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Water reactor fuel damage mechanisms : A review

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ABSTRACT

A review is given on the LWR fuel damage mechanisms with specific emphasis on the cases of degradation processes having only minor contributions on the behavior during standard irradiation, but that can lead to drastic degradation in particular conditions. With that respect, both the oxide (fission gas release) and the cladding (water side corrosion, pellet cladding interaction and growth) are involved. Safety aspects lead to greater attention to failed fuel rod operation (secondary hydriding and major cladding degradation) and to rod behavior during control rod ejection transients or loss of coolant accidents.

INTRODUCTION

The current behavior of the fuel used in power water reactor is extremely good, with a failure rate reported by the utilities as low as 10^{-5} per fuel rod.cycle. Nevertheless, a clear pressure on the fuel manufacturers can be observed for an improvement in cost and quality. Indeed the drastic reduction of the price of fossil fuel reduces, for the short term, the benefit of nuclear energy [1]. In addition safety constrains give rise to reduction in any sources of contamination. This leads to the requirement of clean cores, without any failed fuel rod, as standard operation modes.

The detailed behavior of the fuel rods has been the subject of regular surveys done either by international bodies or by utilities [2]. The current rate of failure remains on an average basis below 2.10^{-5} per cycle, i.e. less than one failed fuel rod per core reload. This means that a large part of the cores are free of fission product contamination. The operation improvement obtained in these power plants is a major driving force to obtain such reduced constrains in other reactors. Indeed, if failed fuel rod reload can be performed without difficulties if enough care is taken, as currently done in France [3], waste level reduction standards persuade the utilities to operate as much as possible non-contaminated cores.

In this review, we will scan the different mechanisms involved in fuel rod failure and the effects of irradiation on the components of it. Therefore, we will describe the failure mechanisms induced by debris, grid-rod fretting and other assembly related mechanisms. With respect to material irradiation induced modifications, corrosion and irradiation defects of Zr alloys will be described, while for UO_2, the relevant points are the increase of fission gas release, the microstructural changes in the RIM region, the decrease of thermal conductivity with burn up. In addition several rising issues are to be considered regarding safety and end of fuel cycle.

ROD FAILURE MECHANISMS

Assembly and core behavior.

In order to analyze the trend in fuel rod behavior, a good source of information is the series of ANS International topical meetings on "Light Water Reactor fuel performance". Hold every second year, they are the place where major changes and evolution can be observed on a near term basis. The last one (Portland, OR; March 2-6, 1997) was the opportunity to stress the progresses obtained in that area: In the recent past, the large majority of the rod failure was due to causes not directly related to core and assembly behavior, like debris damage or grid rod fretting. However adequate and effective corrective actions have been undertaken and failure rates has declined already for standard operations. Among theses external causes, the following will be analyzed: debris fretting, rod vibrations and assembly geometry changes and their consequences.

The *debris failures* occur in any type of reactors. Metallic parts, such as metal-working chips, turnings, wires, clips, electrical connectors ... or ball pen springs, are released in the core during outage operations. The coolant flow transports them to the core, where they are trapped by various obstacles. Among them, the grids of the assembly act as very efficient filters. Indeed, series of pool examinations of fuel assemblies where failure were detected, allowed to conclude that 95 % of the failures due to debris occurred at the lower grid [5]. Once trapped in the low cross section channel of the grid, these parts vibrate, due to highly turbulent flows, and induce very localized wear, leading soon to rod failure. A typical morphology of debris failure is given in Fig. 1. The damage is a narrow, well defined groove, corresponding to the area swept by the debris during its vibration.

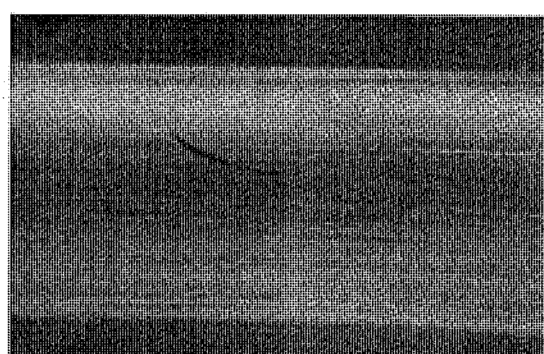

Figure 1: Debris fretting groove in a fuel rod [4].

The corrective actions undertaken by the fuel designers were the addition of an anti-debris filter at the bottom end of the assembly. Some vendors propose also design changes to the lower grid to add more trapping capability: the lower grid is located in front of a lengthened bottom plug. Thus, if a debris passed the debris filter, it is trapped by this grid and, it can only wear the massive bottom plug of the fuel rod, avoiding any leakage [5-7]. In addition, the utilities, in connection with the fuel vendors, have developed quality control procedures during fuel outages and reloads to avoid the release of these debris. All these corrective actions gave impressive results and the current failure rates due to these debris mechanisms are reduced by at least a factor of ten, leading to the majority of the cores to be defect free [1].

The *grid-rod fretting* is a failure mechanism occurring when the grid springs cannot hold anymore the fuel rod in contact with the grid dimples. High vibration amplitudes of the fuel rods induced by peculiarly turbulent flow are the main causes of this fretting mechanism. It occurs in location of high turbulence: inlet close to the bottom grid or areas of significant cross-flow close to intermediate grids in cores with mixed assembly design loading. The grid-rod fretting signature is straightforward as the wear areas due to the springs and the dimples are easily identified. A typical example of such failure is given in Fig. 2 [6]. Rod failure due to baffle jets can be sorted in this category, as well as the failures induced by "fluid elastic instabilities". In all cases, strong cross flows induce excessive fuel rod vibration. In extreme cases, turbulent flow frequency spectrum characteristics can interfere with fuel rod resonant modes.

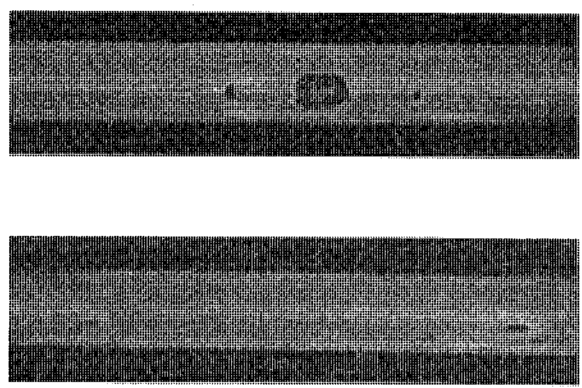

Figure 2: Through cladding grid-rod fretting wear

The corrective actions include improved design of the grid cells and reduction of the turbulent flow induced vibrations. The first point is related to fuel assembly design. It is not a simple task since the spring strength has to be high enough to hold the rod in contact to the dimples without lift-off during vibration, but not to high in order to avoid enhanced fuel rod bowing due to excessive friction between the growing fuel rod and the skeleton of the assembly. In addition, care has to be taken for the relaxation of the cell springs under irradiation.

The last phenomenon to be considered, with respect to the impact of assembly behavior on rod integrity are *the geometrical changes* induced by differential growth of the assemblies or water channel boxes. Mainly induced by the growth under irradiation of the Zr alloys, the assembly bowing is due to the incompatibilities between the different deformations (creep and/or growth) of the components Indeed, usually the fuel rod, the guide tubes or the channels are not made of the same the material, nor irradiated in identical conditions, therefore differential strain may occur. The consequences induced by the bowing affect two main domains: the thermal-hydraulic conditions are degraded, leading to reduction of the dry-out margins [8,9] and the friction of the control rods on the inner surface of the S bent guide tubes reduces the drop time of the control rod bundles. The corrections undertaken are a mixture of better homogenization of flux received by the components by shuffling and rotation of the channel boxes and a careful engineering redesign of the assembly.

Rod failure by Pellet Cladding Interaction

PCI is a fuel rod failure mechanism that is due to a stress corrosion cracking of the cladding occurring when the cladding is circumferencialy stressed due to large UO_2 expansion. This thermo-mechanical strain is induced during fast increases in linear heat generation rate, after that the Zr alloy cladding has crept down to hard contact with the pellet [10,11]. The corrosive species considered is iodine, a fission product having high yield. Frequently observed in BWR's due to the specific neutron control procedures of these reactors, it was also observed in CANDU (induced by on line refueling) and is considered as potentially harmful in PWR's in cases of accidental fast power transients [11].

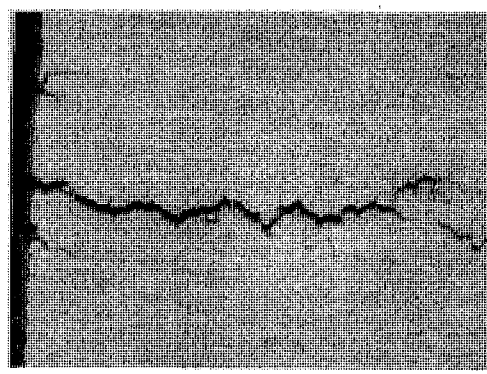

Figure 3: PCI cladding crack after power transient

Two improvements have been used: as far as materials are concerned, the inner surface of the cladding has been made more resistant to SCC crack initiation by adding a soft, low oxygen zirconium liner metallurgically bound to the bulk of the cladding [12]. For the CANDU type reactors, a liner of carbon (Canlub) has been developed. Additional efficient actions were changes in the processing of the cladding tubes, in order to obtain a more radial texture. Indeed, the SCC failures propagate as pseudo-cleavage cracks along the basal planes of the hcp lattice and the plastic strain is obtained at high temperature in Zr alloys by slip along the prismatic or pyramidal planes. Therefore the more tangential the <c> planes, the less susceptible the metal to SCC, since the resolved tensile stress to open the basal planes are reduced and the shear along the prismatic are increased [11,13].

After primary failure either by fretting or PCI, *secondary cracking* may occur. It has been mostly observed in BWR fuel elements: since the coolant pressure is higher than the internal pressure of the fuel rod, once primary failure have occurred, the water flows inside and allows internal oxidation. High radiolysis induced by the fission recoils in the gap drastically alters the water chemistry, increasing the corrosion rate. Both UO_2 and the cladding will reduce water. ZrO_2 and UO_{2+x} will be formed and hydrogen is released in the gap. Depending on the local P_{H2}/P_{H2O} ratio, reviewed in [14], the cladding will either oxidize or pick-up hydrogen. The swelling of oxidizing UO_2 and of the formation of ZrO_2 induces hoop stresses in the cladding and the precipitated hydrides allow axial splitting of the fuel rod. In the specific case of the Zr liner fuel rod described above, the poor corrosion resistance of pure Zr worsens the situation [15]. The recent development of Fe doped, low oxygen Zr liner may improve the behavior with respect to this failure mechanisms of dramatic consequences [6].

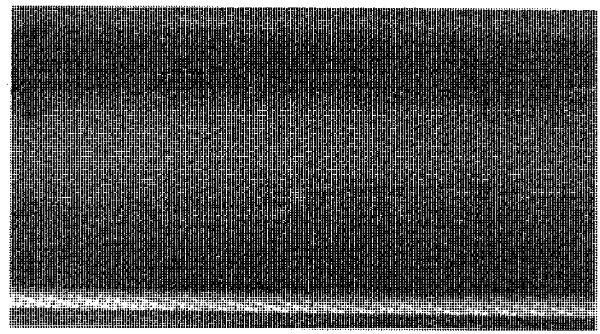

Figure 4: Axial split failure in a fuel rod after primary fretting wear.

MATERIAL BEHAVIOR UNDER IRRADIATION

Irradiation damage in Zr alloys

- Waterside corrosion

In the hot water of power reactors, the Zr alloys used for cladding and structural components develop a zirconia layer by oxidation. The oxidation modes have been observed to be different for the two major coolant conditions. In BWR's the coolant is a mixture of water and steam, in which the chemistry is controlled by the boiling on the fuel rod surface and by the equilibrium between the two phases. Under these conditions, nodular corrosion tends to develop, that can be enhanced under strong radiolytic conditions. In PWR's the coolant remains in a single phase condition, therefore the chemistry is better controlled. Hydrogen additions reduce the concentration of long life oxidizing species. However, boron is added to the coolant to control the nuclear flux, while lithium addition compensates the pH. It has been demonstrated that high LiOH concentrations increase the corrosion rates. In both cases the neutron flux itself affects the structure of the base metal and of the zirconia and, therefore, as a consequence, increases the corrosion rate [16].

The current oxide thickness may reach up to 80-100 μm at the end of life. However, the trend for higher burn ups leads to an intense R&D work for new advanced Zr alloys of high corrosion resistance. New binary or multi-component alloys show promising behavior and several of them have demonstrated excellent performance in several lead use assemblies up to high burn ups [5,7,17]. However, corrosion cannot be eliminated and in any cases reduces the thickness of the remaining metallic cladding. Indeed, the mechanical strength of the zirconia is low and due to its porous structure, cannot contribute significantly to the mechanical resistance of the cladding. In addition, the zirconia layer acts as a thermal barrier and its development increases the mean temperatures of the cladding and of the fuel.

In conjunction with corrosion, hydrogen ingress (*H pick up*) is a phenomenon affecting the Zr alloys. An average of 10 to 50 % of the hydrogen released by the reduction of water is trapped by the Zr alloys. The mechanisms involved are not perfectly understood as the zirconia layer is a perfect diffusion barrier for the proton. Diffusion through intermetallic precipitates or other paths is under investigation. Hydrogen solubility in Zr alloys is strongly temperature dependent. Close to 100 ppm at 300 °C, it is below 1 ppm at room temperature. Also higher oxygen concentration increases the hydrogen solubility. The result is that hydride precipitation occurs when the reactor is cooled. Due to the brittle nature of these phases, high hydrogen concentration drastically reduce the ductility. Recent observations confirm also that the hydride layers, occurring when the solubility limit in temperature is exceeded, enhance the corrosion rate of the alloys [18].

- Irradiation damage in the Zr metallic alloys

In Zircaloy, the transition alloying elements (Fe, Cr, Ni) are almost insoluble in the Zr matrix and are mainly present as $Zr(Fe,Cr)_2$ and $Zr_2(Fe, Ni)$ precipitates. Under irradiation the former are subject to an amorphous transformation, or at high temperature to a dissolution phenomenon. The effect of dose and temperature on this transformation has been studied in detail. Depending on the temperature, the iron released in the matrix can either reprecipitate, often at grain boundaries, or remain partially in supersaturation. The main consequences are the formation of <c> loops (described below) and a reduced corrosion resistance of irradiated cladding, explained by the reduced efficiency of these precipitates in the growing zirconia layer.

The first step of the *fast neutron damage* is the formation of point defects. Under irradiation enhanced thermal diffusion, they combine as larger defects. Due to the low c/a ratio of the Zr hexagonal lattice cell, the dislocation loops obtained are <a> type, located in the prismatic planes. The result is a limited growth (aniso-

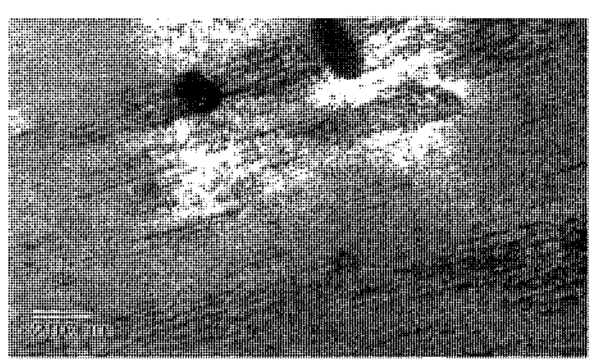

Figure 5: <c> type loops in irradiated Zry 4, responsible for accelerated growth.

tropic strain at constant volume). For high doses, the amorphous transformation of the precipitates occurs, with simultaneous release of alloying elements in the matrix [19]. The resultant high concentration of iron in the matrix has been shown to be liable for the formation of additional <c> type loops [20]. Opposite to the <a> loops, they are very large and are only vacancy type. Their formation, normally not seen in low c/a hcp metal, induces an accelerated growth clearly observed for high burn up recrystallised Zircaloy. In the figure 4, a TEM observation reveals these loops head-on.

- Plastic deformation of irradiated Zr alloys

The formation of the prismatic loops, mainly of <a> type, has a specific effect on the mechanical properties. Indeed, as observed also in bcc metals, once irradiated the material is hardened by dislocation loops. During plastic strain, these loops can be annealed by interaction with the gliding dislocations [21,22]. This gives rise to dislocation loop free areas where the strain is confined. This dislocation channeling explains the strain softening behavior of the irradiated Zr alloys when stressed circumferencialy. Indeed, for this phenomenon to occur, the loading mode should be oriented in such a way that the gliding dislocations have opposite Burgers vectors to the one of the loops, and glide planes different from the habit planes of the loops. In textured Zr alloy tubes, this is the case in transverse loading direction. Therefore axial or circumferencial uniform strains of irradiated Zr alloys are found different. Figure 6 gives an example of such dislocation channeling and localized deformation [23]. This is the origin of the reduced uniform strain of irradiated Zr alloys.

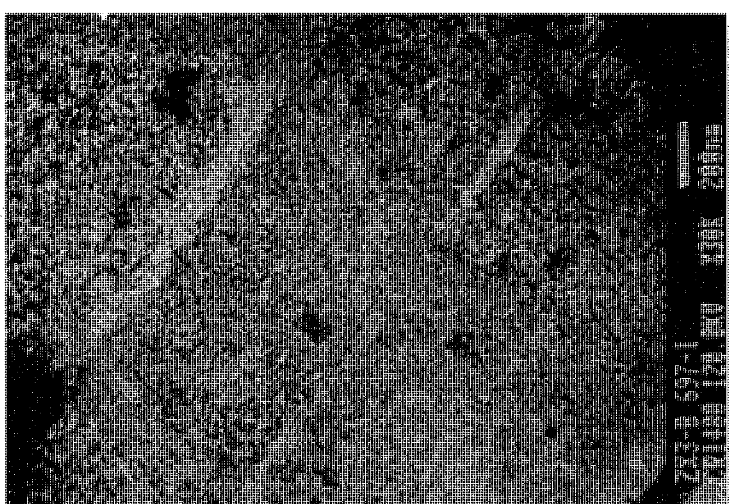

Figure 6: <a> type loop dislocation channeling after 1 % plastic hoop strain in irradiated cladding.

- Thermal conductivity degradation

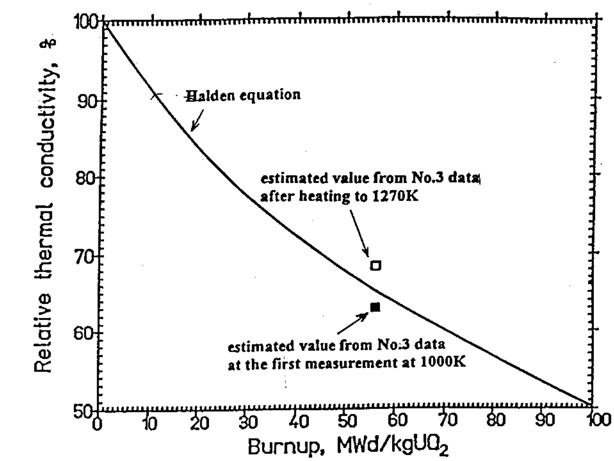

Figure 7: Effect of high BU on the thermal conductivity of UO_2.

Since the thermal behavior is one of the most important design and safety issue, the change of the thermal conductivity of the fuel oxide with irradiation is a matter of great concern. Minor effects like the in-pile densification at beginning of life or fuel swelling have been accounted for since a long time. Due to the trend to higher burn up, a more accurate knowledge of the thermal conductivity of the high burn up fuels became mandatory recently. Specific experiments have been set up and thermal diffusivity measurements have been performed using various techniques. The results are presented in figure 7. In addition to the drastic reduction in thermal conductivity at low temperature, a thermal recovery process has been observed [24]. It can be explained by the fact that the precipitation of the fission products, relaxing the matrix, reduces the density of diffusion centers for the phonons (i.e. fission products in solid solution).

- Fission gas release

Due to the high average temperature of the fuel, the point defects induced by the high energy fission recoils are continuously annealed. However, the fission induces a constant change of the chemistry of the oxide. Starting from a high purity ceramics, the fission and the transmutation lead to the doping by fission products and Pu isotopes. Since they cannot remain in solid solution in the matrix, thermo-chemical equilibrium has to be reached. Many of the numerous possible interactions between all these elements have been measured, simulated or computed. It is clear that, at equilibrium, the fission gases will be released, and that an increase in oxygen potential is expected, with some precipitation of complex oxide or metallic phases. The kinetics of the related precipitations are strongly dependent on the thermal activation, on the transport properties and on the driving force (departure from equilibrium). The balance of them is an increase of the kinetics as the burn-up to a high power (α BU^{3to5})[25].

- Outer RIM structure

For high burn up in water reactors (above 50 $GWd.t^{-1}$), the outer area of the pellet is observed to change in microstructure, with disappearance of the former grains and the development of a

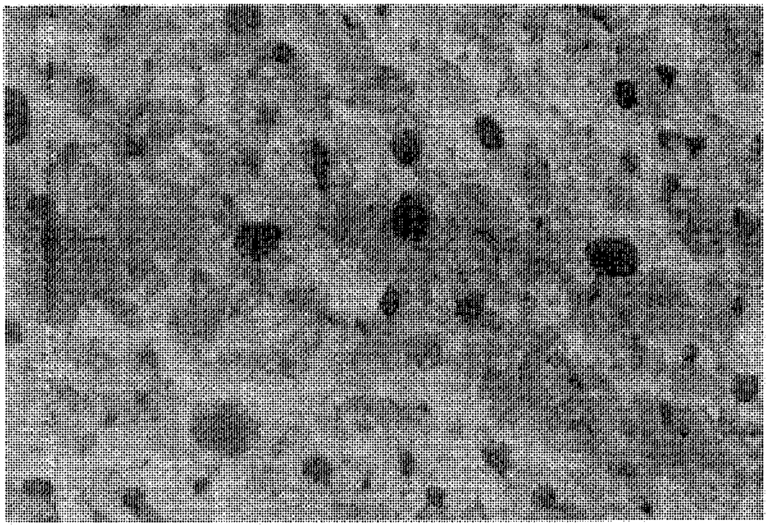

Figure 8: Typical microstructure of the RIM area.

new, very fine microstructure, associated to sub-micron porosities (Fig. 8) [26,27]. The detail mechanism of formation of this structure is still under investigation, but the main processes can already be described: in water reactors, the fast neutrons may return to the fuel after only partial thermalisation. The high capture cross section of ^{238}U for them leads to the formation of a higher concentration of Pu at the periphery of the pellet. This increases the local concentration of fissile isotopes and, therefore gives a higher BU in this area. Using particular fission products as local burn up tracers, typical values above 200 $GWd.t^{-1}$ have been proposed. The analysis of the formation mechanism of this fine grain microstructure by TEM confirms a continuous recrystallisation of the grains. The driving force for it appears to be the high density of dislocations resulting from the climb of the initial loops. Indeed in these areas, the low temperature of the fuel does not allow for long migration distances of the point defects and they anneal out on the intra-granular sinks formed at the beginning of irradiation [28].

- Specific aspects of mixed oxide fuels (MOX)

Currently used for reloads in Europe at an industrial scale, the mixed oxide fuels have properties and behaviors close to those of regular UO_2 fuels. Differences arise mainly from the heterogeneous microstructure obtained by master blend processing. The agglomerates of Pu rich areas, surrounded by the natural or depleted UO_2 matrix, localize the power generation, and therefore the fission product implantation inside them (or close to). In cold outer parts of these fuels, the Pu rich agglomerates behave as the RIM of standard UO_2, but they reach this typical structure much earlier, since the local BU is higher, roughly by a factor of 3, than the average of the pellet. This may be the reason, with higher oxygen balance of the Pu fissions, for the higher fission gas release observed in MOX fuel [29].

RISING ISSUES

As the target burn up increases, new aspects of fuel behavior have to be considered with extended analysis, compared to what was required for standard irradiation conditions.

The safety concerns of high burn up fuels come from two contributions: as the irradiation is associated with oxidation of the cladding by water, the oxide thickness increases with time. For the *loss of coolant accident* (LOCA), a critical point is the thickness of the oxide layer formed during the thermal transient of the accident. A typical criterion would be that a maximum of 17 % of the cladding transforms by oxidation. Since more than 80 μm of oxide are already present for an accident with high BU fuel, a question was risen on the effect of the preliminary oxide on the accidental sequence. Several experiments performed, either with pre-oxidized or irradiated cladding lead to the conclusion that the safety criteria are met for standard oxide thickness [30].

The development of SCC in PWR reactor vessel head penetration [31] could initiate a fast power transient by control rod ejection. The testings of high BU fuel rods during *reactivity initiated accidents* (RIA) in dedicated test reactors have shown that the energy deposition during the transient inducing rod failure and fuel dispersion is drastically reduced for high BU fuel rods. The impact of solid hydrides as crack starters has been demonstrated [32]. However, fine 3-D neutron physics computations have proven that the kinetics of such accident is less energetic and much slower in power reactors, and that the consequences of actual transients will remain very limited [33].

Currently weakly expressed, some aspects of *the end of the fuel cycle* may have to be considered for the future of the fuel design. Depending on the countries and utilities, the policy for end of cycle consists in either recycling or storage. For reprocessing, the dissolution kinetics is critical and, for instance, some specifications are given for MOX fuels. For open end fuel cycle nothing specific is requested, the depository bodies receiving the irradiated fuel "as it is". However, a general consensus arises for a two step depository scheme: intermediate dry storage for less than a century in an environment controlled surface area, followed by final underground depository. For the final stage, as geological time scales are concerned, the state of the fuel assemblies is of very limited concern, however, better performances during intermediate storage could lead to particular design specifications.

CONCLUSIONS

In spite of the extended irradiation conditions of the fuel assemblies in power water reactors, the performances of the fuel rods are extremely high for a large scale industrial product and remain improving continuously. Operations of reactors without failed fuel rod become more and more the standard condition, and the environmental pressure will add more to the fuel suppliers and utilities to do so. The fuel cycle cost benefit gained by higher BU remains still present, but a slower, careful and cautious approach is used now for the implementation of generalized very high BU fuel. Indeed, although the first steps were easy to reach, the BU and maneuvering capabilities already available are closer to the current engineering limits.

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