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Polygonization and high burnup structure in nuclear fuels

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Abstract

The rim effect, i.e., the formation of the high burnup structure typical for the outer zones of LWR UO_2 fuel pellets at extended burnup, is presently being studied in many laboratories. It is caused by a subdivision of the original as-sintered grains of the UO_2 pellets into 10^4 to 10^5 new small subgrains. The aim of the activities presently ongoing in different laboratories is to define the conditions (burnup, temperature, pressure, type of fuel, grain size, etc.) for the formation of the rim structure and to understand the mechanism of this subgrain division, or polygonization. The aim of the present paper is to discuss the existing knowledge on such polygonization processes in other materials, to confront this knowledge with the observations on UO_2 fuel and to discuss the attempts to model the rim structure formation. © 1997 Elsevier Science B.V.

1. Introduction

Polygonization, or grain subdivision of UO_2 LWR fuel is a phenomenon that has achieved worldwide attention in the last decade. It was first evidenced in LWR UO_2 fuel near the outer surface of the fuel pellets and has therefore been called the 'rim' effect. It is found in this 'outer rim' of typically 150 μm width first because neutron resonance capture by U-238 creates fissile Pu-239 in this zone, thus causing locally a much higher burnup. At extended burnup (cross-section average > about 70 GWd/t), the 'high burnup structure' extends much deeper into the pellets. This structure is characterized by the formation of very small grains (0.15 to 0.3 μm) which are depleted in fission gas and, at least in the outer rim zone, are accompanied by increased porosity. The resulting structure has been named a 'cauliflower structure' [1,2].

The more scientific term for grain subdivision is polygonization, or more exactly 'microscopic' polygonization. If a high dislocation density exists in a solid, as is the case in high burnup (e.g., 60 GWd/t) UO_2 fuel, the elastic stored energy increases and acts as a driving force for dislocation mobility thus reducing the stored energy. Some dislocations will annihilate each other. Polygonization is the term used to describe the rearrangement of those dislocations which do not annihilate one another, into walls of dislocations, forming small energy 'sub-boundaries' and rather perfect but slightly disoriented subgrains between those new boundaries.

A similar polygonization can also be produced in UO_2 by irradiation with fission product ions accelerated to different energies between 0.3 to 1.5 MeV [3]. Polygonization has also recently been observed in intermetallics (Zr_3Al, U_3Si), in minerals (e.g., olivine $(Mg, Fe)_2\ SiO_4$) [4,5] and in spinel $MgAl_2O_4$ irradiated with energetic ions [6]. The typical high burnup structure of UO_2 has been observed in high burnup fast breeder advanced fuel (U, Pu)C as well [7] (see Fig. 1). Recent HRTEM work on reactor irradiated UO_2 [8] and also on ion irradiated UO_2 [9] has revealed that the formation of very small subgrains of nanometer size as nuclei for polygonization can be one of the mechanisms causing polygonization in UO_2. This mechanism is related to damage formation. Alternatively, polygonization can also be caused by high concentrations of fission gases forming overpressurized bubbles causing cleavage or cracks on a very small scale. A third mechanism is breakage of larger grains due to the formation of amorphous tracks. Direct nucleation from cascade cores when the irradiation is performed near the critical amorphization temperature has been reported for other materials, but is not operative in UO_2, since UO_2 does not amorphize due to very effective instantaneous defect re-

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Fig. 1. Grain subdivision in pores of highly rated, Na-bonded (U, Pu)C at high burnup (8.3% FIMA, mid-radial position, ~900± 150°C).

combination in damage cascades and in thermal spikes of fission fragments [10]. Recent results of TEM, SEM and HRTEM and of oxygen potential measurements of polygonized high burnup UO_2 and ion irradiated UO_2 illustrate the complex set of parameters affecting polygonization of UO_2. A tailor-made reactor irradiation is aimed at better defining the decisive parameters [11].

Recent new results and modelling activities on the rim effect have been reported during the International Workshop on Interfacial Effects in Quantum Engineering Systems (IEQES 96) in Mito, Japan, August 1996 [12–19] and during a subsequent IAEA Technical Committee Meeting in Tokyo [20,21]. At these conferences, both experimental results on high burnup fuel [12–14,20], relevant data on damage produced by energetic fission products [12,15], results concerning the effects of reactivity-initiated accidents [16,17] and modelling approaches [18,19,21] were reported. Furthermore, there are two recent papers, one containing a careful SEM/TEM study [22] of a specific high burnup fuel which experienced an increased rating and therefore a high temperature phase at about half the final burnup and the second one [23] presenting a fuel performance modelling calculation of this special irradiation. Such fuel can give information on the conditions necessary to initiate grain subdivision, since two possible parameters (fission gas content and radiation damage level) were changed at half the final burnup due to the high temperature phase. The reader is referred to these recent articles and the references contained in them for more details than can be mentioned here.

2. Mathematical models of rim structure formation

As shown above, more and more experimental observations of the rim structure formation are now becoming available. The over-all picture is becoming rather clear. Mathematical models of the structure formation are developed based on several aspects of this picture, with different objectives and from different points of view. Here we review presently proposed models and discuss their use to evaluate fuel performance. The models can be divided into three categories.

(1) Models of category (1) deal with the process of *initiating* the restructuring. The objective of such models is to describe the mechanism of the burnup threshold at which the formation process starts and to clarify the effects of parameters such as temperature, fission rate and some additive component in the fuel (for example Gd) on the threshold. The models should explain the preferential sites for initiating the restructuring: observations show that the process starts at grain boundaries or at pore surfaces. The role of this category of fuel performance analysis is thus to identify extent and local (radial) position of the restructured fuel at a given burnup.

(2) The models of the second category describe the development and growth of the restructured region *after* the initiation of the process. The relative amount of the rim-structured area should be described as a function of the burnup and other parameters such as temperature or mechanical stresses. The effect of the grain size on the development of the restructured region should also be described and modelled. This could lead to improvements in design or fabrication of the fuel in order to control the process evolution during the in-pile life of the fuel.

(3) The models of this category describe the performance of fuel with a known fraction of the rim structure in terms of material property changes. The objective is to assess the effect of changes of fuel material properties of LWR fuel on its performance during transient, accident and steady operation of nuclear reactors, especially for extended burnup. This includes models of thermal conductivity, of density decrease and swelling due to the pore formation and of mechanical properties such as hardness, fracture toughness and thermal and irradiation-induced creep and hot pressing.

2.1. Brief review of the proposed models

Most of the models proposed so far are of the category (1). The first model of Rest [24,25] is based on the evaluation of a fractional fuel volume where some dislocation cells can move freely to recover irradiation damage. This free moving dislocation structure is taken as being a recrystallization nucleus. As the impurity concentration increases due to burnup accumulation, the solute atoms

(burnup dependent) and vacancies (fission rate dependent) combine to move and react with the nucleation cell to eliminate the volume of the free moving dislocations. The initiation of the restructuring is modeled by statistical distributions of stored energy to these moving nuclei. When the energy of a nucleus exceeds a certain limit, the restructuring starts. Experimental observations show accumulation of edge dislocations, distortions of the fuel lattice and resultant elimination of the free moving volume for dislocations as the burnup increases. However, the basic point defect process assumed in this model, i.e., the transport of pairs of a fission product solute and a vacancy, has not been proven experimentally to exist so far.

The new model of Rest [18] is also of category (1). It was inspired by molecular dynamics studies performed by Diaz de la Rubia and Gilmer [26], which suggest that cascade damage produces shock waves which may have an essential role in creating recrystallization nuclei. The shock wave is assumed to produce expanded areas, called centers of expansion (CE), which are locally amorphous. It is also assumed to produce propagating compressed waves, called centers of compression (CC). A rate theory of CE and CC was developed and was first applied to the amorphization and recrystallization of U_3Si_2. Later it was also applied to the rim structure formation of UO_2 to describe the development process of the recrystallization nuclei. Note, however, that there is well documented evidence that UO_2 does not become amorphous [27] and that high energy ions with energy loss values larger than those of fission fragments are needed to produce visible tracks in UO_2 [28].

The model of Lemekhov [29] is also of category (1). It is based on the saturation of fission gas in the matrix related to atomic mixing along fission tracks. The criterion is the stability of capillaries along fission tracks. The pressure of FP gas atoms increases in the capillary-shaped mixing volume of UO_2 lattice sites and when the pressure exceeds the limit of the surface energy, the capillary becomes unstable.

The model of Kinoshita [19] is of category (1). It is based on stability criteria of rate equations with spatial diffusion. The well-known reactions of point defects, gas bubbles and dislocations are analyzed numerically assuming a high dislocation density as was observed near the rim-structured fuel by transmission electron microscope (TEM). The theory shows that spatial instability destroys the uniform distribution of point defects and that the local density of dislocations can increase significantly.

The model of Baron et al. [21] covers not only category (1) but also category (2). The model assumes that the excess matrix energy is converted into surface energy of newly created boundaries of the sub-divided grains. The extent of the sub-division process and the resultant fractional area of the sub-divided grains are given as a function of the accumulated energy.

The model of Chkuaseli and Matzke [30] assumes overpressurized fission gas bubbles to be the main source of stress, causing loop punching. In minimum stress regions, i.e., halfway between bubbles, the concentration of interstitials is highest leading to the formation of a high dislocation density, of dislocation tangles and walls. At the threshold burnup, subgrains form, with a size of the order of the average distance between bubbles. In this model, bubble sweeping is not the main mechanism for depletion of xenon fission gas. Rather, resolution destroys the bubbles very effectively once restructuring has occurred, since restructuring provides a large sink concentration and since gas atoms can reach the many new subgrain boundaries easily via radiation enhanced diffusion. Some larger bubbles are less subjected to resolution. These bubbles continue to capture fission gas atoms and grow into the observed μm-size pores containing most fission gas. This model of Chkuaseli and Matzke [30] thus also covers categories (1) and (2).

2.2. *Verification and use of the present models and direction of development*

One of the challenges of models of the category (1) is to predict the effects of parameters such as temperature or fission rate. The experimental observation shows that the grain subdivision starts at around 7% FIMA (or around 70 MWd/kg U) local burnup and below 1200°C. Recently, however, in an accelerated irradiation experiment using thin rods, the initiation burnup was found to be only around 60 MWd/kg U [31]. The fuel was irradiated in the Halden heavy water reactor with higher enrichment and the fission rate was two times higher than that of standard commercial LWR fuel. Although the experiment indicates an effect of fission rate, there is no proposed model which shows a definite fission rate effect at present. The model of Rest indicates fission rate effects in U_3Si, but not in UO_2.

Another application of the models of category (1) should be to understand the formation of similar structures in other advanced fuels such as (U, Pu)C or (U, Pu)N. At around the same burnup of 7% FIMA, these materials show the development of similar structures of sub-micron grains around pore surfaces as UO_2 does (see Fig. 1). The models should thus describe the restructuring process as a universal process. If the dominating factor of the process could be identified, a delay in the initiation or a spread of the period of the restructuring over a burnup range and a reduction of the transition effects to avoid drastic changes of the fuel performance could possibly be achieved.

For the models of category (2), the task is to describe the development of the high burnup structure as a function of increasing burnup. A driving mechanism could be the defect flux generated by fission damage. At the rim of the pellet, thermal activation is not the major process because of the low temperatures of < 600°C. Therefore, the development process, which consumes the generated defects, could be fission rate dependent. For instance, it is known that athermal, radiation-enhanced diffusion of the less

mobile lattice atoms in UO_2, i.e., the U-atoms, is proportional to the fission rate [32]. The organizing process of the restructuring involves reactions of short-lived defects such as interstitials and vacancies and long-lived structural defects such as dislocations and clusters of fission products in the lattice matrix. The rate equations for these reactions could be a useful tool to describe these processes.

As the process of the structural development is directly related to the gas bubble development, fission gas precipitation at, and possible transport along, the grain boundaries of sub-micron sized grains should also be considered. Bubble growth and possible mutual interaction and coalescence, hence reduction of the number of bubbles, should be taken into account. The Pu-rich agglomerates in high burnup MOX fuel, which show high porosity and mixtures of different phases of fission products, could give an indication of the final structure.

The model of Baron et al. [21] was developed along this line for the category (2). The objective is to simulate bubble development during and after the restructuring. The progress of grain sub-division is quantified by the increase of grain boundary surface area per unit volume (S/V). The bubble concentration is derived from interactions of dislocations and intragranular gas atoms. The intragranular fission gas concentration is determined by balancing the flux of fission-induced resolution from bubbles along the boundary surfaces, which is increased by the restructuring and the counter flux from a resolution layer, i.e., a layer which has the width of the resolution distance in the fuel grain. The FP gas concentration in the bubbles is due to diffusive trapping of the gas atoms from the matrix into the bubbles. The increase of the number of bubbles and resolution according to the Nelson scheme [33] are also included. The average size of the bubbles is presently estimated by assuming thermal equilibrium. This model's aim is to evaluate the swelling potential and the fuel behavior during and after the rim structure formation.

For the category (3), a first generation of models should be based on simplified correlations deduced from the experimental data. Presently, some projects are under way to provide such data, e.g., the Nuclear Fuel Industrial Research (NFIR) project organized by EPRI and the High Burnup Rim Project (HBRP) organized by CRIEPI [11]. For these projects, the thermal diffusivity of the irradiated UO_2 will be measured with the laser flash method. Heat capacity measurements will also be performed to obtain the thermal conductivity from the diffusivity data. The degradation of the thermal conductivity with burnup and due to restructuring depends on several factors: the existence of different phases in the fuel, precipitates, nm-sized grain boundary bubbles, μm-sized pores containing most fission gas, oxides of fission products, dissolved fission products, new grain boundaries and radiation defects. These effects could be clarified by similar measurement on SIMFUEL (e.g., [34]). It is thus also necessary to evaluate the effects of temperature history during irradiation. The models should be developed to account for processes of both damage production and thermal recovery of damage.Precipitation of gas during the measurements causes an effect similar to that of damage annealing, but it occurs in a different temperature region. This contribution was modeled by White [35] and modified by Turnbull [36].

For fuel material performance models, fission gas swelling, gas release and cracking by highly pressurized bubbles are main issues. The swelling during steady operation could be largely dependent on the mechanical constraint by the cladding as observed during PIE of PWR fuel [21]. Bubble development models of the category (2) may contribute to assess this swelling performance.

Rim structured fuel contains a large inventory of fission gas which is mainly stored in μm-size bubbles surrounded by the sub-μm sized new grains. As the gas pressure is proportional to the temperature, large enough temperature transients may cause the fuel to crack into small pieces, and burst release of the gas may occur. The behavior is expected to depend on the rate of the temperature increase. Moreover, the mechanical constraint is also expected to affect the cracking behavior. The recent transient experiments for simulating reactivity initiated accident (RIA) conditions indicate such cracking and gas release behavior from the rim structured area of high burnup pellets [16,17].

2.3. Discussion and comments on modelling

All the proposed models, except those of Lemekhov, are based on rate equations. However, rate equations of the defects request some abstraction to use quantities such as point defect concentrations and diffusion coefficients. For high burnup fuel, these quantities are not well defined. In particular, defects, in contrast to the fission gas atoms, can annihilate when they react with each other or migrate to grain boundaries or surfaces. Also, concentrations of interstitials and vacancies (both occur in two sublattices, i.e., the oxygen and the uranium sublattices) are not directly observable with the present experimental techniques, in contrast to, for example, fission gas bubbles. Because of the simplifications which have to be made, the results calculated with the equations may not be completely realistic so that verification by experiments is also, sometimes, impossible. When non-linearity is introduced to the rate equation framework, the theory may be restricted to one specific objective. Presently, it is not mathematically clear what happens when many different non-linearities are placed into one set of equations. Simulation using high computing power, with many degrees of freedom (for example use of the Monte-Carlo method), may lead to new, exciting results.

The difficulty is partly due to experimental restrictions, as mentioned above and to lack of more quantitative investigations. For improved rim structure modelling, more data on fission gas concentration and on the statistics of bubbles would be very helpful. The number density, size

distribution and internal pressure of bubbles for burnups below and above the threshold are key observations to develop models, as are exact statistics on subgrain sizes, dislocation densities and pore sizes.

3. Gross structural changes due to radiation damage, or to other physical processes

In a pioneering study of Xe implantation in different oxides, one of the authors has shown more than 30 years ago [37] that two types of gross structural damage can occur due to high dose Xe-implants:

(i) A phase change from the crystalline to the amorphous state (c–a phase transformation, called metamictization in minerals containing U or Th, i.e., actinides decaying by α-decay).

(ii) A phase change from the single crystalline to the polycrystalline state.

Amorphization occurred in anisotropic (non-cubic) oxides such as TiO_2, Al_2O_3, U_3O_8, etc., whereas single crystals of MgO developed a polycrystalline state. In this early work, polycrystalline UO_2 (and ThO_2) were shown to remain crystalline and not become amorphous. The technique used to observe the phase changes, i.e., reflection electron diffraction, was not suitable to detect polygonization or grain subdivision of polycrystalline sintered specimens [37].

3.1. The crystalline-to-amorphous (c–a) phase transformation

Amorphization, i.e., the c–a phase transformation, is by now a frequently observed consequence of accumulated radiation damage. Different criteria were elaborated (e.g., based on crystal structure, bonding conditions, thermodynamic properties, etc.) to explain why some substances amorphize, whereas others do not (e.g., review [38]). The criteria show that UO_2 is one of the oxides which do not become amorphous (e.g., [27,39]). Fig. 2 shows the different energies (from 40 keV to 700 MeV) and energy loss values dE/dx (nuclear and electronic energy losses from 2 to 20 keV/nm) which were used by one of the authors and his co-workers for irradiation of UO_2 with Kr- and I-ions. For other heavy ions (e.g., Rb, Te, Cs, Xe and U), even wider ranges of up to 2.7 GeV and $\simeq 60$ keV/nm were used. No amorphization was observed for any of these conditions (which include fission energy). Amorphous UO_2 can be produced by evaporation on an amorphous substrate. Upon annealing, it recrystallizes at 675 ± 15°C [39]. UO_2 thus fulfills the criterion, based on a thermodynamic model, stating that amorphization does not occur if the ratio of the crystallization temperature to the melting point, T_c/T_m, is ≤ 0.3 [38,39]. It also fulfills the other existing criteria. Though the c–a transition does not occur in UO_2, it is illustrative for the present purpose to

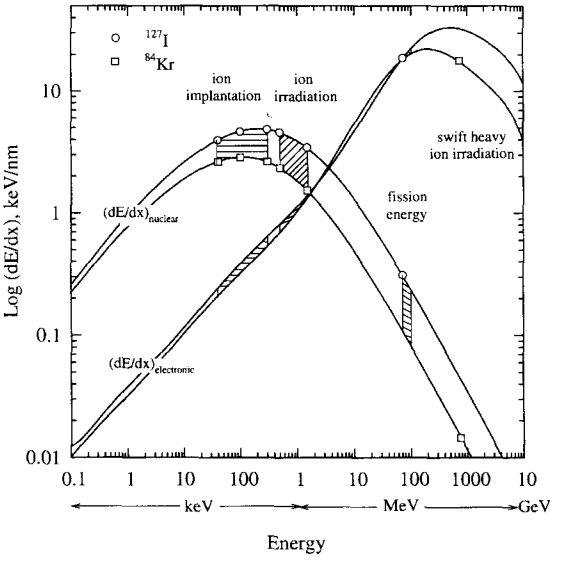

Fig. 2. Electronic and nuclear energy loss of a heavy and a light fission product atom (iodine and krypton) in UO_2 as a function of energy. The experimentally used ranges are also indicated (TRIM-code calculations).

briefly summarize the existing knowledge. c–a phase transitions cannot only occur by energetic particle irradiation, including α-decay (e.g., zircon $ZrSiO_4$ and other matrices for radioactive waste solidification) and fission (e.g., U-silicide), but also by a variety of other solid-state processes, including interface interdiffusion, hydrogen charging, thermal annealing, mechanical deformation and high-pressure compression [40]. Wolf et al. [41] have recently emphasized the parallels between amorphization and melting. A cylindrical molten zone is formed in UO_2 along the path of each fission fragment [28,42]. Recent observations of ion tracks in UO_2 [28] can be used to calculate temperature–time conditions: the results are compatible with maximum temperatures of 6000 K and molten volumes of about 8 μm length and some 10 nm diameter. This process contributes to radiation-enhanced diffusion [32], but does not cause amorphization of UO_2 either, just as ion impact does not amorphize UO_2, not even at high doses and very low temperatures, e.g., at 5 K [43]. This fact excludes also one of the possible mechanisms for polygonization (see Section 3.2). The remarkable radiation stability of the fluorite-structured UO_2 is convincingly demonstrated by the fact that the threshold for polygonization of about 7 at% burnup implies that no gross structural damage has occurred before the corresponding damage level of some 2000 dpa (displacements per atom) is reached. Note that each fission produces two fission fragments which in turn produce a total of about 100 000 displaced lattice atoms (about 27 000 U-defects and 73 000 O-defects, 80% of which recover instantaneously within the life-time of the displacement cascades, about 10^{-10} s [32]). Note also, that

for the same burnup, each UO_2 molecule in the fuel was within the molten central part of a fission track more than 10^4 times.

3.2. Grain subdivision or polygonization

As the c–a phase transformation can be due to a number of different processes, polygonization can also be caused by a variety of solid-state mechanisms. As mentioned above, MgO single crystals were rather early [37] shown to develop a polycrystalline structure due to high dose Xe-ion implantation. Recently, the same observation was made with UO_2 and Xe- or I-ions [3]. If similar experiments are done with sintered, polycrystalline specimens of conventional grain size (5 to 10 μm), grain subdivision occurs, i.e., as the big single crystal, the individual grains of the sinter are polygonized.

Polygonization has been observed in a number of materials and it can have many reasons. It has been observed in ion irradiated intermetallics (Zr_3Al, U_3Si), in ceramics (olivine $(Mg_{0.88}Fe_{0.12})_2SiO_4$, neptunite $Na_2KLi(Fe,Mn)_2Ti_2(SiO_3)_8$), as well as in UO_2 not containing fission products, but rather only damage (e.g., [4,5,9]). Wang et al. [5] suggested that randomly oriented nanoscale crystallites could either originate from the breakage of the original single crystal (or big grain) due to the formation of amorphous volumes in the irradiated material or could be directly nucleated from the cascade core when irradiation is performed near the critical amorphization temperature, hence that polygonization is the result of competition between amorphization and crystalline recovery. This mechanism is plausible for the above materials studied by Wang et al. [4,5], but is apparently not the reason for polygonization in UO_2, since UO_2 does not become amorphous. Polygonization can also be related to damage *and* impurity accumulation, e.g., in UO_2 implanted with Xe when fracture between overpressurized bubbles can produce small UO_2 blocks with a slight misalignment [3].

In addition, impurities alone, in the complete absence of radiation damage, can cause polygonization, an example being 'diffusion-induced recrystallization', or 'DIR' of Zn in Cu single crystals [44]. Furthermore, deformation or bending can cause polygonization as well (see also below for UO_2).

The term 'recrystallization' has been used for polygonization in UO_2, i.e., for the formation of the 'cauliflower'-type rim structure. This is a less suitable nomenclature. Recrystallization is the phenomenon observed in cold-worked metals involving motion of grain boundaries. The moving grain boundaries absorb dislocations and develop more perfect grains at the expense of the work-hardened matrix, developing through the solid from a small number of nuclei [45]. Note also, that grain boundary mobility is known to be slowed down by impurities as they exist in large quantities in irradiated UO_2.

One of the authors has previously investigated the phenomena occurring in rare gas-implanted annealed and in cold-worked metals (Ag and Cu) for conditions which cause polygonization in UO_2 [46–48]. No instantaneous recrystallization due to gas and damage was observed. Rather, the usual increased temperature was needed to start recrystallization and this was unaffected by gas content and damage. The results are reproduced schematically for Ag and in some detail for Cu in Fig. 3. For Ag, recrystallization swept out 90% of the rare gases (Kr or Xe). The process was thermally activated with $\Delta H = 1.35$ eV. This is in good agreement with $\Delta H = 1.4$ eV for recovery of the electrical resistivity which occurs in the same temperature range and is conventionally used to study recrystallization. Note that no sweeping of Kr was observed in cold-worked Cu. In conclusion, recrystallization, normally observed in cold-worked metals but not in ceramics, needs increased temperatures and does not necessarily sweep gases. Grain subdivision in UO_2 occurs without temperature increase and does transport (not necessarily sweep, see above) the fission gases into the newly formed pores. One should note, however, that a nucleation and growth process was seen in the rim zone of high burnup UO_2 by Nogita and Une [8] and it forms an important mechanism in their model to explain the formation of the rim structure.

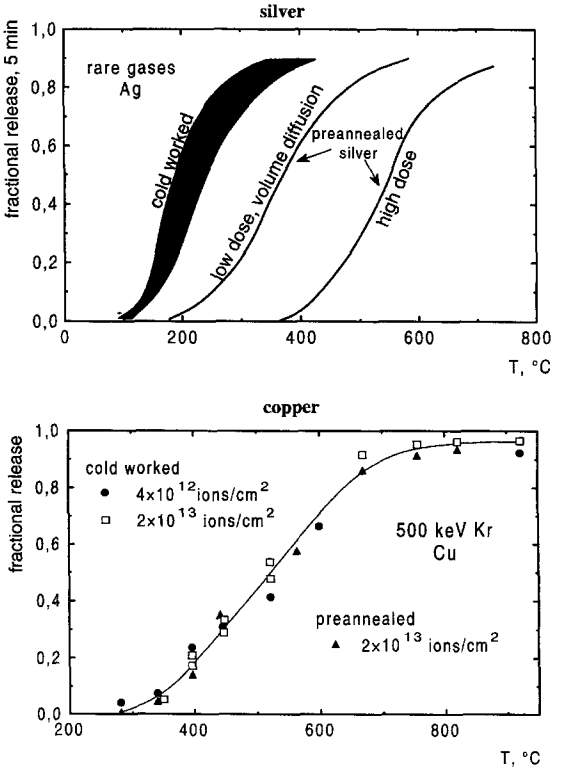

Fig. 3. Xenon and krypton release from ion implanted cold-worked and annealed Ag and Cu. Recrystallization causes gas release in Ag but not in Cu [46–48].

Finally, one should note that in addition to fission damage which can create dislocation cell structures of the size of the new grains, simple deformation can cause the formation of the same cells. As an example, in UO_2 single crystals deformed by 7% in compression at 1000 to 1200°C, a well defined very regular dislocation structure with cells of 0.15 to 0.2 μm size is formed, in the absence of any radiation and any impurities (fission products).

3.3. Suggested experimental work to solve open questions

As a first step to better modeling and understanding of the threshold values for grain subdivision in UO_2 fuel, the parametric field temperature–burnup-pressure-type of fuel should be investigated, as will be done in the tailor-made irradiation mentioned in Section 2.2 [11]. The quantities summarized in Section 2.3 should be determined for the different irradiation conditions.

As a second approach, single effect studies simulating reactor conditions should be continued. Ion implantation at higher temperature with volatiles (e.g., Xe and I) should be done to observe the temperature threshold for given damage and impurity levels. Parallel work with soluble fission products (e.g., Zr, Ce, or La) should be performed to check whether rare gases are necessary for polygonization, etc. Such work has also been started.

4. Summary

It is the intention of this paper to discuss the present state of knowledge on mechanisms causing gross structural damage, grain subdivision and polygonization in UO_2 (and in other materials). It is shown that polygonization can be caused by a series of different solid state processes including reactor irradiation, but also ion implantation with and without rare gas atoms in the damaged zone, deformation, impurity introduced by diffusion, etc.

Since many of these processes occur in parallel in reactor-irradiated UO_2, the ongoing investigation on high burnup UO_2 should be extended to more separate effect studies. The aim is to find ways to delay the grain subdivision process, if possible.

Modelling activities to describe the complex processes of the rim structure formation are summarized and suggestions are made for further modelling activities. The paper is thus meant to extend the large number of experimental and modelling results presented at the IEQES and IAEA meetings held before and after the present IUPAC Conference.

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