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Comments on the threshold porosity for fission gas release in high burn-up fuels

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Abstract

This letter challenges two recent papers in this journal, suggesting that the high burn-up structure of LWR-fuels would evolve towards an open pore system, facilitating gas release. In contrast, recent experimental results and supporting calculations reviewed here as well as new evidence from a 3D pore-reconstruction strongly suggest that the materials in question would show closed porosity conditions and hence reduced probability of gas release, at least up to porosity fractions of about 25%. This value is most likely conservative.

In the strategy for extending the burn-up in LWR fuels, a matter of concern remains the release of fission gases from the rim zone, as the released fraction increases just at the burn-up where the rim structure appears (about 40 GWd/t M pellet average) [1]. However, the more thoroughly the rim material and related processes are characterized, the less probable seems a high fission gas release (FGR) from this part of the fuel. Support for this are the ostensible closed character of the rim porosity [2] and the predominance of thermally released gases (i.e. of non-rim origin) in the rod free-volume [1]. In spite of this, two recent publications in this Journal [3,4] postulate the rim material as evolving towards an open system. According to [3] this transition will happen steadily as a linear function of the rim width, and according to [4], for values of porosity $\geqslant 24\%$, as a power function of the local porosity. However, these results do not correspond to the current experimental evidence.

In [3], the porosity increase and the grain subdivision of the rim zone are considered per se to result in an enlarged free surface area of the system, and as a consequence in an enhanced athermal gas release (via recoil and knockout). However, fine-grained and porous materials do not always exhibit large open areas. For example, closed cell solid foams, a class of materials to which the fuel rim would belong [2], can sustain considerable pore fractions (>80%) under sealed conditions [5]. Therefore, the surface-to-volume ratio (S/V) increase postulated in Ref. [3], with a slope of 75 000 cm^{-1} per mm of rim width as well as the assumed maximum S/V value at the pellet edge ($\simeq 5000\ cm^{-1}$), require experimental proof. For comparison, measured S/V values for UO_2 range between <500 cm^{-1} for as-sintered pellets with 95% density [6] and $\leqslant 2000\ cm^{-1}$ for highly irradiated polycrystals (about 70 GWd/t M burn-up) [7].

Linked to the conclusions in [3], it is stated in [4] that the fuel rim will show extensive pore channelling and boosted gas release after exceeding 24% porosity. This comes from a Monte Carlo calculation, in which the rim structure is simulated by distributing variable amounts of 1 μm (or larger) hole-cubes (pores) in a finite mesh of 0.5 μm solid-cubes (grains) [4]. However, these results contradict our experimental observations and supporting calculations using Ronchi's percolation model for regular arrangement of pores [8], both indicating low

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pore-interconnection probability for the rim material up to high porosity fractions [2,9].

In fact, analysis of 'hardness vs. porosity' data shows that, in terms of the load-bearing area fraction, the rim material is best represented by a system of (quasi-spherical) pores in a continuous matrix (mimic of foam materials) [2]. For such a system, the critical porosity for the open pore transition ranges between about 30% for randomly distributed pores [10] and about 52% for cubic-packed pores [11]. In agreement, calculations applying the cited percolation model of Ronchi for the rim configuration (where the grain-to-pore size ratio is 0.2–0.3) indicate pore-interlinkage probabilities well below the percolation threshold even for porosity fractions of 50% or larger [2,9].

On the other hand, the hardness analysis for the non-rim fuel material showed this as being best represented by a system of intersecting solid particles (mimic of sintered solids) [2]. This system can exhibit the open pore transition as early as at about 3% porosity [2,10,11]. Coincident with this, the application of Ronchi's model for the as-sintered fuel conditions (the grain-to-pore size ratio is 5–7) showed pore-interlinkage probabilities above the percolation threshold already at porosity fractions below 3% [2,8,9].

Compared to these results, the calculations in [4] appear to underestimate the open pore transition for the rim case ($P_{\text{open-rim}} \simeq 24\%$, grain-to-pore size ratio = 0.1–0.5 [4]) and to overestimate this for the as-sintered fuel case ($P_{\text{open-as-fabricated}} \simeq 12\%$, grain-to-pore size ratio = 10 [4]). A general explanation for these differences cannot be given at this stage. However, for the rim material, it appears that in [4] a too large grain size was assumed (0.5 μm). By assuming smaller grain sizes closer to the real rim (0.2–0.3 μm), thus allowing for a larger (numerical) pore-wall thinning at constant rim volume, it is possible that a larger porosity fraction before pore-contact would have been obtained.

As further support for the findings in [2,9] we present here new results of a 3D-reconstruction of the rim and the pellet-centre pore systems of a LWR-fuel with about 100 GWd/t M average burn-up, respectively, in Figs. 1 and 2. The reconstruction was done from a series of 14 optical-micrographs, taken after stepwise polishing with submicron suspension at about 0.2 μm-distance from each other. Stacking of the digitalized optical image planes and spatial interpolation of pore features with appropriate isosurfaces were performed using the PC-softwares Image-Pro® and 3D Constructor®.

For the rim case, it is shown that no relevant interconnecting paths between pores were present, despite the high measured porosity (22%). Only rare pore coalescence and few isolated pore-channels on apparently former cracks or grain boundaries were found (Fig. 1).

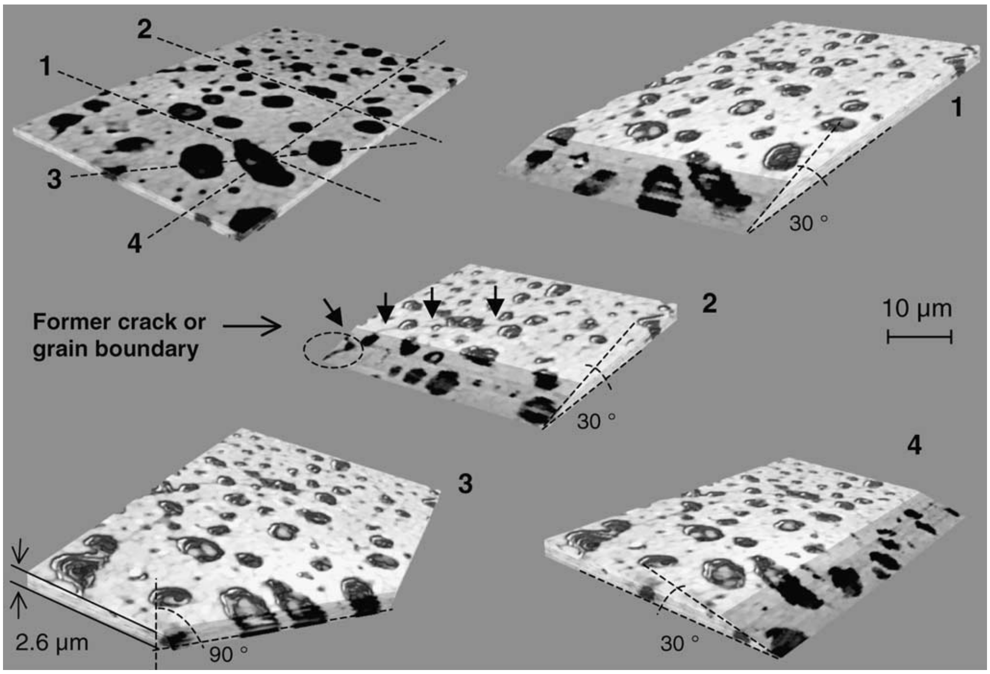

Fig. 1. 3D-reconstruction of the pore space at the pellet rim of an LWR fuel with 100 GWd/t M average burn-up showing a manifested lack of pore interconnection in oblique and perpendicular cuts. The isolated pore-channel in Section 2 lies apparently on a previous crack or grain boundary. The local porosity measured with Image-Pro® is 22%.

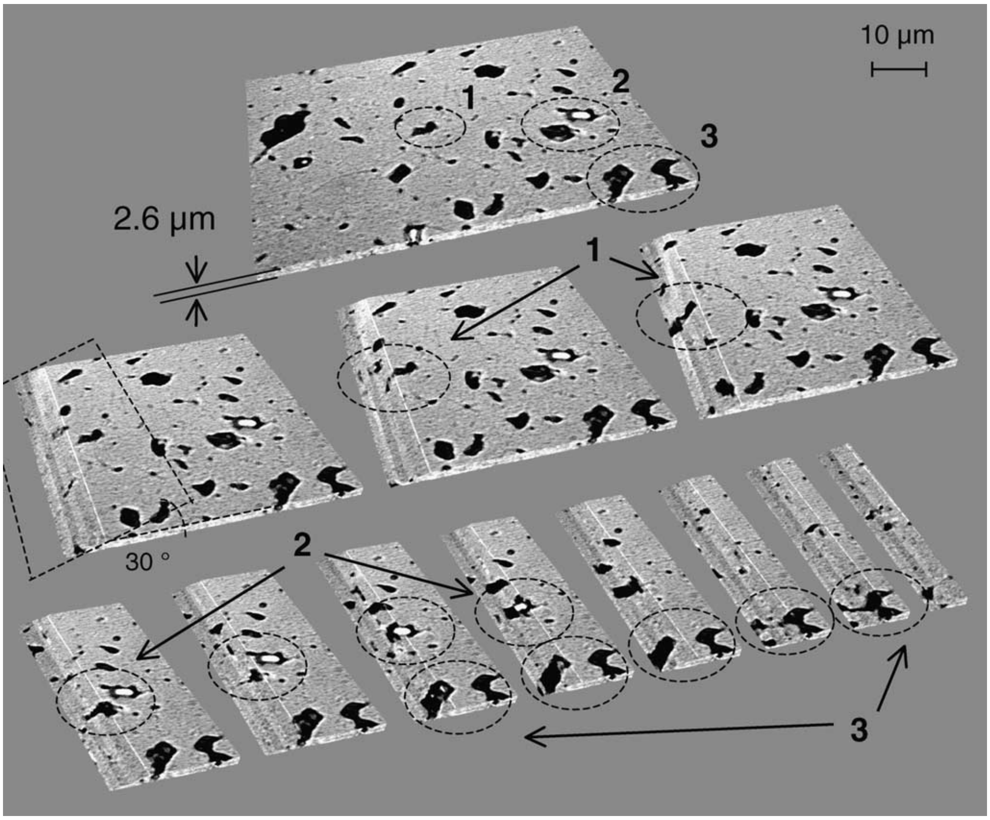

Fig. 2. 3D-reconstruction of the pore space at the pellet centre of a LWR fuel with 100 Gwd/t M average burn-up. Sequential oblique cuts reveal abundant pore channels in the region. The local porosity measured with Image-Pro® is 6.6%.

In contrast, rather abundant inter-pore channels could be recognized in the fuel centre case, despite the low measured porosity (6.6%). Three of these cases are shown in the cuts of Fig. 2.

Besides the specific section shown in Fig. 1, other sections examined in this fuel yielded a maximum porosity at the pellet edge of 25.3%. Again, at this porosity also the lack of pore interconnection was verified. Although we do not have data available of rim zones at higher porosity values, the trend suggested by the micrographs is that apart from some pore coalescence and enlargement (Fig. 1), boosted pore channelling would not occur immediately above $P = 25\%$. In line with this, similar high burn-up structures in U_6Fe [12] and U_3Si [13] dispersion fuels show an onset of bubble interlinkage (i.e. break-away swelling) at porosity fractions >30–35% [14].

Thus, for conditions below and probably beyond the presently characterized limits, the main sources of FGR would continue being the central and intermediate fuel regions (as-sintered microstructure), due to the higher operating temperatures and the more likely formation of pore channels. Nevertheless, FGR is not to be excluded from the high burn-up fuel regions, as observations suggest a variable decrease of the retained amounts of Xe and Kr relative to the theoretical inventory [1,15,16]. As we also mentioned in [9], a plausible reason for this gas escape would be the activation of release channels on the still present prior grain boundaries. Once the rim-structure is formed, and prior to the onset of extended pore interlinkage, the gas release from this region occurs most likely only by long-range diffusion of occluded gases.

As a conclusion, beside the exposed initial release and minor possible gas-loss by out-diffusion, it is proposed that the LWR fuel rim will maintain gas-tight conditions at least up to 250 GWd/t M local burn-up and 25% local porosity. These values are most likely conservative. Because of the undeniable impact on fuel performance, further measurements are needed to exactly determine these limits under the specific reactor situations.

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