation. Nonetheless, in spite of moderate central temperatures and in spite of no obvious intergranular bubble interlinkage, fission gas release rates clearly increased at high burn-up.

In 2016, we replaced our W filament electron gun PHILIPS XL30 SEM with a field emission electron gun Focused ion beam–scanning electron microscope (FIB/SEM) with improved

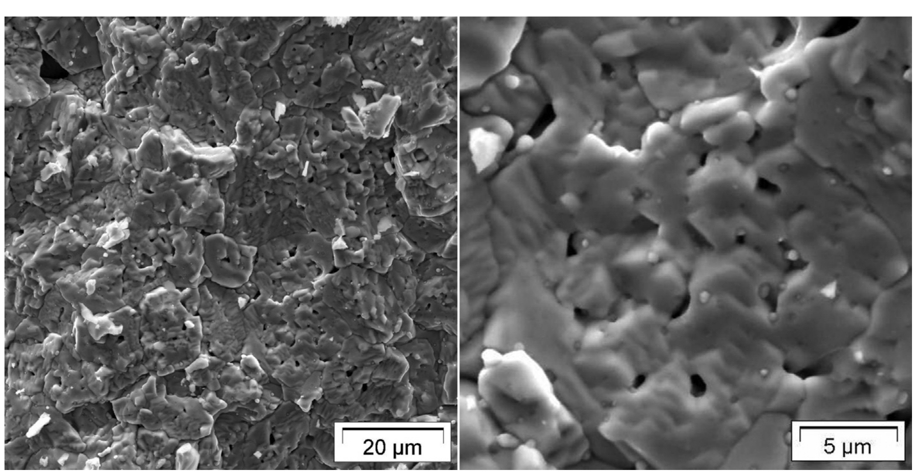

Fig. 2. SEM fractograph of a 73 GWd/t_U UO_2 sample at the center of the pellet, from the studies by Noirot et al. [10,11]. SEM, scanning electron microscope.

Please cite this article in press as: J. Noirot, et al., Focused ion beam–scanning electron microscope examination of high burn-up UO_2 in the center of a pellet, Nuclear Engineering and Technology (2018), https://doi.org/10.1016/j.net.2017.12.002