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Letter to the Editor

Lattice contraction in the rim zone as controlled by recrystallization: Additional evidence

J. Spino *, D. Papaioannou

European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, D-76125 Karlsruhe, Germany

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Abstract

Additional micro X-ray diffraction measurements of the lattice constant are described in the rim zone of a BR3-fuel irradiated under LWR-fuel conditions up to an average burn-up of 68.5 GWd/tM. Though signs of the so-called rim-transformation, with peripheral local burn-up enhancement, porosity increase and Xe-depletion, were found in the fuel, the traditionally accompanying lattice contraction verified in standard high burn-up LWR-fuels, was not detected. The absence of this feature is attributed to the lack of pellet-edge recrystallization in the present case, which likely impeded the release of accumulate strains.

During service in the reactor and furthermore after discharge, the lattice spacing of the UO_2-matrix of LWR-fuels is modified by a number of processes. The main chemical effects associated with burn-up, namely dissolution of fission products, dissolution of Pu and incorporation of excess-oxygen in case of Zr-cladding- and Mo-oxidation saturation [1], cause the fuel lattice to contract. On the other hand, point and finite-size-defects induced by radiation- and α-decay damage, the latter occurring mainly during storage, cause the fuel lattice to expand. These lattice expansion effects, which are burn-up and decay-time dependent and reach saturation, generally override the lattice contraction effects. As a result, non-annealed irradiated fuels examined at room temperature usually show a net lattice expansion, which compared to non-irradiated UO_2 can reach a fraction of up to 0.2% [2].

Sections of non-annealed base-irradiated LWR-UO_2-fuels with extraction burn-ups below about 40 GWd/tM, for which a constant radial burn-up profile is expected due to still negligible rim-effect, do not show, however, a constant lattice parameter profile throughout the radius, but decaying values towards the pellet centre [3]. This drop towards the centre is attributed to the thermal healing of irradiation defects by exposure of the fuel to temperatures superior to ≅800 °C in the reactor [2]. Even in the mentioned decreased region, the lattice parameter of the fuels at room temperature normally exceeds the value of non-irradiated UO_2 (547 pm), or that correspondingly corrected for incorporation of fission products and Pu at the given burn-up [2,3]. This difference reflects primarily the accumulated α-decay damage during storage, as well as the eventual remaining neutron-damage after the in-pile healing.

In a previous work by the authors describing first micro X-ray diffraction studies of high burn-up LWR-fuels [2], it has been shown that for fuels with average burn-ups beyond 60 GWd/tM, which suffered the so-called rim- or HBS (high burn-up structure)-transformation, an additional lattice contraction took place at the pellet periphery, which followed approximately the slopes of the porosity increase and the Xe-depletion profiles. For such fuels, a flat maximum of the lattice constant was verified at the onset of the rim zone [2]. Further measurements reported in [4] and analysed in the accompanying paper of Ref. [1] have shown that the lattice parameter values at very high average burn-ups (≈100 GWd/tM) might be affected by fuel oxidation. The shape of the lattice parameter profile, especially the

* Corresponding author. Tel.: +49 7247 951 233; fax: +49 7247 951 590.
E-mail address: jose.spino@itu.fzk.de (J. Spino).

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decrease of it in the rim zone for burn-ups above the threshold, may persist independently of the oxidation state of the fuel [1].

The slope of the referred to lattice contraction in the rim zone per unit burn-up, after surpassing the threshold local burn-up of ≈70 GWd/tM, was shown to be much higher than that expected from the local increase of burn-up [2]. For this reason, the behaviour was attributed principally to the release of lattice strains during the rim-transformation. The relaxation mechanism addressed was intersection of dislocation loops with the walls of the dislocation cells formed in the rim zone (recrystallization) [2]. A similar explanation, based in the X-ray scattering theory of Krivoglaz [5] for crystals with finite-size-defects, has been supplied for the lattice expansion and contraction effects observed in He-implanted metals [6,7]. In agreement, models of the lattice parameter changes during recrystallization of high burn-up UO_2 and U–Mo fuels recently developed by Rest [8,9], also addressing dislocation loops/dislocation cell interaction processes, satisfactorily match the measured data. However, an experimental evidence of unequivocal correspondence between lattice contraction and recrystallization in UO_2 fuels was not available. This is provided by measurements of the present communication.

Figs. 1 and 2 show the variation of the lattice parameter, local burn-up and porosity as a function of the radial position in the rim zone, for two UO_2 fuels with similar discharge average burn-ups, respectively, 67 and 68.5 GWd/tM, though with basically differing grain structures. The data of Fig. 1, already published in [2] and given here just for comparison, correspond to a standard LWR-fuel with well developed rim-structure. Typical power history and irradiation temperatures were provided in [10]. Fig. 2, in turn, corresponds to a BR3-fuel irradiated under LWR-fuel conditions, which, having more than twice as larger initial grain size and ^{235}U-enrichment as the former one, developed just an incipient rim-structure, with still persisting initial grain structure almost up to the interface with

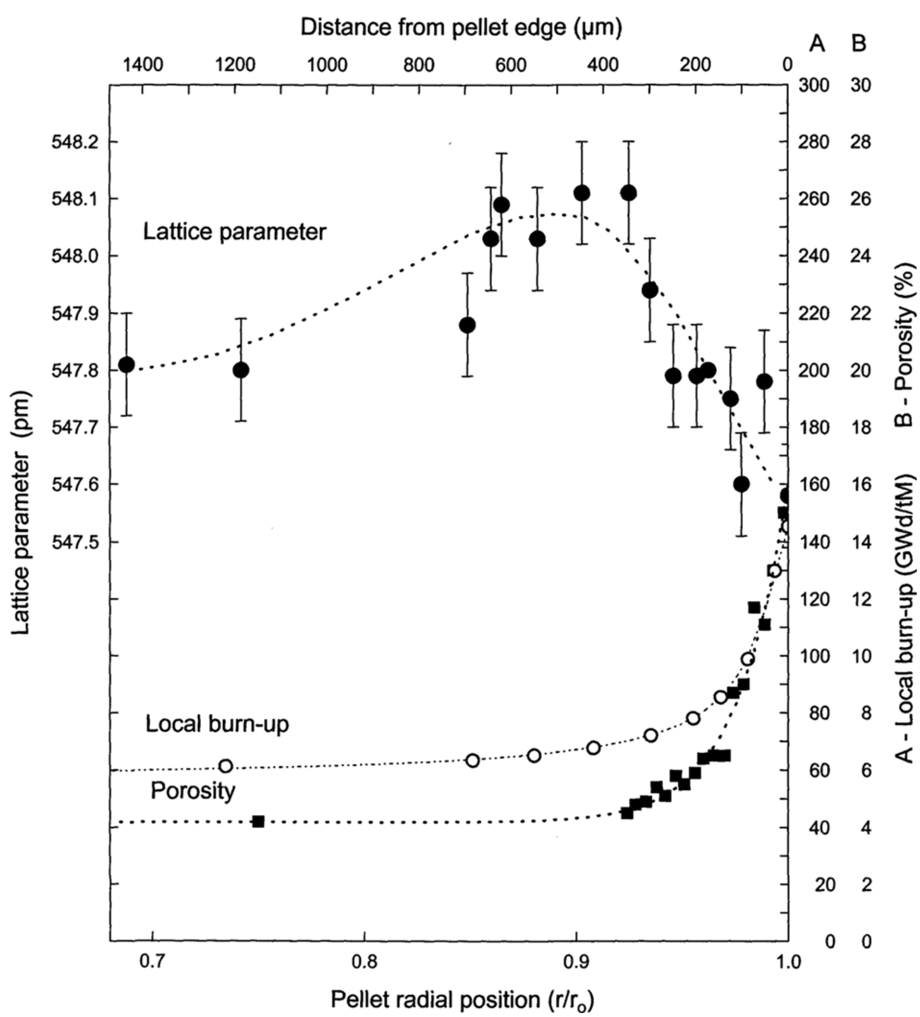

Fig. 1. Radial lattice parameter, local burn-up and porosity profiles of a standard LWR-fuel with 67 GWd/tM average burn-up. Initial ^{235}U-enrichment: 4.2 at.%. Initial fuel-matrix grain size: 5–10 µm. Local burn-up calculated with Appolo 2-Code. Typical power ratings and central temperatures [10]: at average burn-up ≈35 GWd/tM, $\lambda \approx 275$ W/cm, $T_c \approx 1150$ °C; at average burn-up ≅60–65 GWd/tM, $\lambda \approx 175$ W/cm, $T_c \approx 920$ °C. Original lattice parameter, porosity and burn-up data from Ref. [2].

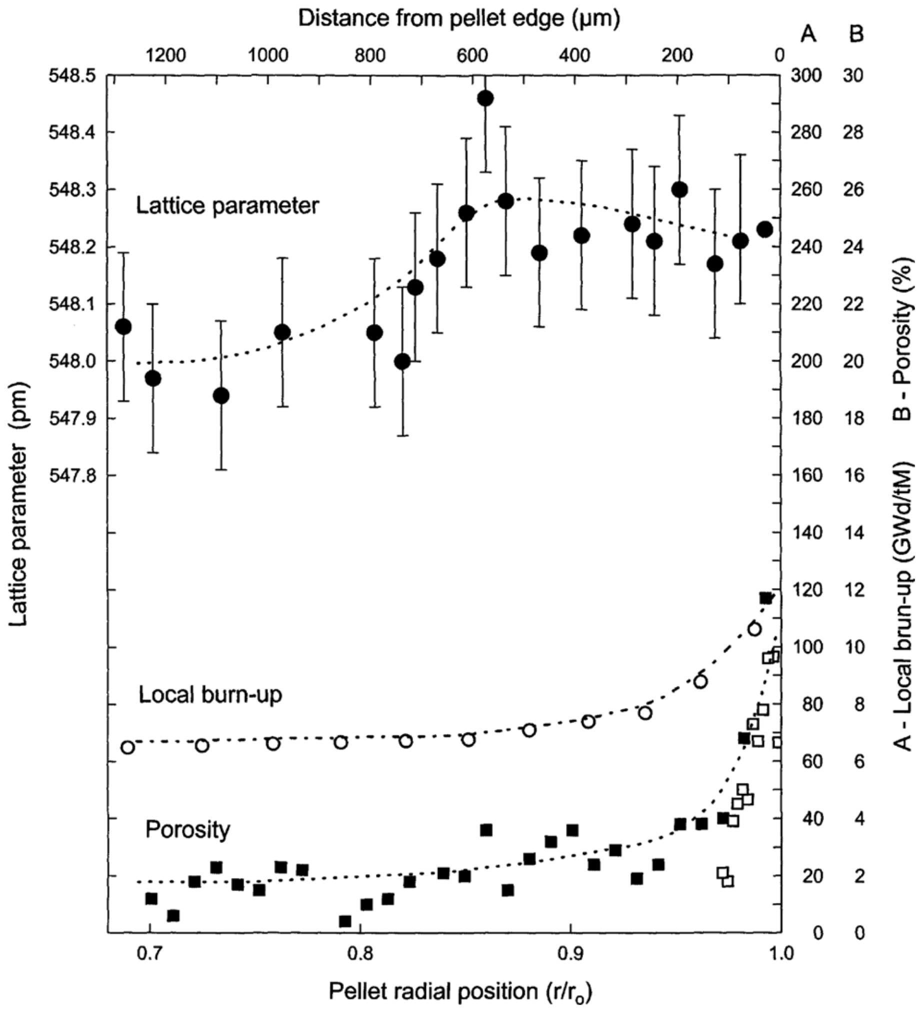

Fig. 2. Radial lattice parameter, local burn-up and porosity profiles of a BR3-fuel with 68.5 GWd/tM average burn-up, irradiated under LWR conditions. Initial ^{235}U-enrichment: 8.65 at.%. Initial fuel-matrix grain size: 20 μm. Local burn-up calculated with Appolo-2-Code. Typical power ratings and central temperatures [11]: at average burn-up ≈35 GWd/tM, $\lambda \approx 310$ W/cm, $T_c \approx 1320$ °C; at average burn-up ≅55–68 GWd/tM, $\lambda \approx 195$ W/cm, $T_c \approx 1050$ °C. Original porosity and burn-up data from Ref. [11].

the cladding. Characterization of this special fuel was provided in [11]. The here reported lattice parameter measurements of the BR3-fuel sample were performed by micro XRD-diffraction with the same method as described in [2]. The experimental error, according to examinations of Si, Au and UO_2 standard specimens, was around ±0.01 pm [2]. Though less pronounced for the BR3-fuel, Figs. 1 and 2 show similar increasing trends in the local burn-up and porosity profiles for both kinds of fuels. In turn, the previously described lattice contraction in the rim zone appears remarkably diminished in the case of the BR3-fuel (Fig. 2).

The above situation can be further visualized in Fig. 3, where the lattice parameter of the two fuels of Figs. 1 and 2 are plotted for the outer pellet zones ($r/r_o > 0.9$) as a function of the local burn-up, in correspondence with the respective microstructures. The figure also shows the lattice contraction slope of UO_2 due to the dissolution of fission products, as measured for simulated fuels with nominally stoichiometric oxygen composition (≈−0.13 pm per 10 GWd/tM) [1,3]. The BR3-fuel evidences just a slight decrease of the lattice spacing in the rim zone, which is visibly smaller than the expected change due to dissolved fission products (Fig. 3, upper curve). The incipient recrystallization of this fuel, only scarcely detected at the outermost radial positions ($r/r_o > 0.99$) (Fig. 3, upper micrographs), had no influence on the lattice parameter. In turn, the variations in the equivalent LWR-fuel markedly surpass the lattice contraction effects assigned to burn-up. This is particularly noticeable for the narrow burn-up range ≈70–85 GWd/tM, in which for this fuel the recrystallization was almost completed (Fig. 3, bottom curve and micrographs).

As for the above evidences, it seems that only under conditions of pronounced recrystallization, i.e., under considerable loss of the original grain boundaries, a measurable lattice contraction would be recognizable in the rim zone of LWR-fuels in addition to that caused by the dissolution of fission products. Incipient recrystallization, as observed in the examined BR3-fuel, would not have an impact on

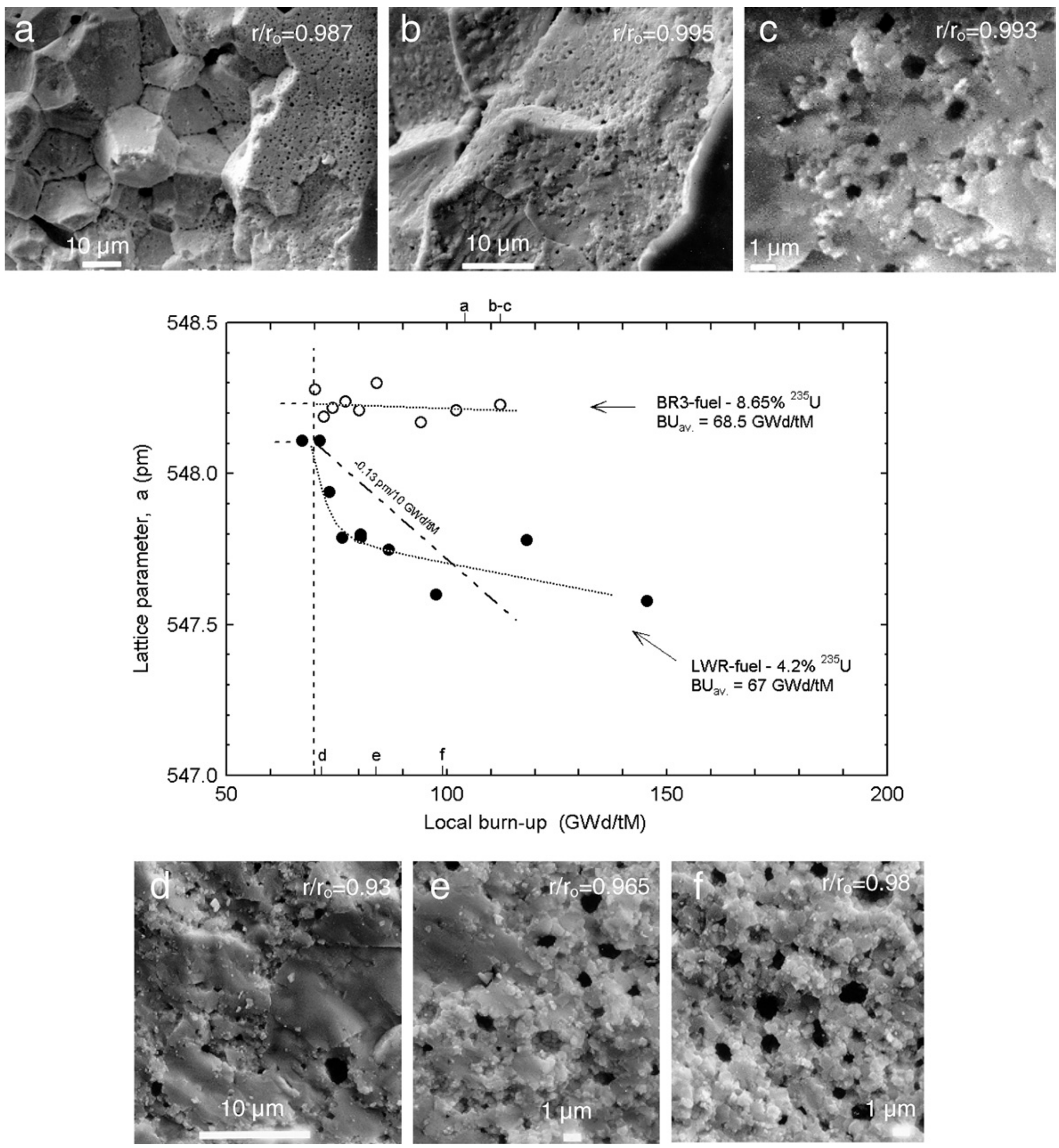

Fig. 3. Comparison of the lattice parameter versus local burn-up data of the fuels of Figs. 1 and 2, with corresponding microstructure features. Dotted line: expected lattice contraction slope of nominally stoichiometric fuels due to the local increase of burn-up (fission products concentration) (−0.13 pm per 10 GWd/tM [1,3]).

the lattice parameter. Why in the last case the measured lattice spacing in the rim zone did not fully respond to the expected decrease due to the local variations of burn-up (fission products concentration), is not completely clear.

A possible explanation refers to the higher power ratings and centreline temperatures of the BR3-fuel compared to standard LWR-fuels (values provided in captions of Figs. 1 and 2). As the irradiation temperatures are increased, the oxygen getter action of the Zr-cladding, and so the likelihood of the fuel to acquire hypostoichiometric O/M ratios at the pellet edge, are increased [12]. This effect could have caused flattening of the 'lattice parameter vs. burn-up' curve in the rim zone of the BR3-fuel, as it is observed in annealed irradiated and simulated fuels when the oxygen potential is decreased [1]. ΔGO_2-measurements of the BR3-fuel are unfortunately not available.

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