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Journal of Nuclear Materials 352 (2006) 246–253

journal of nuclear materials

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Specific outcomes of the research on the spent fuel long-term evolution in interim dry storage and deep geological disposal

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Abstract

This paper presents an overview of the main results of the French research on the long-term evolution of spent fuel. The behavior of the spent fuel rods in the various conditions likely to be encountered during dry storage and deep geological disposal, i.e., in a closed system, in air and in water were investigated. It appears that in a closed system the effects of helium production on the mechanical stability of grain boundaries remain the major unanswered question. In air, microscopic characterization of the UO_2 oxidation leads to introduce a new phase in the classical oxidation scheme. The limiting step assumption on which the oxidation kinetics are based is only partially valid. In water, the effect of the alpha radiolysis which accelerates UO_2 dissolution was demonstrated for anoxic conditions. However this effect could be counteracted by the environmental conditions, such as the presence of H_2 produced by the container corrosion. The effects of the environmental parameters on the fuel matrix dissolution still need to be assessed.

PACS: 28.41.Kw; 23.60; 82.50.Gw; 83.50.Bw

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1. Introduction

For the interim storage of long-lived radioactive waste, the practical concern is the retrievability of fuel assemblies after the period of storage, the feasibility of treatment in facilities such as those at La Hague (COGEMA) and the compatibility with ultimate disposal. Moreover, the safety analysis of the storage facility requires definition of the radionuclide source terms. This is also true for geological disposal. In this case, the release of radionuclides versus time must be known in order to evaluate the radiological impact on the environment. Most of research in France on that topic is integrated in the PRECCI project (acronym of *Programme de Recherches sur l'Evolution à long-terme des Colis*

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doi:10.1016/j.jnucmat.2006.02.061

de Combustibles Irradiés). Its aim is to investigate the spent fuel behavior under the various boundary conditions encountered during storage and geological disposal. These are:

- The evolution in a closed system. This corresponds to the nominal scenario during storage and to the first confinement phase during disposal. The effects of residual temperature and high radioactivity on the chemical and physical properties of the spent fuel were evaluated. The studies focused also on the evolution of the irradiated cladding during transport and long-term dry storage, due to creep and hydride re-orientation upon cooling under internal pressure.
- The evolution in air. This condition corresponds to the incidental loss of confinement during dry storage or to a rupture of the canister before the site re-saturation in geological disposal. UO_2 is thermodynamically unstable in air. The formation of the U_3O_8 phase which leads to significant swelling could damage the fuel rod and enhance release of radionuclides to the atmosphere.
- The evolution in water. In reducing conditions as expected in deep geological disposal, dissolution of UO_2 is controlled by the low solubility of this solid phase. However, radiolysis of water caused by residual spent fuel radioactivity generates oxidants which by creating local oxidizing conditions at the water/fuel interface could accelerate matrix dissolution.

The major difficulties inherent to the long term behavior are the time frames of interest: some decades for dry storage but over 100000 years for geological disposal. These time frames are not directly accessible through laboratory experiments. The studies performed in the PRECCI project were based on various experimental methods coupled with modeling, from the macroscopic to the microscopic point of view in order to depict the mechanisms of evolution which can be extrapolated to the time frame of interest [1]. This paper presents an overview of the major results obtained for each scenario.

2. Evolution of spent fuel rods in a closed system

The chemical and physical properties of the spent fuel may evolve even without external exchange of matter due to the temperature which remains relatively high during dry storage and to the high radioactivity. Three main processes of evolution of the ceramic fuel were identified at the beginning of the project:

(1) Chemical evolution associated to β disintegration which produces elements with different valence states and the temperature decrease could change the oxygen potential in the spent fuel.
(2) Release of radionuclides from the spent fuel grains by diffusion. This process could enhance the inventory of radionuclides rapidly released when water contacts the fuel.
(3) Physical evolution associated with helium production and accumulated damage due to α-decay.

The metal cladding properties may also evolve at the temperature of dry storage due to creep under internal pressure and to hydride reorientation during cooling under stress. The internal pressure in the fuel rod should decrease in time with the decrease of temperature and creep deformation. However, these effects could be counteracted over the long-term by potential release of helium and fission gases to the void volume of the fuel rod.

2.1. Evolution of the spent fuel pellet

Thermodynamic calculations have shown that the oxygen potential is relatively unaffected by the β-decay and diminishing temperature, but is buffered by oxidation of Mo, one of the most abundant fission products and the presence of the internal zirconia layer in MOX fuel. The buffering role of Mo was confirmed by its valence state revealed by X-ray absorption spectroscopy of implanted UO_2 pellets [2].

2.1.1. Diffusion of radionuclides in the spent fuel grains

In accordance with the results obtained by Prussin et al. [3] at high temperatures ($T > 1973$ K), the fission products can be classified in order of decreasing mobility as follows: (1) electronegative elements which are insoluble in UO_2 such as I and Te, (2) non electronegative insoluble elements such as Cs, Xe and (3) non electronegative soluble species in UO_2. Lanthanides and actinides belong to the third class.

Studies were performed to check the validity of this classification at lower temperatures. Diffusion profiles in implanted UO_2 pellets were determined

Table 1
Diffusion coefficients in UO_2 grains ($m^2\ s^{-1}$) for fission products

Element	Temperature (K)			
	1273	1223	1173	1073
O [5]	1.3×10^{-15}	5.0×10^{-16}	1.7×10^{-16}	1.6×10^{-17}
^{127}I [6]	1.5×10^{-19}		6.0×10^{-20}	5.5×10^{-21}
^{125}Te [6]	1.6×10^{-19}		2.0×10^{-20}	1.3×10^{-21}
^{133}Cs [6]	7.0×10^{-21}	1.6×10^{-21}		
Xe [4]	8.0×10^{-21}	2.4×10^{-21}	5.0×10^{-23}	
U, Pu [7]	3.2×10^{-28}	6.3×10^{-29}		

by secondary ion mass spectroscopy after annealing at temperatures between 1073 and 1273 K [4]. The results corroborate Prussin et al. classification (Table 1). Extrapolating the Arrhénius laws deduced from these data to lower temperature (<673 K) shows that the thermal diffusion process can be considered as negligible within the time frame of interim storage (a hundred years) or disposal (>10000 years) and at the expected temperatures. The maximal temperature at the beginning of storage is lower than 673 K and rapidly decreases with time to reach a value lower than 473 K after a few decades. For disposal, the expected temperature is lower than 473 K.

Two mechanisms that can be described by Fick's diffusion account for the mobility of fission products in the reactor: One is temperature-driven and the other is due to the effect of fission. The effects of α radioactivity in UO_2 are very similar to those of fission. By analogy with what is observed in the reactor, the existence of a diffusion process due to the effects of α decay was examined. This mechanism should be lower by some orders of magnitude than the one measured under fission in reactor. As the diffusion coefficients are expected to be lower than the available detection limit (about 10^{-21} $m^2\ s^{-1}$), direct measurements of the diffusion enhanced by α self-irradiation is not possible within the time frame of laboratory experiments. Various theoretical approaches have been developed to enable rough estimates to be made. A first approach was based on the extrapolation from in-pile data given in Ref. [8]. It led to a diffusion coefficient $D^*_\alpha (m^2\ s^{-1})$ which is supposed to be proportional to the volume α activity A_α ($Bq\ m^{-3}$) as [9]

$$D^*_\alpha \approx 2 \times 10^{-41} A_\alpha. \qquad (1)$$

The mean value of the diffusion coefficient of fission gases deduced from this relationship is 10^{-25} $m^2\ s^{-1}$ during the first hundred years of cooling for a UO_2 fuel with a burnup of 60 MWd kg^{-1} (of initial heavy metal). It then decreases with time as the α activity declines indicating a limited effect of diffusion. Moreover, other models based on atoms enhanced mobility induced by the α particle due to the electronic excitation and by the recoil atom due to generation of displacement cascades lead to a proportionality factor between D^*_α and A_α which is about three orders of magnitude lower [10].

2.1.2. Physical evolution of the spent fuel pellet

The physical properties such as the density of the UO_2 pellet evolve in reactor due to formation of fission products and fission gases (Kr and Xe) which cause solid and gaseous swelling. Some fragmentation of the fuel pellet occurs also in reactor and is essentially due to the thermal gradient which develops in the pellet under irradiation. Out of reactor, the thermal gradient is negligible but solid swelling due to α decay accumulated damage and gaseous swelling induced by the production of helium may change the fuel pellet properties over time.

The irradiation damage generated in the UO_2 matrix is due mainly to displacement cascades created by α radioactivity, especially by the recoil nucleus emitted with each disintegration. From a review of literature data illustrating the lattice parameter evolution versus the mean number of displacements per atom (Fig. 1), the cumulative effect of irradiation damage is limited in the UO_2 matrix because of extensive annealing of the defects. No amorphization is expected [15] and the value of the lattice parameter should remain constant for damage exceeding an equivalent of about 1 displacement per atom. The saturation value of the lattice parameter (about 0.4%) corresponds to a relative volume increase of about 1%. Following irradiation damage in reactor, saturation of the lattice parameter should occur very rapidly after the fuel is unloaded from the reactor.

Gaseous swelling depends on the helium behavior in the irradiated fuel particularly in MOX fuel where its concentration reaches about 1 at.% after 10000 years for a burnup of 47 MWd kg^{-1}. Studies on unirradiated UO_2 to determine the helium solubility [16–18] and its diffusion coefficient [19] indicate a low solubility limit of He (<1 at.%) and a thermal diffusion coefficient lower than 10^{-26} $m^2\ s^{-1}$ for temperatures below 573 K. The present state of knowledge suggests that in a first step, helium should accumulate in intragranular bubbles possibly causing microcracks. This phenomenon should accelerate the helium release to grain boundaries and by increasing

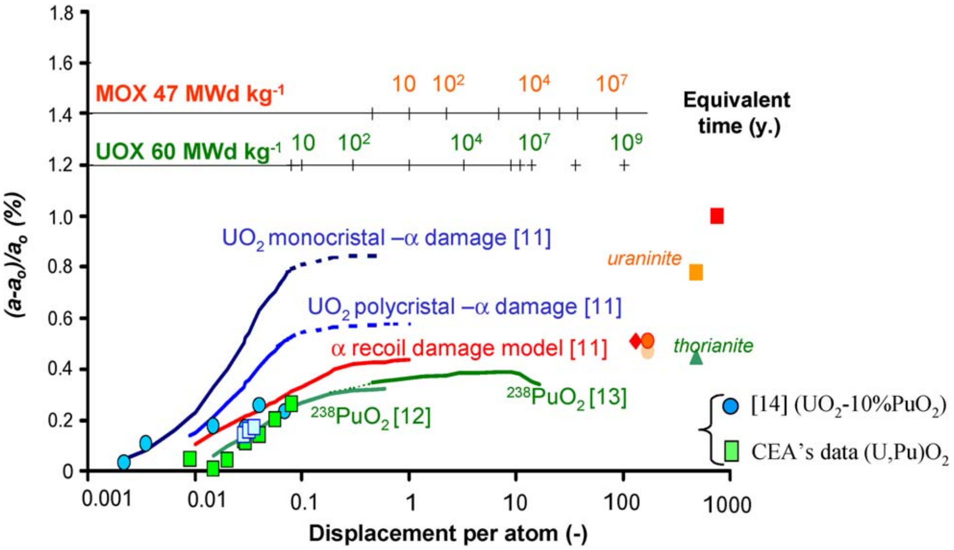

Fig. 1. Variation of the lattice parameter of UO_2, PuO_2 and $(U,Th)O_2$ matrices versus the mean number of displacements per atom for different irradiation sources: α alone, α + recoil nucleus. The correspondence with the time after removal from the reactor is also indicated for UO_2 fuel (60 MW d kg^{-1}) and a MOX fuel (47 MW d kg^{-1}) [11–14].

the preexisting fission gas bubble pressure lead to opening of grain boundaries as observed in old PuO_2 pellets (Fig. 2). The long term fate of the helium in the spent fuel is the major open question concerning the behavior of spent fuel in a closed system.

2.2. Evolution of the cladding properties

Short term creep tests (up to 60 days) led to propose creep laws for CWSR Zircaloy-4 [20,21]. However, these laws underestimate the creep deformation observed in the most severe conditions i.e. longer time and higher temperature or stress. Transmission electronic microscopy observations have revealed a partial annealing of the irradiation defects (⟨*a*⟩ loops recovery) during creep which should explain the gain of cladding ductility. These analyses allowed development of a new formulation of the creep law taking into account this mechanism with an extended domain of validity compared to the former laws (Fig. 3).

The conditions under which hydride reorientation occurs in Zircaloy-4 during cooling under internal pressure were studied as a function of stress and temperature. The influence of hydride orientation

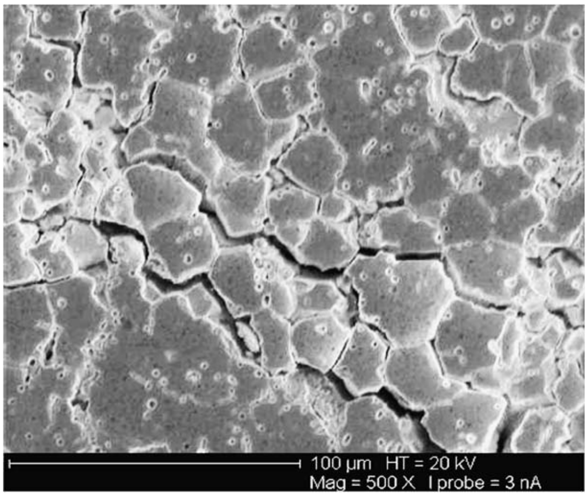

Fig. 2. Scanning electron microscopy analysis of $^{238}PuO_2$ pellet (after cutting and polishing) stored during 30 years of storage in inert atmosphere (D. Roudil: personal communication).

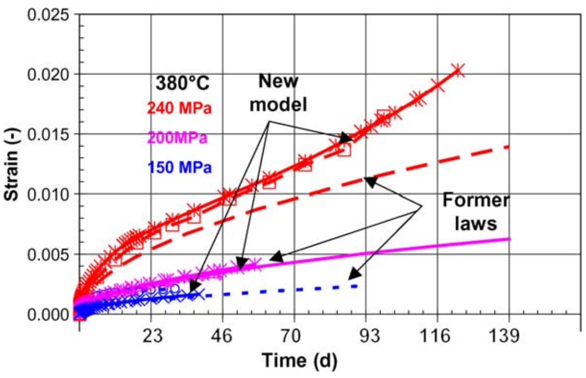

Fig. 3. Comparison between creeps tests and models (at 653 K and for circumferential stress of 150, 200 and 240 MPa). The former laws are described in [20] and [21].

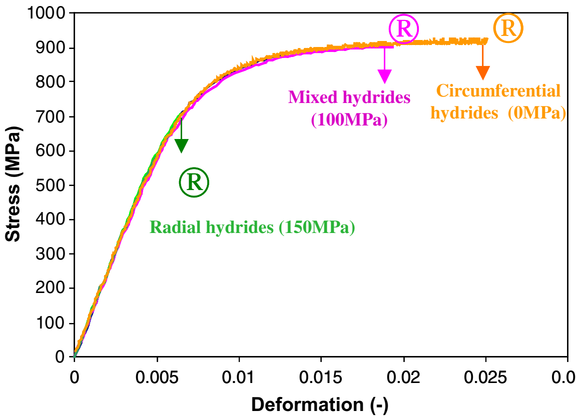

Fig. 4. Comparison of bursting failure stresses at 293 K versus the hydride orientation for a hydrogen concentration of 200 ppm in Cold Worked Stress Relieved Zircaloy-4. (® indicates a rupture).

on the intrinsic brittleness of the unirradiated cladding material at low temperature has also been investigated by conventional burst tests and by more elaborate bending tests simulating a handling incident during retrieval of spent fuel assemblies. The results obtained with both types of tests show that the hydride orientation has no effect on the material strain law. However, the burst tests revealed a lower circumferential deformation at failure that was more notable with higher radial hydride fractions, indicating embrittlement of the material (Fig. 4).

3. Evolution of the defective spent fuel rod in air

UO_2 is thermodynamically unstable in air. The formation of the orthorhombic phase U_3O_8 which causes an important volume swelling of about 36% can lead to the ruin of the defective spent fuel rod. Oxidation kinetics based on the thermogravimetric analysis (TGA) which assumes the existence of a limiting step during oxidation reaction lead to Arrhénius type laws [22,23].

Under long-term dry storage, the expected temperature of spent fuel rods after a few decades is below 473 K. Microscopic studies were performed on unirradiated UO_2 in order to determine the mechanisms of oxidation and to check the validity of kinetic laws in the temperature range of storage. The approach combined in-situ X-ray diffraction analysis with TGA during oxidation of UO_2 powder to analyze structural evolution versus the macroscopic O/U ratio. The validity of a rate limiting step was verified by applying a method based on jumps of temperature [24,25]. According to the limiting step assumption, the ratio of the reaction rates before and after a rapid temperature increase depends only on the temperature difference and is constant regardless of the degree of reaction progress. The kinetic study indicates four separate kinetic domains and only two where the rate-limiting step approximation is applicable. Moreover the high resolution of the synchrotron diffraction revealed a four transition sequence from UO_2 to U_3O_8 with the formation of the secondary cubic phase U_4O_{9-y} at the earliest stages of oxidation [26]:

$$UO_2 \rightarrow U_4O_9 \rightarrow \beta\text{-}U_3O_7 \rightarrow U_3O_8.$$

A first modeling approach of the oxidation of UO_2 powder based on these observations was proposed in [27].

In parallel, oxidation tests were carried out at 473 K on UO_2 and MOX fuel fragments with a burnup of 41 MWd kg^{-1} to compare the behavior of the two types of fuel at the same burnup (Figs. 5a and 5b). The data show that in UO_2 fuel the weight gain remains parabolic up to 1600 h, which is coherent with the oxidation kinetic proposed in the literature [28]. Then it varies only slightly and in a nearly linear manner up to 6000 h. Electron microprobe analysis confirmed that U_3O_8 did not form during this oxidation interval. By analogy with the observed behavior of unirradiated UO_2, the linear weight gain phase can be attributed to the formation of U_3O_7 or to oxygen incorporation in nonstoichiometric U_4O_9, for which the existence range is extended by the presence of fission products.

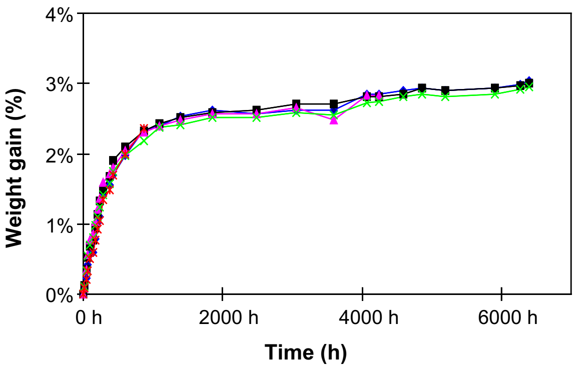

Fig. 5a. Measured weight gain of UO_2 fuel fragments irradiated at 41 MW d kg^{-1} as a function of heating time (473 K in air with a relative humidity of 30% at 293 K). The experimental curves correspond to oxidation tests performed on different fragments of the same spent fuel pellet.

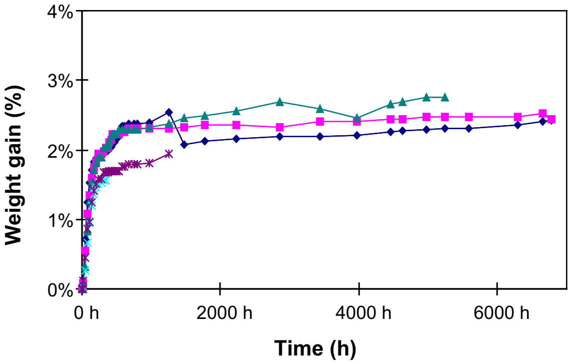

Fig. 5b. Measured weight gain of MOX fuel fragments irradiated at 41 MW d kg^{-1} as a function of heating time (473 K in air with a relative humidity of 30% at 293 K). The experimental curves correspond to oxidation tests performed on different fragments of the same spent fuel pellet.

In MOX fuel a distinction must be made between the UO_2 matrix and the Pu-rich aggregates. The UO_2 matrix appears to be completely transformed into U_3O_7 after 400 h of heat treatment in MOX, whereas 2500 h are necessary for UO_2 fuel. Conversely, the Pu-rich aggregates are not completely oxidized after 1255 h of heat treatment, and the onset of the second step appears to be delayed with respect to UO_2 fuel. In samples analyzed between 5000 and 6000 h of oxidation, the oxygen concentration was higher in the Pu-rich aggregates than in the matrix. These results show that the conversion of UO_2 to U_4O_9 is slower at high Pu concentrations as observed in Pu-rich aggregates of MOX fuel or at high burnup values. This is consistent with the observations that extraneous elements in the matrix tend to retard its oxidation. They also show a greater oxygen incorporation capacity when Pu concentrations or burnup values are high.

Metallographic examination during oxidation of spent fuel showed that some grains in UO_2 fuel are oxidized to mid-radius while others are fully oxidized. The oxidation kinetics thus appear to be controlled by a complex oxidation front inside the fragment, but not inside the grains. After 6000 h of oxidation, the UO_2 fragment is cracked, but the cracking occurs inside the fragment. No satisfactory explanation is currently available for this internal cracking. Microprobe observations of UO_2 spent fuel show that complete conversion to U_3O_7 (O/M = 2.4) does not modify the fission product distribution after up to 6000 h of oxidation at 473 K: they remain uniformly distributed in the matrix.

The effect of the cladding defect which should limit the access of oxygen and the release of volatile radionuclides is currently under investigation.

4. Evolution of the spent fuel in water

The environmental conditions in deep geological disposal are reducing. In these conditions, UO_2 is thermodynamically stable. However, radiolysis of water at the surface of the spent fuel can create local oxidizing conditions which accelerate the dissolution of the fuel matrix. Studies performed in the project first focused on the effect of the water radiolysis on the fuel alteration. After about 1000 years, the α activity is dominant in the fuel (see for example [29]). Two experimental methods have been explored to understand and evaluate the impact of α radiolysis of water on fuel matrix dissolution. One involves exposing an undoped UO_2/water system to external α radiation. The other uses UO_2 pellets doped with α emitters at different concentrations to reproduce the α flux emitted at the surface of the spent fuel with various ages.

External irradiation experiments at high flux values were carried out using the CERI–CNRS cyclotron in Orléans, which generates a $^4He^{2+}$ ion beam with an initial energy of 45 MeV. The α particles passed through a uranium oxide disk of a thickness adjusted to obtain an emission on the surface with an energy of about 5 MeV [30,31]. The experiments were carried out in deionized water at pH 6, in aerated (2×10^{-4} M dissolved oxygen) and deaerated (1.2×10^{-10} M dissolved oxygen) media. The solution was renewed after each one-hour irradiation cycle at constant α flux (between 3.3×10^{10} cm^{-2} s^{-1} and 3.3×10^{11} cm^{-2} s^{-1}). The uranium and hydrogen peroxide concentrations in solution and the pH were measured at the beginning and end of the experiment. In both conditions (aerated and deaerated), the uranium concentration in solution increased by three to four orders of magnitude under irradiation. The higher the flux, the higher the uranium and H_2O_2 concentrations in solution and the greater the solution acidity. These findings were compared with those obtained simply by adding hydrogen peroxide to the solution to obtain the same bulk concentration as measured under external α irradiation (10^{-3} M). The uranium concentration measured under irradiation was two orders of magnitude higher than with H_2O_2 additive alone. This observation can demonstrate the role of the radicals produced by α radiolysis on UO_2 dissolution. However it could be explained by a local concentration of H_2O_2 in the water layer adjacent to the solid surface generated by irradiation which is higher than the mean bulk concentration. The characterization of the UO_2 disks surface by XRD revealed the presence of uranium peroxide which is related to the high concentration of H_2O_2 in these experiments.

Leaching of UO_2 pellets doped with α-emitters (^{238}Pu and ^{239}Pu) was also used to determine the effect of the α activity on UO_2 dissolution. In these tests, the ^{238}Pu and ^{239}Pu concentrations were adjusted to reproduce the α activity of spent fuel with a burnup of 47 MWd kg^{-1} at different time intervals (15, 1500 and 40000 years) after discharge. The total $^{238/239}$Pu content was maintained constant at 0.217 wt%, however, to avoid modifying the material chemistry. Leach tests were conducted at room temperature in a deaerated medium (<0.1 ppm O_2) and deionised water for 30 days. The results reported in [32] indicate a correlation between the α flux at the reaction interface and the rate of uranium release in solution. The effect of gamma irradiation on the spent fuel matrix dissolution has also been investigated [33].

However, recent studies have revealed a threshold value below which the dissolution rate is not affected by the α activity. The threshold value depends on the redox capacity of the environment. Moreover, the radiolysis effect should be inhibited by the presence of hydrogen produced by the corrosion of the container in anoxic conditions [34]. Additive work is needed to study the effect of the near field on the spent fuel dissolution (composition of water in contact, presence of corrosion products and hydrogen).

5. Conclusions

The main results and major questions issued from the PRECCI project are summarized below.

In a closed system, no chemical change is expected in the spent fuel. Thermal diffusion is not relevant at the temperatures and within the time frame of storage and geological disposal. The diffusion enhanced by α self-irradiation appears as limited but this still needs to be confirmed by experiments currently in progress. The consequences of the formation of intra- and inter-granular bubbles of He on the mechanical strength of grain boundaries embrittled by irradiation in reactor have still to be quantified.

As for the cladding, studies are based on a semi-empirical approach combining long-term creep tests (>1 year) coupled with microstructural analyses. They have allowed development of a new creep model for Zircaloy-4 with a domain of validity extended in time.

When exposed to air, the spent fuel rod alteration is governed either by the UO_2 oxidation kinetics, or by the oxygen access through the failed cladding. Oxidation of UO_2 is commonly described via two main steps for which the oxidation kinetics are based on the weight gain curves. Various microstructural observations disagree with the simplified formalism applied in the literature. Thus its extrapolation to other types of fuels such as MOX or to another temperature domain seems inappropriate. Investigating the oxidation mechanisms at smaller scale should diminish the uncertainties affecting the macroscopic approach proposed.

In water, various findings of PRECCI have shown that the UO_2 dissolution rate increases with α and β(γ) radiolysis of water. In deaerated media the dissolution rate increases with the dose rate. β(γ) radiolysis, which enhances the formation of radical species, appears to be more reactive under comparable irradiation conditions (same dose rate and cover gas). However after a few hundred years only α radiolysis of water will persist at the fuel surface. Recent results indicate that the effect of α radiolysis should be limited in time. Studies have now to focus on the effect of the near field conditions on the spent fuel matrix dissolution.

Acknowledgements

The research performed in the PRECCI program is supported by the Commissariat à l'Energie Atomique (CEA), the French Electricity Utility (EDF) and FRAMATOME-ANP. The authors thank the reviewers for their constructive comments.

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