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# On the rim effect in high burnup $UO_2$ LWR fuels

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Ion implantation experiments with energetic Xe-ions were performed in order to better understand the possible mechanisms leading to the formation of an outer shell of small grains and large porosity in high burnup $UO_2$ (the so-called "rim effect"). It is shown that grain subdivision related to the formation of highly pressurized (probably solid) Xe-precipitates occurs at implantation doses corresponding to fission product concentrations reached at 7 to 8 at% burnup. Fracture or cleavage of $UO_2$ grains lead to a structure of small grains similar in size to those observed in irradiated high burnup $UO_2$. The limitations of such simulation experiments are discussed and the parameters are defined for conclusive reactor irradiation experiments to finally understand the underlying mechanism.

## 1. Introduction

In recent years, the burnup of light water reactor, LWR, $UO_2$ fuel has frequently been increased to more than 40 GWd/t U, and 50 to 60 GWd/t U have been reached, both in experimental and in power reactor fuels. At burnups above about 45 GWd/t U, a porous outer ring is formed with a typical thickness of 100 to 200 μm (e.g. refs. [1,2]). In this region, the local burnup is largely increased due to Pu-formation by resonance absorption of neutrons. The phenomenon of Pu-formation is known for long (e.g. refs. [3–6]). It is due to the very high capture cross section of $^{238}U$ for neutrons of certain energies which causes the neutrons to be absorbed in the fuel close to the surface, resulting in a gradient in Pu-concentration that falls off approximately exponentially into the fuel volume. The formation of the porous zone in high burnup LWR fuel is a much more recent observation. These two phenomena are so far usually being considered to be identical in spatial extent *. The question whether or not they are only related to one another for the specific irradiation conditions (burnup range, linear power or fission rate) of the so far published results on LWR fuel postirradiation examinations in the above burnup range is obviously of large scientific and technological interest. The porous zone shows very small grains of up to 1 μm diameter and a high porosity of up to 30%. The author has introduced the term "cauliflower" structure to described the appearance of the fuel, other authors have named the phenomenon the "rim effect". Note, however, that the term "rim effect", is being used to describe three (or even four) separate observations and their extent into the fuel volume: (i) Pu- and burnup increase, (ii) (a) the porous zone, (b) the large reduction in grain size, and (iii) a decrease in the content of fission xenon within the $UO_2$ grains as measured with electron microprobe analysis [7].

It is important to realize that though this zone appears to be thin, its apparently small thickness of 100 to 200 μm corresponds to 4 to 8% of the fuel volume. We thus deal with a fuel volume. The term "surface effect" sometimes used is therefore misleading, and when using the word "rim" one should keep in mind that a fuel zone or shell is involved. This rather important volume fraction is located near the fuel surface, hence at a location of utmost importance for two reasons: the large porosity and the high burnup cause a low thermal conductivity and hence a thermal barrier at the clad [8]. Therefore, fuel temperatures may be increased with the unwanted consequences of potentially larger fission gas release, etc. Secondly, in the scenario of direct storage of spent fuel in a repository,

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studied by a number of nations and in the unlikely event of any groundwater possibly having access to the repository and to the fuel, the water will first interact with the Pu-rich, high burnup, radiation-damaged rim zone of small grain size, large porosity and thus large surface area available for leaching, before leaching the remaining $UO_2$. Note that even if this zone occupies only 5% of the fuel, the contained radioactivity (Pu and fission products) will be about 10% of the fuels inventory, a fact of importance for a spent fuel program in which the ceramic fuel is regarded as barrier against activity release.

Though this "cauliflower structure" has been seen in a number of laboratories, there seems to be no consensus on the mechanism of its formation and on its importance for higher burnups. For instance, the new structure was suggested to be formed by chemical reactions with fission products such as Cs [9] and frequently it is thought to be entirely due to Pu-formation (and thus locally increased burnup) by resonance neutron capture (which only occurs near to the fuel surface). If this would be correct, the rim effect would remain localized in the vicinity of the fuel surface and would not necessarily present a barrier to increasing the average burnup. However, a very similar effect was already seen in early irradiations of highly enriched $UO_2$ (see below) and is therefore not entirely due to Pu-formation.

It is the intent of this paper to report further evidence from an ion implantation study indicating that the formation of the porous zone is not necessarily a phenomenon restricted to a shallow fuel rim (as Pu-enrichment is). These studies indicate that the cauliflower structure is due to a grain subdivision process of the $UO_2$ structure which is related to burnup (fission gas content) and low local fuel temperatures (absence of fission gas release). Some important aspects of both the old and the new evidence will be described in the following.

## 2. Observations on the rim effect

It was realized rather early [3] that the strong resonance absorption of neutrons in $^{238}U$ leads to a surface enrichment in $^{239}Np$ and therefore also in $^{239}Pu$ and thus in fission rate and local burnup. Fig. 1 shows three different typical observations from different periods: in its right part, early measurements of 1958 are shown, obtained with stacks and spirals of U-foils [3]. An increase by about a factor of 2.5 in $\gamma$-activity emitted from $^{239}Pu$ in the first 100 $\mu$m is obvious. Fig. 1

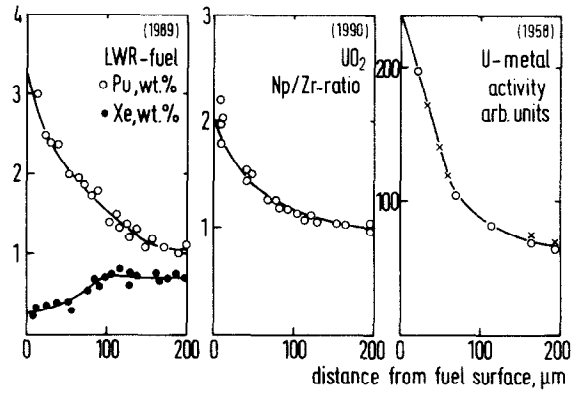


Fig. 1. Typical analyses of the rim zone performed between 1958 and 1990: Pu- and Xe-content in high burnup LWR fuel [1], Np-formation near the surface of $UO_2$ [4] and early measurements of n-resonance capture in U-metal [3].

includes two other measurements: that obtained with chemical etching of $UO_2$ discs irradiated for a short time [4] in the middle of fig. 1 where the ratio of $^{239}Np$ to the fission product $^{95}Zr$ is plotted and where the solid line is obtained with a model to calculate the effective absorption cross section of the $^{238}U$ (n, $\gamma$) reaction as a function of depth *. Finally, in the left part of fig. 1, electron microprobe measurement [1] of the Pu-content at the rim of a high burnup LWR fuel are shown. Ref. [1] contains similar profiles of e.g. Cs proving that (i) the burnup follows the shape and extent of the Pu-profile and that (ii) there is no enrichment of Cs (except possibly locally at cracks). Shown is also the decrease in the content of the fission gas xenon within the fuel grains. Similar profiles have been measured at other laboratories providing confirming evidence [14].

Fig. 2, also first shown in ref. [1], illustrates the "cauliflower" structure observed on a fuel (with 45 GWd/t average burnup) as that used to measure the

* The total neutron absorption in $^{235}U$ is small. In $^{238}U$, there are six strong resonances in the $^{238}U$ (n, $\gamma$) cross section at energies of 6.67, 20.90, 36.80, 66.15, 102.47 and 116.85 eV [5]. The resonance capture in the rim is dominated by the first three resonances, in particular by the one at 6.67 eV since for the higher energies, attenuation by scattering becomes important as a contribution to the total resonance interaction rate [4]. This phenomenon of neutron physics has recently been modelled with a multispace-dimensional code using multigroup cross sections and a continuous Monte Carlo method [7].

Pu-profile on fig. 1. A small grain size of 0.5 to 1 μm is seen together with a large porosity of up to 30%. Due to Pu-formation the burnup in this zone is ~ 10 at%. Many similar pictures were obtained in the past years in the hot cells of the European Institute for Transuranium Elements [11] and elsewhere. As mentioned above and included in the left part of fig. 1, a third type of observation exists, i.e. a depletion of the $UO_2$ grains in Xe, the fission gas being located in the new formed big pores (e.g. refs. [1,7,10–14]).

## 3. Simulation experiments with ion implantation to investigate the probable cause of the rim effect

The small grain size and the appearance of the rim structure have led to the proposal that this structure is the result of a reaction between $UO_2$ and the fission product Cs [9]. This, however cannot be generally true. For instance, the Cs-uranates that can be formed at the conditions of temperature and oxygen potential in LWR fuel ($Cs_2U_4O_{12}$, $Cs_2UO_4$, $Cs_2UO_{3.5}$) contain 0.5 to 2 Cs-atoms per U-atom. There is no evidence of radial transport of Cs (except possibly along cracks) and of significant depletion of Cs in the centre (except for transient-tested fuel). Even at a burnup of 10% as it occurs in the rim, the Cs-content is just large enough to convert 1% of the $UO_2$ to e.g. $Cs_2UO_4$. Also, some solubility of Cs exists in $UO_2$ [13] and the temperatures at the fuel surface are usually too low to reach thermodynamic equilibrium. Cs-uranates may thus form locally but cannot explain the extent of the porous zone.

On the other hand, the structure of Pu-rich particles in MOX fuel experiencing a local high burnup was reported to resemble to some extent that of the rim structure [15], and transmission electron microscopy near the rim shows a very high dislocation density indicating significant stresses in the fuel [16]. It is therefore plausible to investigate the possibility of burnup and fission gas content being the driving forces for the structural changes. Consequently, a study was started in 1988 [17] to "simulate" the phenomenon of the rim effect with ion implantation of rare gases. Previous work with ion implanted $UO_2$ measuring gas

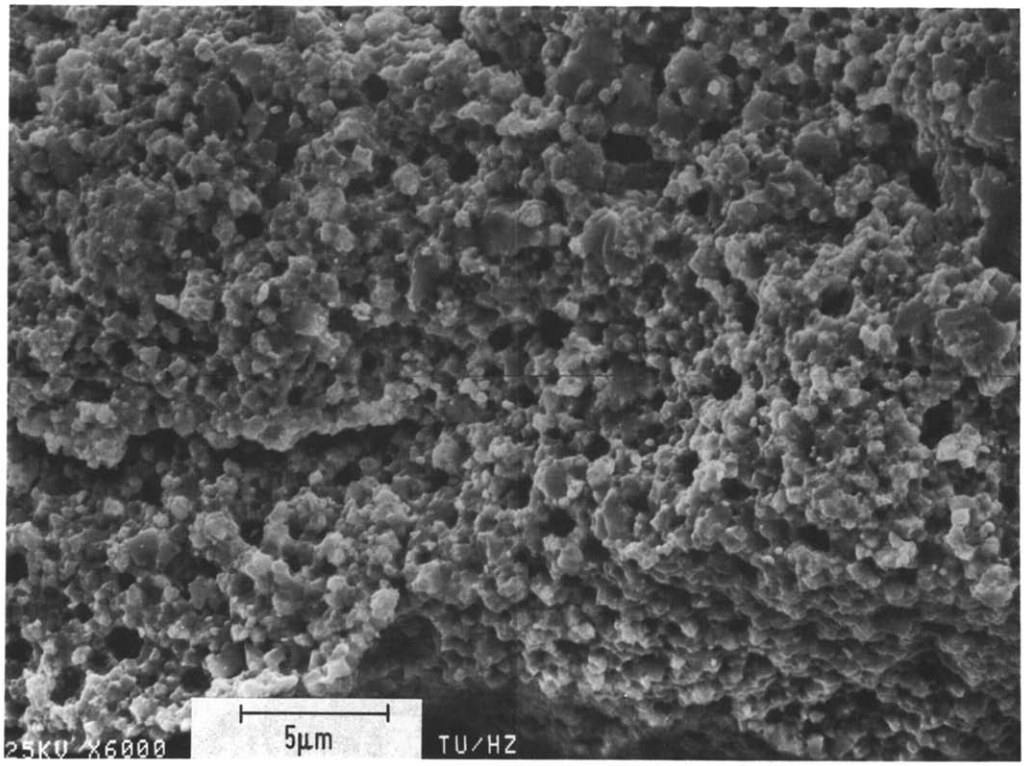


Fig. 2. Typical "cauliflower structure" of the outer porous zone in high burnup $UO_2$. SEM micrograph [1].

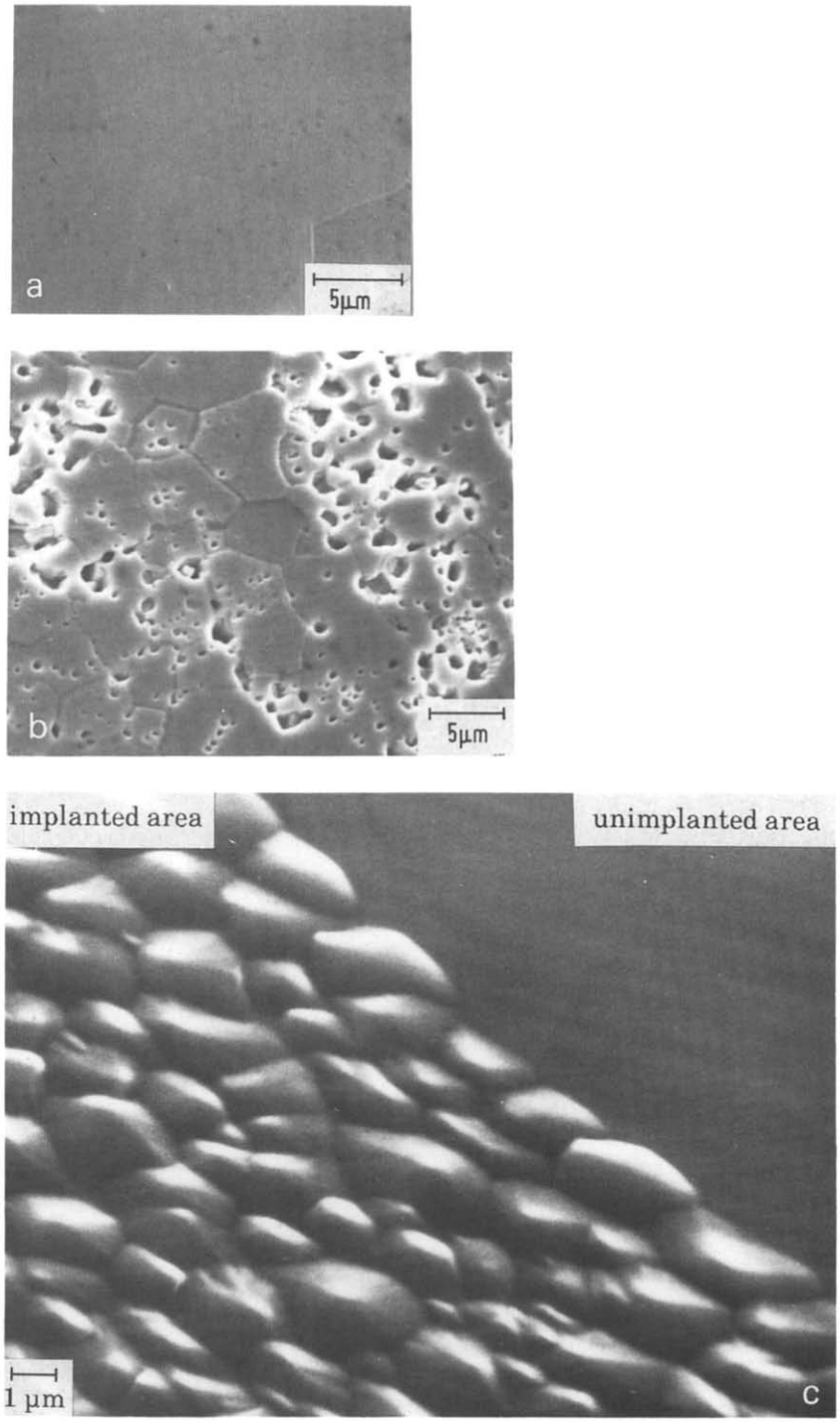


Fig. 3. Simulation of grain subdivision by observing fracture of $UO_2$ grains implanted with Xe-ions (300 keV energy, $5 \times 10^{16}$ ions/cm$^2$). The SEM micrographs show (a) unbombarded $UO_2$, (b) implanted $UO_2$ annealed to 500°C in reducing conditions ($Ar/H_2$), and (c) the edge between implanted and unimplanted area of $UO_2$ annealed to 500°C in very slightly oxidizing conditions ($CO/CO_2$).

release and using transmission electron microscopy [18,19] has shown that shallow implants (40 keV Xe-ions, median range 13 nm) with doses up to $2 \times 10^{16}$ ions/cm$^2$ do not change the $UO_2$ structure towards the "cauliflower" appearance. The same is true for similar shallow implants with Cs [20], iodine or Rb [21]. This observation is, however, not conclusive: at such a comparatively low energy, sputtering of the surface and radiation-enhanced rare gas diffusion and release prevent accumulation of large gas concentrations. Such large concentrations can be achieved at higher energy at which all of the implanted gas remains in the solid. Therefore, $UO_2$ was implanted with 300 to 500 keV Xe-ions. At a dose of $2 \times 10^{16}$ ions/cm$^2$, scanning electron microscopy of the implanted surfaces showed no surface changes, neither in the as-implanted state (at ambient temperature) nor following anneals at a number of temperatures up to 1500°C. However, at the higher dose of $5 \times 10^{16}$ ions/cm$^2$, a very pronounced breakup of the $UO_2$ structure was seen (fig. 3). Many of the cracks of the fractured pieces were aligned, probably following cleavage planes. This evidence is a clear indication that the $UO_2$ grains break up into smaller pieces at and above a certain inert gas and damage concentration under these conditions of damage formation *. It is not completely straight forward to relate the ion doses to fuel burnup since energy and damage rate differ. Though the gas concentration for a dose of $5 \times 10^{16}$ ions/cm$^2$ at 300 keV energy corresponds to the fission product concentration $\approx 7\%$ burnup, all foreign atoms are rare gas atoms – in contrast to fission. Yet, the evidence is clear: there exists a process of subdividing $UO_2$ grains at a critical gas content. This fracture process is probably caused by the high pressure of athermally-formed small fission gas bubbles. Recent channelling–Rutherford backscattering results [23] are compatible with the athermal formation of solid epitaxially oriented rare gas precipitates in $UO_2$. Such solid precipitates of rare gases can only exist at high pressures (order of $10^4$ bar) at room temperature.

* In channeling–Rutherford backscattering experiments [22], the rearranged originally single-crystalline $UO_2$ gives even the appearance of an amorphous state indicating disordering of the newly formed broken-up pieces by more than the critical angle for channeling ( ~ 1.5°). Parallel X-ray diffraction studies confirm this statement: disordered small crystals rather than an amorphous layer are formed. The phenomenon was seen in implantations performed at 77 K, room temperature or 500°C [22].

## 4. Discussion

The present results extent early experiments at the Bettis Atomic Power Laboratories of Westinghouse Electric Corporation by Bleiberg et al. [24–26] which have shown this process of "grain subdivision" in highly-enriched $UO_2$ plates irradiated to high burnup. The original 10–20 μm grains subdivided into grains of less than 1 μm at about $2 \times 10^{21}$ fissions/cm$^3$ (about 8.5 at% burnup) at low temperature. This grain subdivision affected some individual grains only at the critical burnup [27]. At the same dose, fuel swelling and gas release increased as well. Rest and Hofman [28] have very recently modelled these early gas release measurements and found that a grain size of 0.5 μm best reflects the experimental data, in agreement with the microscopical appearance.

These early experiments though being very helpful, suffer from not well-defined temperature conditions. Also, the "complete absence of any X-ray diffraction structure" at 9% burnup, and "retained crystallinity at 12.5% burnup with little peak broadening" were both reported. Note that $UO_2$ is not expected to be subject to radiation-induced amorphization (or metamictization) [29] but the stresses and deformations of the unit cell suggested to cause grain subdivision will certainly be dependent on fuel temperature during burnup and damage formation. Radiation-enhanced surface diffusion aided by the chemical environment could cause the final appearance: The pores formed during grain subdivision will also contain Rb, Cs, I and Te which facilitates rounding of the sharp edges of the smaller pieces formed by fracture and cleavage. The well known athermal in-pile sintering process could cause the new grains to form a coherent structure.

There are possible alternative explanations to grain subdivision by cleavage due to overpressurized fission gas bubbles, and there are possible arguments against the simulation by ion implantation. Radiation-induced largely athermal plasticity is known to exist in $UO_2$ during fission [29,30]. It is known to be proportional to the fission rate, and it has been observed very early to lead to e.g. creep of very highly rated fuel discs ($(U, Pu)O_2$ reaching 8% burnup in about 2 months at about 1000°C) into notches in the wall of the metal irradiation capsule [31]. Pronounced plasticity would prevent the buildup of high pressure bubbles and would cause swelling instead. However, at low fission rates and in the absence of large free space, as in a LWR fuel, the effect of radiation-induced plasticity will be smaller than in the above experiments. Ion implantation produces collision cascades which also cause the

implanted material to become plastic. As an example, ion implantation is being used as a simulation technique which permits rapid determination to examine neutron irradiation creep in Zircaloy [32]. However, in the present simulation with $UO_2$, plasticity occurred only in the bombarded surface layer and not in the underlying crystal, in contrast to reactor irradiated fuel. Also, again in contrast to LWR fuel, the gas escaped from the subdivided zone, and the final product was not coherent as the cauliflower structure is, since following the subdivision process there was neither stress nor a radiation environment to cause in-pile sintering.

A possible alternative explanation to grain subdivision by cleavage due to overpressurized fission gas bubbles would be a type of recrystallization process similar to that known for cold-worked metals. The driving force for recrystallization is usually the increased energy of the plastically deformed matrix. Radiation-induced plasticity is known to exist in $UO_2$ during fission (see above) and is also evidenced by the high dislocation density in the cold outer part of high burnup $UO_2$ [16]. In cold-worked metals, the grains evolving from recrystallization are strain free and the migrating boundaries can absorb dislocation lines and sweep (accumulate) insoluble fission products out of the new grains. This phenomenon is well documented for some cold-worked metals. It was, for instance found in Ag [33] but not in Cu [34]. However, ceramics cannot be cold-worked as metals can, there is no cold-work process in a reactor, and there is no annealing treatment either as applied to recrystallized cold-worked metals. Furthermore, the fact that the implantation work shows fracture of $UO_2$ grains at high gas concentration though the dpa-levels are smaller than during fission is another argument against this alternative *.

## 5. Open questions

The cauliflower structure as so far observed at the rim of LWR fuel is related to enhanced fission rates and enhanced local burnup due to Pu-formation by neutron resonance capture of $^{238}U$. Thus, three parameters are different from those of an enriched $UO_2$ fuel reaching similarly high average burnups of $\sim 9$ at%: (i) the rim area is rather cold, (ii) it experiences a high fission rate, and (iii) most of the fissions are due to Pu. Pu-fission in $UO_2$ is known to be more oxidizing than fission of $^{235}U$ since much more metals (Pd, Ru, Rh) are formed in the fission of $^{239}Pu$, which do not bind oxygen. The results of Rest [28] indicate that the fission rate may be important and it is well known [30] that kinetic processes in $UO_{2+x}$ are enhanced compared with $UO_2$. On the other hand, fission gas solubilities are probably higher in $UO_{2+x}$ and the increased plasticity might reduce stresses due to fission gas bubbles in slightly oxidized $UO_2$.

It is thus difficult to predict the exact values of the parameters determining to which extent and at what average burnup grain subdivision will affect $UO_2$ fuel further in the pellet and independently of Pu buildup. The indication of a burnup of $\sim 9\%$ being critical of the early US results [24–27] suffers from the lack of exact knowledge of irradiation temperatures. Recent work based on measuring Xe-depletion in $UO_2$ grains [7] has revealed a threshold local burnup of 70–80 GWd/t for formation of the rim-zone defined as zone with grains depleted in Xe, and a very wide zone was reported extending deep into the $UO_2$ pellet ($\approx 1.2$ mm, hence comprising $\sim 50\%$ of the fuel volume) in fuel with a high average burnup of 85 GWd/t [7]. A second observation of this type was recently obtained in $UO_2$ irradiated to 70 GWd/t average burnup [35]. In contrast, $UO_2$ with an average burnup of 5.9% showed still the "normal" $\sim 150$ μm wide zone of Xe-depleted grains. More data of this type are needed. A parametric study investigating the effects of fuel temperature, fission rate, oxygen potential, and Pu-content would solve the open questions and would allow a prediction of the parameters for the burnup threshold for $UO_2$ in the absence of increased Pu-formation.

## 6. Conclusions and summary

The "cauliflower" structure observed in the outer zone of LWR fuel at burnups in excess of 45 GWd/tU is shown to be probably due to an intrinsic property of the $UO_2$ crystal structure: at a given damage level and inert gas content corresponding to about 8% FIMA, the heavily stressed grains subdivide into small crystallites of $\leq 1$ μm size thus forming about 1000 small grains from one original grain. Simulation by ion-implantation with xenon shows that fracture occurs in

* See note added in proof.

$UO_2$ grains at high fission gas concentrations **. The critical burnup is at ~8% FIMA, it may depend on Pu-content and it certainly depends on temperature. The phenomenon is known to be absent in highly-rated and thus hotter (U, Pu)$O_2$ fast breeder fuel since most of the fission gas is released. On the other hand, it is known for long [36] that isostructural thorianite-minerals containing ~2 at% He accumulated over geological ages disintegrate if heated to 950°C. A parametric study on the effects of fuel temperature, fission rate, oxygen potential and Pu-content of the fuel on the critical burnup is needed to define the burnup/temperature conditions for LWR $UO_2$ to develop the structure independently of the Pu-enriched zone.

The consequences for fuel operation and, in particular, for possible direct spent fuel storage are obvious. The increased heat production at the rim flattens the temperature profile while the increased burnup and porosity in the rim reduce its thermal conductivity and create a barrier for heat flow thus increasing the central temperature. For the latter, even todays rims are of interest: the oxidizing Pu-fission, the concentration of fission products and of Pu near the fuel surface and the large porosity at the rim where leaching will start ought to be considered in relevant programs regarding the ceramic $UO_2$ as a barrier: oxidation, porosity (i.e. larger available surface area) and radiation damage will all increase fuel dissolution. Recent ion implantation work [37] has shown significantly increased leaching of $UO_2$ damaged at low temperatures.

## Note added in proof

Since this manuscript was submitted, the author, together with Dr. G. Schumacher, KfK, has organized a conference (Symposium on Nuclear Materials for Fission Reactors, E-MRS, Strasbourg, France, November 5–8, 1991). Experts were invited who actively investigate different aspects of the rim effect. The Proceedings are a special issue of Journal of Nuclear Materials (volume 188; Guest Editors Hj. Matzke and G. Schumacher). The readers interested in more details are referred to this volume, and in particular to the articles presented by K. Une (NFD, Japan), M.E. Cunninghan and L.E. Thomas (PNL, USA), C.T. Walker (CEC) and M. Kinoshita (CRIEPI, Japan), I.L.F. Ray and Hj. Matzke (CEC). The information contained in these article elucidates the rim phenomenon further though by far not to the extent necessary for a complete understanding of the underlying mechanisms.

** Indications of formation of a polycrystalline structure in a single crystal due to Xe-impact and -content were, in fact, already noticed in the very first work [38] reporting absence of metamictization in $UO_2$, but formation of amorphous phases in other noncubic oxides. The example was single crystalline MgO which showed a polycrystalline pattern in reflection electron diffraction following implantation with $2\times10^{16}$ Xe-ions/cm$^2$.

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