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DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS : RECHERCHES ACTUELLES

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

Nuclear fuels: design constraints, irradiation induced changes and end of cycle impact

Clément Lemaignan

CEA-Grenoble, Direction de l'énergie nucléaire, DEC Dir, 85X, 38041 Grenoble cedex, France

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Note presented by Édouard Brézin.

Abstract Nuclear fuel is the material in which the fission reactions occur. As such, it has to support many constraints induced by heat generation, fission product creation, as well as interaction with the coolant. These constraints are taken into account and fulfilled by the design. However, during irradiation, many changes are known to occur. Among them, irradiation damage, OD oxidation and changes in chemical or physical characteristics are to be considered. In addition, intermediate storage adds new constraints that the fuel has to support. With emphasis on the water reactor fuels, by far the most important group, a review will be given of major points, focusing on a few specific areas. ***To cite this article: C. Lemaignan, C. R. Physique 3 (2002) 763–772.***

nuclear fuel / engineering properties / irradiation effects / thermal behaviour / corrosion

Le combustible nucléaire : contraintes de conception, évolutions sous irradiation et impacts sur la fin de cycle

Résumé Lieu de la fission et donc de la source d'énergie, le combustible nucléaire constitue l'un des composants les plus critiques des réacteurs. Il présente d'autre part plusieurs caractéristiques prises en compte lors de la conception (génération de puissance thermique, création de produits de fission, contact avec le caloporteur…). Celles-ci évoluent au cours de l'irradiation (dommage d'irradiation, oxydation externe, évolution de composition et de propriétés…). Enfin le comportement après irradiation en réacteur, en particulier lors du transport et de l'entreposage, doit être considéré comme induisant une série de contraintes additionnelles qu'il doit pouvoir subir sans dommage.

En se focalisant sur le cas des combustibles des réacteurs à eau, très largement majoritaires, l'objet de cet article est de décrire les caractéristiques générales du combustible nucléaire et d'en illustrer en détail certains points particuliers. ***Pour citer cet article : C. Lemaignan, C. R. Physique 3 (2002) 763–772.***

combustible nucléaire / propriétés d'emploi / effets d'irradiation / thermique / corrosion

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E-mail address: lemaignan@cea.fr (C. Lemaignan).

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1. Introduction

In a nuclear reactor, fission is obtained in the fuel rods loaded in the core. In the case of water reactors, it consists of Zr alloy tubes filled with UO_2 or MOX pellets. For handling purposes the fuel rods are clustered in assemblies, in which the fuel rods are spaced in order to allow the water to flow around, acting as coolant and moderator. As the fissile material is transformed, the fuel rods are replaced annually by new fuel elements. After irradiation the fuel rods are considered either as waste, in the case of an open end fuel cycle, or as a potential reservoirs of energetic material in the case of reprocessing. In any case, their behaviour after irradiation for either medium- or long-term storage has to be considered.

The purpose of this paper is to give an overview of the general characteristics of nuclear fuel, its behaviour under irradiation and to analyse with deeper insight a few examples of scientific questions remaining open in this field of high technical impact.

2. General characteristics

Among the various aspects of fuel behaviour that have to be considered for the design of a reliable fuel, the following are of specific importance [1]: energy release and thermal behaviour; nuclear reactions and changes in chemistry (Pu conversion, fission product creation); behaviour of the Zr alloy claddings. These principal points are now introduced.

2.1. Thermal behaviour

A typical water reactor fuel rod generates fission energy leading to an average linear heat generation rate (LHGR) of $20\ \mathrm{kW \cdot m^{-1}}$. Due to the low thermal conductivity of the oxide, the centreline temperature is then in the range of 900–1000 °C, i.e. inducing a thermal gradient of more than $100\ \mathrm{K \cdot mm^{-1}}$. For such a brittle ceramic, this leads to the development of a network of cracks in the fuel pellets. During power changes, chips may relocate and induce local hard contact points in the inner side of the cladding. In addition, the very different temperatures, along the radius of the fuel pellet, activates or not various processes relevant to thermo-chemical processes. This will be the case for the important mechanisms of fission gas release, described later.

2.2. Nuclear reactions and changes in chemistry

The current design burn-up (BU) of the fuel rods frequently exceeds $50\,000\ \mathrm{MW \cdot d \cdot} t_{\mathrm{U}}^{-1}$ for UO_2 and is aimed to be similar soon for MOX fuels. This is equivalent to the fission of 5% of the initial uranium atoms. For each fission, two new atoms are created, with a large spectrum of mass distribution, leading to a major change in chemistry of the fuel. Indeed, the initial fuel (UO_2 or MOX) is an oxide of very high purity (less that 100 ppm total impurity for current fuel oxides), and, after irradiation, will form a mixture of more than 35 chemical species, 20 of them having a significant contribution.

In addition, the fertile ^{238}U is continuously transformed into ^{239}Pu. This has two consequences: a positive one is to reduce the loss of reactivity of the core as the ^{235}U is burned (for instance, at the end of life, half of the power is due to the fission of the Pu atoms), and a less favourable one is the unavoidable remaining Pu content after irradiation.

2.3. Zr alloy cladding

Zr alloys are always used as cladding and structural materials in thermal reactors, due to their low neutron capture cross section, good corrosion resistance in the reactor environment, and satisfactory mechanical properties. However, irradiation induces major changes in the microstructure of these alloys, with significant impact on their behaviour. Among these, an amorphous transformation of the precipitates, known to be responsible for the initial good corrosion resistance, is observed after one to two years of irradiation. This phase transformation is associated with a release of Fe from the precipitates to the matrix, enhancing the formation of a new type of dislocation loop, leading to a large increase in the growth rate. This growth is a length increase under irradiation induced by the anisotropic distribution of the irradiation induced dislocation loops, and macroscopic strain. In the case of the Zr alloys under irradiation, a specific corrosion mechanism can be observed in BWR environments, the so-called ‘shadow corrosion’. It will be described in detail later.

3. Scientific aspects of specific behaviours

In this section, only a limited number of processes having significant industrial or scientific interest will be described.

3.1. Fission gas release

Among the various fission products, gases amount for a large part (about 2 at.% at a BU of 50 000 $\mathrm{MW{\cdot}d{\cdot}}t_{\mathrm{U}}^{-1}$). The rare gases are non-soluble in the fuel oxides and the high content induced by the fuel irradiation leads to a super-saturation of them. Typically, for standard BU, the fission gases trapped in solution within the UO_2 amount about 15 cm^3 STP per cm^3 of UO_2. The high driving force for their escape from the fuel is counter-balanced by the very low diffusivity of these gases at the operating temperatures of the fuel, making the oxide ceramic a good storage medium for the fission gases. However, significant increases in the pellet temperature, as induced by a local increase in fission power, may allow the activation of atomic transport, and the diffusion of Xe atoms from the bulk of the grains to the grain boundaries. Depending on the local temperatures, this process is more or less efficient. As an example, Fig. 1 describes the differences in morphology observed using an SEM in a hot cell laboratory after irradiation, at two different locations along the radius of a pellet, power ramped to a LHGR of 34 $\mathrm{kW{\cdot}m^{-1}}$. In the center of the pellet the fuel reached a temperature of 1650 °C and a clear precipitation of Xe bubbles is observable at the grain boundaries, allowing the formation of a percolating network of 3D-channels surrounding the triple boundary lines [2]. In order to reduce the fission gas release induced by such atomic diffusion processes, advanced microstructures, with large grain sizes may be used. This could be obtained either by longer sintering times, a method hardly acceptable to manufacturers, or by very small amounts of additives and sintering in controlled atmospheres [3].

At lower temperature, other processes for fission gas release have to be considered, that are less controlled by thermal activation: under the flux of neutrons and fission products, two ballistic mechanisms are operating. The diffusion is enhanced under irradiation, but, in addition, specific release mechanisms occur at the free surfaces. Indeed, as the fission products leave the oxide by recoil, sputtering occurs, allowing a few hundred atoms around the emergence point to be released into the He filled gap. Recent studies have analyzed in detail this process, using heavy ions to simulate the fission products and have shown a strong dependency of the sputtering yield on the electronic stopping power of the ions, without a very clear understanding of the details of the process [4].

3.2. The RIM structure

For high BU in water reactors (above 40 000 $\mathrm{MW{\cdot}d{\cdot}}t_{\mathrm{U}}^{-1}$), the outer area of the pellet is observed to change in microstructure, with the disappearance of the initial grains and the development of a new, very fine microstructure, associated to sub-micron porosities [5,6]. 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 pellets. This increases the local concentration of fissile isotopes with higher Pu content, giving rise to a higher BU in this area. Using particular fission products, such as Nd as local BU tracers, typical values above 200 $\mathrm{GW{\cdot}d{\cdot}}t^{-1}$ have been measured at the outer surface, decreasing to the mean value a few hundred microns inside. The analysis of the formation mechanism of this fine grain microstructure by TEM confirms a continuous

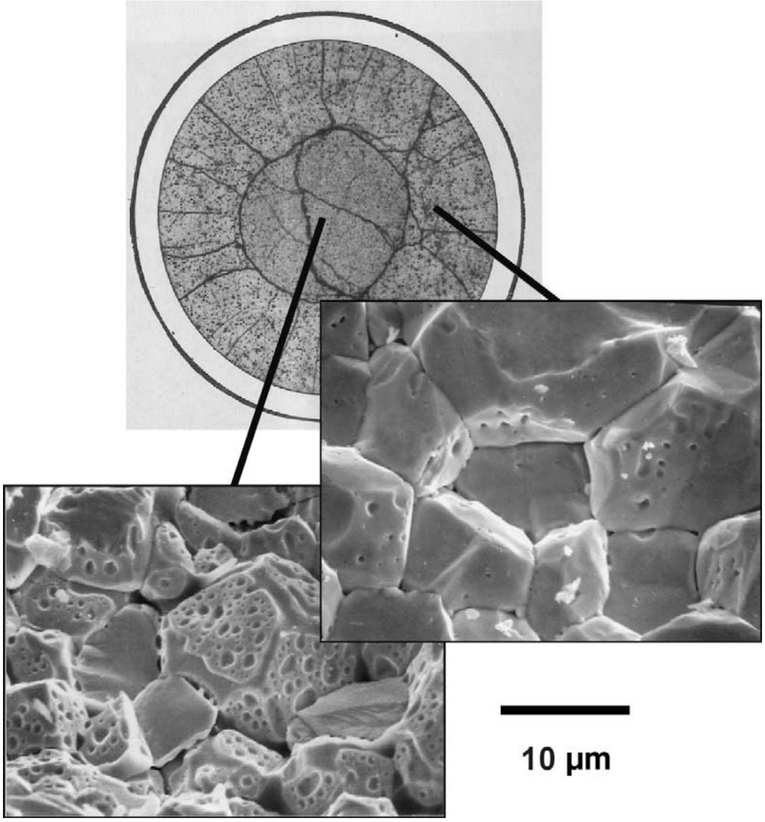

Figure 1. Mechanisms of fission gas release after a power transient, as observed by SEM after irradiation.

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 these intra-granular sinks formed at the beginning of irradiation [7].

3.3. Specific aspects of mixed oxide fuels (MOX)

Currently used for reloads in Europe at an industrial scale, the mixed oxide fuels have properties and behaviours close to those of regular UO_2 fuels. Differences arise mainly from the heterogeneous microstructure obtained by the 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 the cold outer parts of these fuels, the Pu rich agglomerates behave like the RIM in 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 for the pellets. In addition to these microstructural effects, the neutron physics behaviour of the MOX fuels leads to higher power generation at intermediate BUs, and thus to higher centreline temperatures. At the higher temperature centre zone, the diffusion processes are effective and the precipitation of fission gases as bubbles and other fission products as small precipitates may be observed (Fig. 2). These are the reasons, with the higher oxygen balance of the Pu fissions, for the higher fission gas release observed in MOX fuel [8].

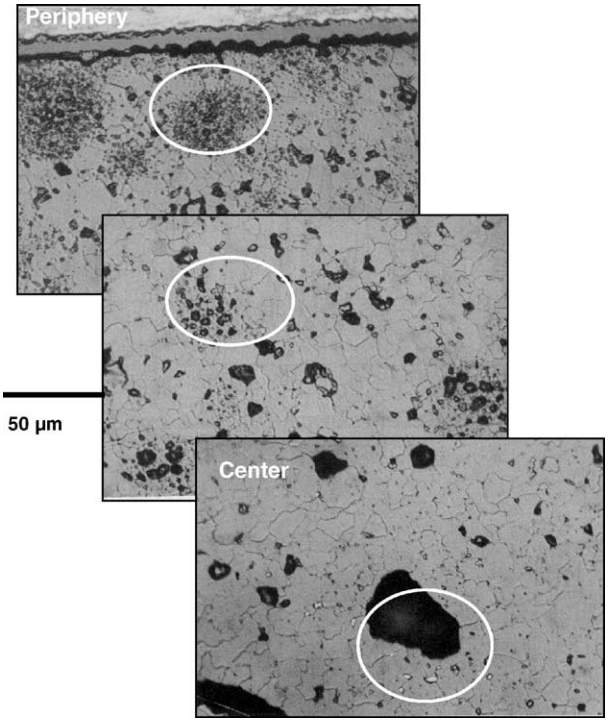

Figure 2. Changes in fission gas release and microstructure of the MOX around the Pu rich aggregates.

The behaviour of these Pu rich areas allow one to anticipate the behaviour of UO_2 at very high BU or the situation of an oxide irradiated with heavy ions, like the inert matrixes, such as the Cer–Cer fuel considered for minor actinide transmutation.

3.4. Effects of irradiation on Zr alloys

Under irradiation, the damage induced in the cladding and in the growing Zr oxide is also to be considered: in Zircaloy's, 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 (Fig. 3). 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 $\langle c\rangle$ 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 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, the dislocation loops obtained are of $\langle a\rangle$ type, located in the prismatic planes. The result is a limited growth (anisotropic strain at constant volume). For high doses, the amorphous transformation of the precipitates occurs, with a simultaneous release of alloying elements in the matrix [9]. The resultant high concentration of iron in the matrix has been shown to be liable for the formation of additional $\langle c\rangle$ type loops [10]. Opposite to the $\langle a\rangle$ loops, they are very large and are only of vacancy type. Their formation, normally not seen in low c/a hcp metal, induces an accelerated growth clearly observed for high burn up recrystallized Zircaloy.

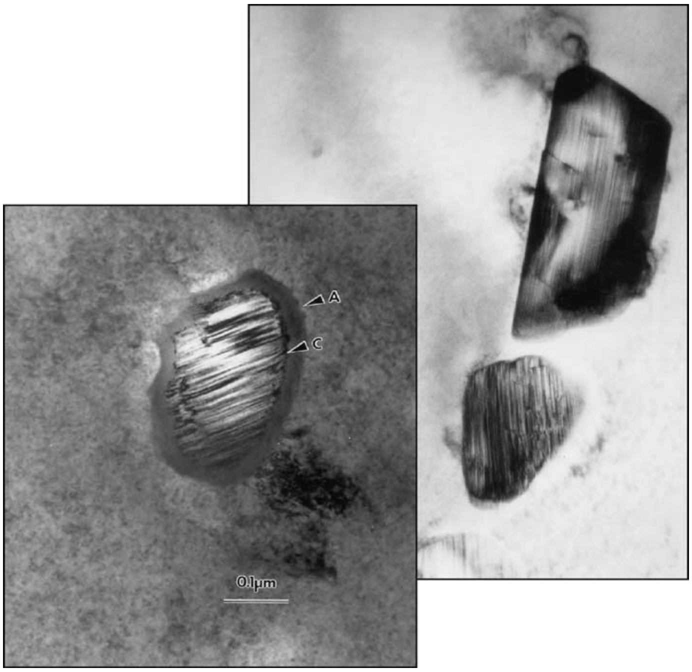

Figure 3. Irradiation induced amorphous transformation of the intermetallic precipitates in Zry-4 (back: unirradiated, front: after irradiation).

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 the dislocation loops. During plastic strain, these loops can be annealed by interaction with the gliding dislocations [11,12]. 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 circumferentially. 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 the transverse loading direction. Therefore, axial or circumferential uniform strains of irradiated Zr alloys are found to be different. Fig. 4 gives an example of such dislocation channelling and localized deformation [13]. This contributes to the reduced uniform strain of irradiated Zr alloys.

3.5. Zr alloy cladding corrosion

Under irradiation, the corrosion rate of zirconium alloys is significantly increased and constitutes one of the main limitations to the use of high burn-up fuel rods in Light Water Reactors (LWR) [14]. The oxidation process is usually described in two steps, with a transition between a cubic to a linear oxidation rate after a few micrometers of zirconia. Many research teams have pointed out the role played by two different types of parameters: the metallurgical ones directly correlated to the cladding microstructure and those specific to reactor environment, such as the coolant chemistry or the neutron and gamma fluxes.

It was found that the size, chemical composition and distribution of intermetallic precipitates $Zr(Fe, Cr)_2$ and ($Zr_2(Fe, Ni)$ in Zircaloy-2) significantly alter the oxidation process by retarding or anticipating the transition of the oxidation process or by enhancing pre- and post-oxidation rates [15–17]. This is the reason why the neutron irradiation, modifying the microstructure of these precipitates, affects the resistance to

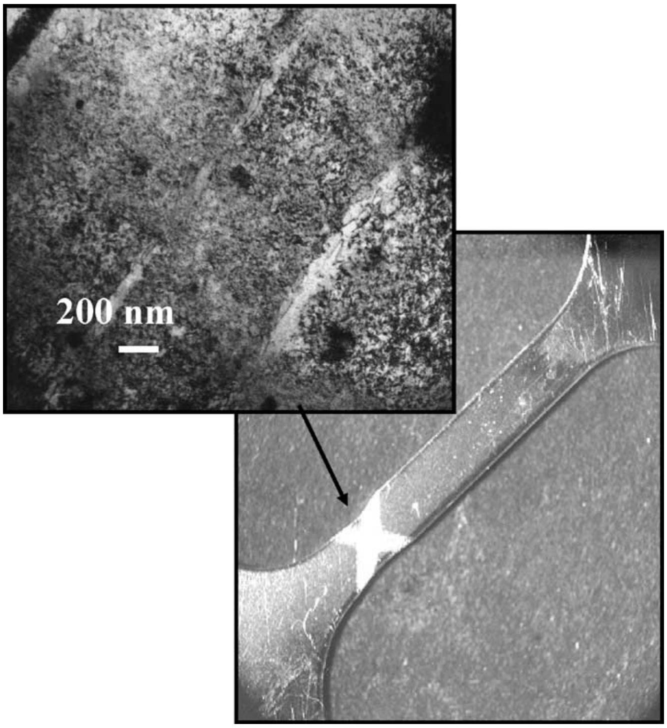

Figure 4. Localized plastic strain, at microscopic and macroscopic scale, in Zry-4 after irradiation.

water side oxidation. In addition, the reactor coolant chemistry, and more precisely Li content, appears to have a great influence either on the oxidation rate [18,19] or on the oxide structure [20].

Typical values of the outer ZrO_2 layer thickness after irradiation may be above 50–70 µm, for a design limit corresponding to about 100 µm. In order to avoid operation limitations, new alloys are developed, such as Zr 1% NbO ($M5^{TM}$), where the kinetics transition is reduced, as well as the overall corrosion rates.

A specific type of corrosion has been observed in different BWR reactors. The ‘shadow corrosion’ is observed in Zr alloys near another metallic alloy, such as stainless steel. The oxide thickness may there be much higher than just a few centimetres away, where the thermo-chemical conditions are similar (Fig. 5). A detail review of all the cases of the shadow corrosion occurrence stressed our poor understanding of the physical phenomena involved in the process. To occur, shadow corrosion requires a synergy between a cathodic electrochemical potential, possibly modified by electron emission due to β^- decay, an oxygen rich environment, a strong irradiation with β^- or γ, and a close geometry with the facing alloy. The various experiments performed in and out of reactors, in order to better understand the process, hardly led to reproducible results and pointed out our lack of understanding of the various interactions between these phenomena [21].

3.6. Zr alloy cladding hydriding

In conjunction with corrosion, hydrogen ingress (called H pick up) is a phenomenon affecting the Zr alloys in reactors. An average of 10 to 50% of the hydrogen released by the reduction of water is indeed trapped by the Zr alloys. The mechanisms involved are not perfectly understood as the zirconia layer is expected to be a perfect diffusion barrier for the protons. Diffusion mechanisms through intermetallic precipitates or other paths are under investigation. Hydrogen solubility in Zr alloys is strongly temperature dependent, being larger than 200 ppm at 350 °C and below 1 ppm at room temperature. The result is that

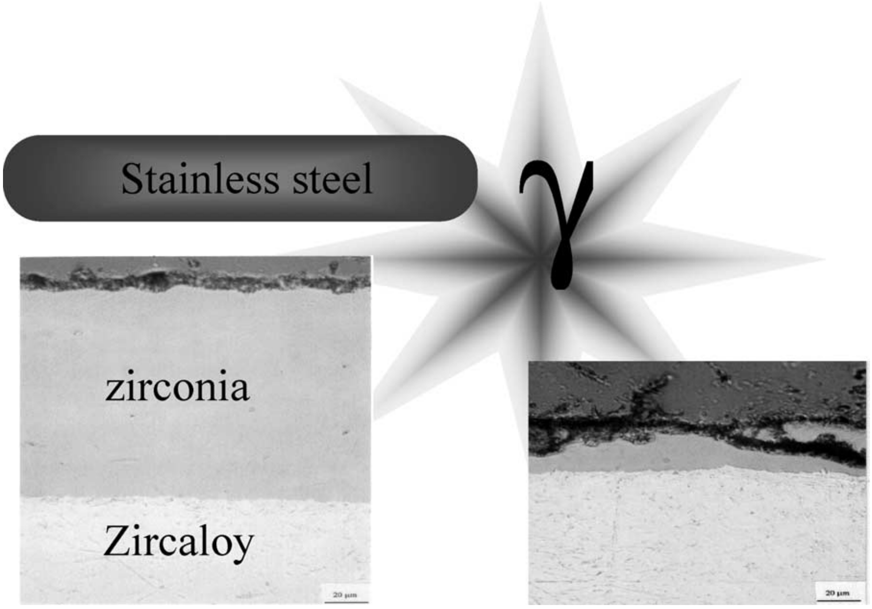

Figure 5. Irradiation enhanced localized corrosion (shadow corrosion) in front of a stainless steel plate (BWR environment).

zirconium hydrides ($ZrH_{1.66}$) precipitate when the reactor is cooled. Due to the brittle nature of this phase, a high hydrogen concentration drastically reduces the ductility at room temperature of irradiated and oxidized Zr alloys. This can be detrimental for handling of the fuel assemblies after irradiation.

On the other hand, it has been shown that a high concentration of hydrogen reduces creep rate after irradiation. This is a positive point, with respect to the design limitations of the stain occurring during transport in dry casts from the plant to the intermediate storage location, as the fuel could reach temperature high enough to induce significant creep during the corresponding time.

4. Elements of conclusions

In order to satisfy the high reliability of the fuel elements in power reactors, only minor improvements are implemented step by step in the fuel design and fabrication. Indeed large technological jumps from proven technical choices could lead to the occurrence of unexpected physical events. These potential problems have clearly to be considered, as the metallurgical processes induced by irradiation are known to arise frequently after a given threshold dose, as it has been observed in the case for the swelling and the growth.

Under irradiation, the fuel materials – both cladding and fuel – are subject to various transformations that affect all the properties of the components of the fuel rods. A safe design of the fuel elements should take into consideration all these transformations, with additional margins as provisions for unexpected events, in the case of tentative extensions of the irradiations conditions.

A new common trend is to include in the service life of the fuel elements, the back end of the fuel cycle. This new constraint is limited to the behaviour in the time span following directly the irradiation, i.e. transport and intermediate storage. Usually, the new operating conditions are weak compared to the irradiation in the reactor itself, but particular conditions may require additional knowledge, such as during a dry transport.

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Discussion

Question de C. Madic

La shadow corrosion du Zircaloy a-t-elle – pu être reproduite en laboratoire ?

Réponse de C. Lemaignan

Plusieurs tentatives ont eu lieu pour reproduire les caractéristiques de la « shadow corrosion ». Si certains aspects de cette corrosion localisée ont parfois pu être dupliqués, il semble que cette forme de corrosion nécessite une synergie entre une irradiation intense (au moins γ, mais très probablement β^- aussi, voire sous neutrons), des conditions oxydantes dans le caloporteur, et un potentiel électrochimique anodique. Il est extrêmement difficile de reproduire l'ensemble de ces conditions hors d'un réacteur. Une revue récente de ces points peut être trouvée en réference [21].

Question de C. Devillers

Quelles assurances a-t-on de la tenue de la gaine des combustibles usés dans une piscine d'entreposage à long terme ?

Réponse de C. Lemaignan

Pour les gaines en alliages de Zr, les conditions présentes en piscine d'entreposage ne correspondent pas à un milieu agressif, par rapport à celui qu'elles ont eu à subir en réacteur. Les vitesses d'oxydation attendues à 100 °C restent de l'ordre de 10^{-3} $\mathrm{nm \cdot an^{-1}}$, ce qui conduirait à des épaisseurs d'oxydation non mesurables. Tout au plus y aurait-il à considérer les dommages mécaniques induits lors de manœuvres brutales, en raison d'une fragilisation à basse température induite par la précipitation d'hydrures de Zr.

Question de É. Brézin

La durée de vie des pastilles est-elle limitée par le taux d'atomes fissionnés, ou par l'oxydation des gaines de zirconium ?

Réponse de C. Lemaignan

Le crayon combustible constitue une entité en lui-même, dont il est pratiquement impossible de séparer les limitations imputables à un composant particulier. L'un des rôles du concepteur est justement d'optimiser un compromis entre les limitations des composants pour une utilisation dans un réacteur particulier. Ainsi, pour une conception donnée, la durée de vie maximale du crayon sera limitée par le relâchement des gaz de fission dans un réacteur particulier, alors que dans un autre réacteur, plus chaud, elle le sera par l'épaisseur maximale de zircone.

Question de É. Brézin

L'effet des gaz de fission est-il plus important dans le MOX ?

Réponse de C. Lemaignan

Il est observé que le taux de sortie des gaz de fission hors de la pastille combustible est plus élevé dans les MOX que dans l'UO_2, et en particulier que l'accélération du taux de relâchement apparaît pour un taux de combustion plus faible. Ceci est lié à deux phénomènes particuliers : d'une part la microstructure du MOX, étant hétérogène, conduit à un très fort taux de combustion local dans les agrégats de « mélange mère », pour le même taux de combustion moyen, augmentant la force motrice thermodynamique de sortie, et, d'autre part, les historiques d'irradiation obtenus avec ces combustibles, conduisent à de plus fortes puissances linéiques, favorisant, à même taux de combustion, un relâchement plus important.