ASSESSMENT OF DRY STORAGE PERFORMANCE OF SPENT LWR FUEL ASSEMBLIES WITH INCREASING BURNUP

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Abstract

Although the safety of a dry long-term spent fuel store is scarcely influenced if a few fuel rods start to leak during extended storage - since all confinement systems are designed to retain gaseous activity safely - it is a very conservative safety goal to avoid the occurrence of systematic rod defects. To assess the extended storage performance of a spent fuel assembly (FA), the experience can be collated into 3 storage modes: I - fast rate of temperature decrease $\delta_{max} \geq \delta \geq 300°C$, II - medium rate of decrease for the fuel rod dry storage temperature $300°C > \delta \geq 200°C$, III - slow to negligible rate of temperature decrease for $\delta < 200°C$. Mode I is typical for early interim storage, mode III covers extremely long-term storage. Mode II dry storage is characterised by the fact that all creep deformation of the spent fuel cladding can already be regarded as terminated as well as the corrosive attack of the cladding. Reviewing the fission product behaviour results shows that the fission products in the UO_2-fuel are practically immobile during storage. Consequently all fission-product-driven defect mechanisms will not take place. The leading defect mechanism - also for fuel rods with increased burnup - remains creep due to the hoop strain resulting from the fuel rod internal fission gas pressure. Limiting the creep to its primary and secondary stages prevents fuel rod degradation. The allowable uniform strain of the cladding is 1 - 2%. Calculations were performed to predict the dry storage performance of fuel assemblies with a burnup ≤ 55 GW·d/tHM based on the fuel assemblies end of life (EOL)-data and on a representative curve T = f(t). The maximum allowable hot spot temperature of a fuel rod in the CASTOR V cask was between 348°C (U FA) and 358°C (MOX FA). The highest hoop strain predicted after 40 years of storage is 0.77% proving that spent LWR fuel dry storage is safe.

1. INTRODUCTION

Nuclear fuel supply and disposal in Germany is characterised by a situation whereby key cost components are assessed in specific terms, i.e. per kg of processed fuel [1]. It thus becomes immediately clear that, apart from the respective specific costs, it is above all the mass of fuel required to generate a given quantity of electric power that has the greatest influence on the total cost. A reduction in the mass of fuel that is in circulation means, in particular, significant savings in storage and disposal costs. The key to any reduction in the mass of fuel in circulation within the nuclear fuel cycle is the fuel assemblies themselves and the in-core fuel performance of which determines how much fuel (enriched uranium or MOX) is needed to generate a given quantity of electric power [2]. Figure 1 uses Siemens fuel assemblies for PWRs as an example to illustrate developments over the last three decades.

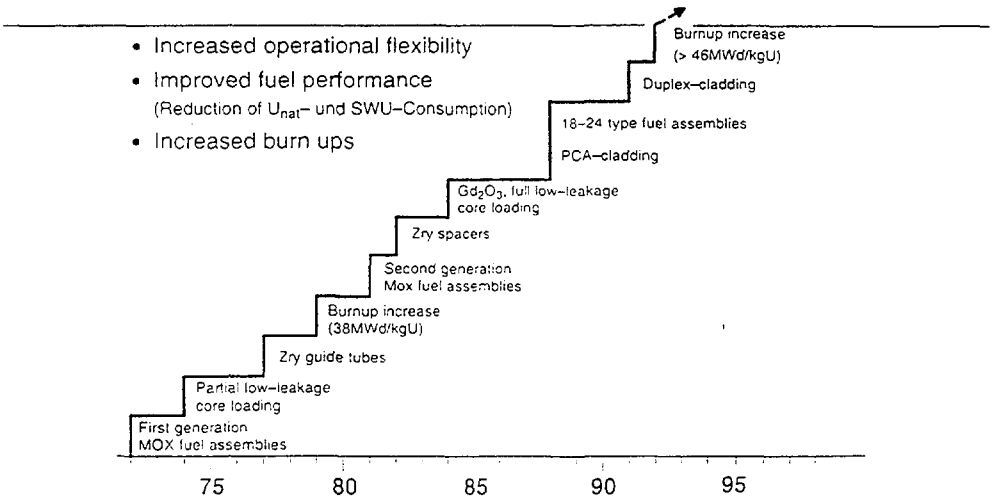

FIG. 1. Answer to increasing spent fuel management costs: PWR fuel assembly technology improvements

Fuel utilisation and operating flexibility optimise uranium and separative work requirements and in-core fuel management. However, the most important part of optimising fuel performance lies in the possibility of achieving higher burnup, which have an inversely proportional effect on the mass of spent fuel to be disposed of per generated $kW \cdot h_e$. Advances made in fuel assembly design and manufacture have now made batch average discharge burnup of over 55 MW·d/kgU achievable.

Figure 2 shows the corresponding development in the average discharge burnup of the Siemens reload fuel assemblies most frequently supplied for BWRs and PWRs. It can be concluded from this figure that, from the point in time at which a sharp rise was experienced in specific disposal costs, higher burnup enabled the mass of fuel requiring disposal to be reduced by:

- approximately 28% in the case of PWRs and even by around;
- 42% in the case of BWRs.

This also explains how it has been possible to achieve continual reductions in fuel cycle costs per $kW \cdot h_e$ since the mid-1980s, in spite of disposal costs that continue to rise. For direct disposal of spent nuclear fuel there is, however, the need to store the spent fuel for an extended period of time. The necessary interim storage might last between 40 and 100 years. In Germany, the dry storage technology had been chosen to store the spent fuel after its removal from wet storage in the pool of the reactor [3]. This paper reviews the dry storage performance of spent LWR fuel with a burnup reaching values up to 55 GW·d/tHM.

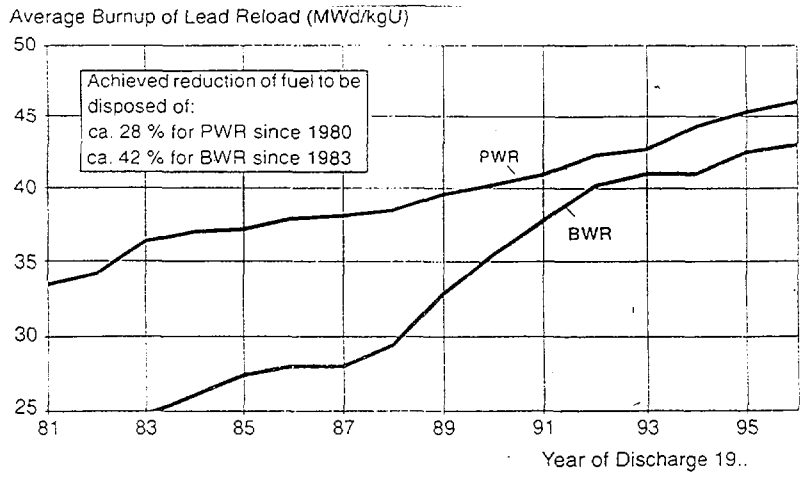

FIG. 2. Burnup increase reduces the circulating fuel quantity and thus reduces the primarily the spent fuel management costs

2. DRY INTERIM STORAGE OF FUEL ASSEMBLIES WITH INCREASED BURNUP

2.1. Definition of storage modes with regard to spent fuel dry storage performance

To assess the performance of spent LWR-fuel in extended storage, the available experience can be collated into 3 storage modes:

- **mode I:** fast rate of decrease in temperature between maximum of licensed dry storage temperature and 300°C;
- **mode II:** medium rate of decrease for in the fuel rod dry storage temperature between 300°C and 200°C;
- **mode III:** slow to negligible rate of decrease in the fuel rod dry storage temperature for temperatures less than 200°C.

Mode I is typical for early interim storage, **mode III** covers extremely long-term storage which is encountered presumably for nearly all dry storage extensions to be considered. **Mode II** dry storage is characterised by the fact that all creep deformations of the spent fuel cladding can already be regarded as terminated as well as the corrosive attack of the cladding. Under the assumption of air ingress to the inert system there - if the oxidative condition holds for a longer period of time - UO_2-fuel-oxidation needs to be considered in more detail. If the UO_2 will be converted in such a case to U_3O_8 the fuel will swell and may lead to fuel rod splitting by the mechanical stress applied to the cladding by the oxidised fuel.

2.2. Source term considerations

Higher fuel assembly burnup leads to higher decay heat power levels and higher n- and γ-source terms [4] (Fig. 3). This correlation is not generally linear in nature, but is frequently characterised by a disproportionate increase with rising burnup levels. These two aspects are in fact in conflict with each other here:

- on the one hand, the cask walls must be as thick as possible and contain sufficient moderator material in order to provide shielding against n- and γ-radiation;
- on the other hand, however, it is precisely this kind of configuration which hampers heat removal, which would be better with thin cask walls.

Cask design thus incorporates a compromise between these two requirements, which ultimately results in restrictions on how spent fuel assemblies can be loaded in the cask.

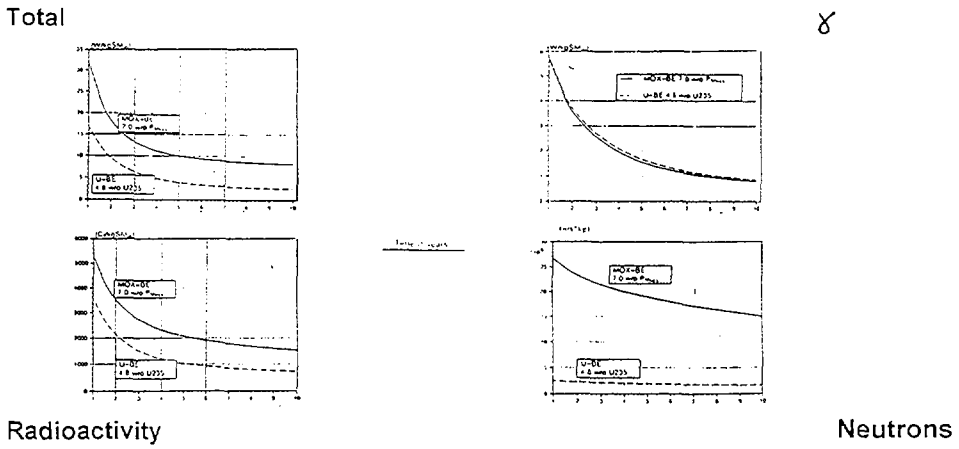

FIG. 3. Thermal and nuclear source terms of spent LWR FA with 65 GW·d/tHM burnup

2.3. Spent fuel rod integrity criteria in dry storage

The integrity of the fuel rod cladding tubes is a crucial factor in interim dry spent fuel storage, and particularly their function as a barrier over the duration of interim storage. Figure 4 compiles all noteworthy fuel rod degradation mechanisms under dry storage conditions:

2.3.1. Oxidative corrosion

Oxidation of the Zircaloy (Zry) is a thermally-induced process. Dry storage under inert gas conditions leads to no further increase in the oxide layer over and above the condition upon final discharge from the reactor, since the storage conditions rule out the presence of oxidising substances. Limited access of air can be assumed if the sealing system of the dry storage system had failed as an

off-normal event. If the spent nuclear fuel stored is free of defects no alteration of the UO_2-fuel will occur since the Zry-cladding can easily withstand the oxidative corrosion at the temperature level to be expected in the storage mode II and mode III period. There is also experimental evidence that oxidation will not defect the cladding for temperatures as high as the primary spent nuclear fuel insertion temperatures licensed when first loading the system after the wet storage period (storage mode I) . In the temperature range of 400°C to 200°C the rate of cladding and fuel oxidation decreases by an order of magnitude if the temperature drops about 10% [5]. If there is a defective fuel rod in the storage system and the temperature is less than 200°C (mode III) still no fission product release is expected, since the UO_2-fuel will practically not oxidise. If the fuel does not oxidise no restructuring takes place and hence no fission product release need be expected. If the temperature exceeds 200°C (mode I and mode II), UO_2 will be converted to U_3O_8 predominantly and Kr will be released [6].

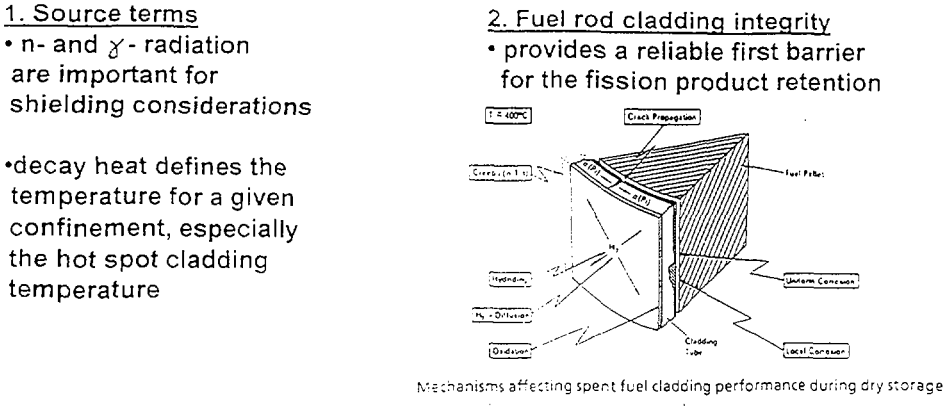

FIG. 4. Source terms of spent FA and spent FA integrity are essential criteria for dry storage

2.3.2. Fission-product-induced cladding corrosion

Fission-product-induced stress corrosion cracking (crack propagation) occurs only within a particular temperature range in the presence of chemically-active iodine and adequate stresses. In dry spent fuel storage, fission products are not present in a chemical form which could trigger any kind of corrosion, and also the stress conditions required for the occurrence of stress corrosion cracking are absent.

Why fission products are stabilised in the UO_2-lattice? UO_2-fuel crystallises in a lattice from CaF_2-type and contains a plurality of vacancies which aid in the retention of fission products generated during the burnup. Additionally it must be considered that each fissioned U-atom leaves a further U-vacancy. Thus even in a fuel with a higher burnup not all vacancies will be occupied by fission products indicating that the UO_2 crystal provides excellent capabilities to retain the fission products.

Why the fission product chemistry stabilises the fission products in the UO_2-lattice? The thermodynamic assessment in the system „U-O-fission products" provides a further argument for the stability of fission products in the UO_2 fuel (Fig. 5). The fission product isotopes from Ce and Zr occur in the tetravalent state and can therefore easily replace a fissioned tetravalent U. Y and the lanthanides may occur in a trivalent state and need - if positioned on a U side - another neighbouring atom in the monovalent state. The Pt metals are stable in the non-ionised metallic state. Since the O potential in a fuel rod is high enough to oxidise Zr and to keep the UO_2-fuel in an almost stoichiometric, the Cs released from the fuel may occur in the form of a Zirconate. Mo as a fission

product has a nearly identical free energy function as UO_2-fuel. Establishing a valancy balance for the fissioned UO_2-atoms and their related fission products without Mo results in a slight surplus of negative valences. The Mo will therefore balance the system by occurring partly in the metallic state and partly in oxidised form.

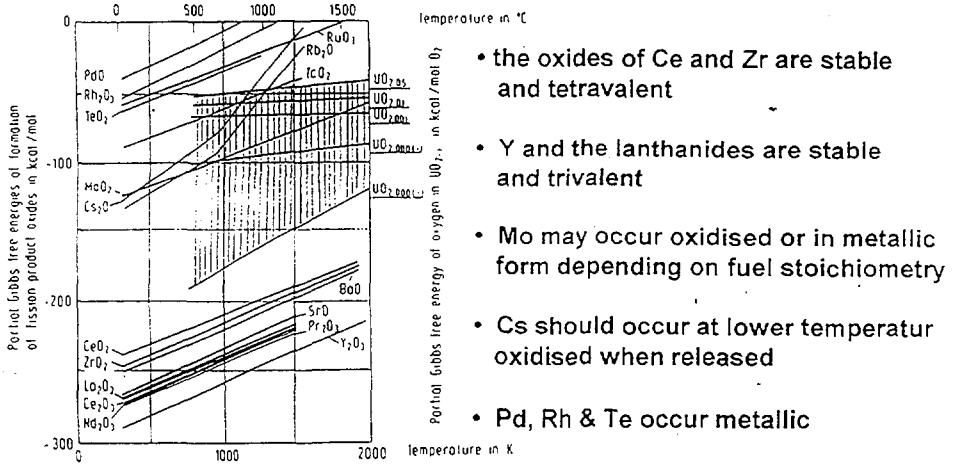

FIG. 5. Gibbs free energies of formation of FP oxides and partial Gibbs free energy of the O in UO_2 of various stoichiometries

Why the fission product atomic radii stabilise the fission products in the UO_2-lattice? Another interesting aspect for the stability of the fission products in the UO_2-crystal results from the comparison of the atomic and ionic radii of the tetravalent U with those of the fission products in their characteristic chemical state. The ionic radii of La, the lanthanides and of the tetravalent Ce are practically identical to those of the tetravalent U. Most of the other fission products including the noble metals lie a +/- 30% scatter around U (Fig. 6). The incorporation of such atoms into vacant sites of the UO_2-crystal is energetically favourable. The atomic radii of the noble gases as well as those of the Rb-, Cs and I-atoms however are exceeding largely those of the tetravalent U ions. The situation is even worse for the Cs and Rb in ionic form. Therefore it must be concluded that the noble gases, the earth-alkali-metals and iodine will tend to be released from the UO_2-crystal if the relevant kinetic processes will allow.

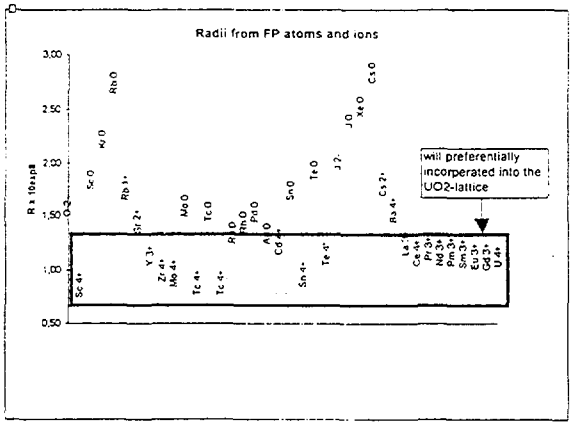

FIG. 6. Virtual atomic and ionic radii of the FP in the UO_2 lattice

Why the are the fission products in the UO_2-lattice so stable? The release of meta-stable implanted fission products occurs by more or less complicated diffusion processes. Since the diffusion under spent nuclear fuel dry storage conditions is a thermally activated process the temperature distribution in the fuel will be of great importance. A large data base and extensive knowledge is available to describe the release of the fission gases from the fuel under in-service conditions during reactor operation. Typically, the fuel temperature during reactor operation on its surface is around 400°C and in its centreline between 1,300°C and 1,800°C depending on the heat rating and burnup. For temperatures above 1,000°C, the diffusion coefficients are only dependent on the temperature. Those coefficients decrease between 1,800°C and 1,000°C from 10^{-12} cm^2/s to 10^{-16} cm^2/s by 4 orders of magnitude. Between 1,000°C and 400°C the pure thermal diffusion is combined in-pile with a radiation supported term only decreasing by 2 orders of magnitude. Since in storage the radiation level is negligible in comparison to in-pile conditions, only the pure thermal diffusion will contribute to fission gas release. Therefore, the diffusion coefficient at storage temperature is at least 8 orders of magnitude less than compared with those at fuel centreline temperature, or 5 orders of magnitude less than that of the fuel average temperature. Therefore, it can be concluded that under dry storage conditions for fuel temperatures less than 400°C, no fission gas will be released at all even under extended storage periods. Investigation of the Cs and I release from irradiated fuel has shown [7], that the release is diffusion controlled and the diffusion coefficients of those fission product isotopes are very similar to those of the fission gases. It was found that below a specific temperature there was no release at all. This temperature decreases with increasing burnup and will be above 800°C and 900°C (Fig. 7). This corresponds to the findings for the fission gas release as discussed above. Therefore, it can be concluded that neither Cs nor I will be released under dry storage conditions from the fuel to the gap of a spent LWR fuel rod.

In summary it can be stated that the fission products generated in the UO_2 fuel under in-service conditions are practically immobile in the UO_2 fuel lattice during storage. Consequently all fission-product-driven defect mechanisms such as:

- stress corrosion cracking (SCC);
- uniform fuel rod internal fission product corrosion of the cladding;
- localised fuel rod internal fission product corrosion of the cladding will not take place.

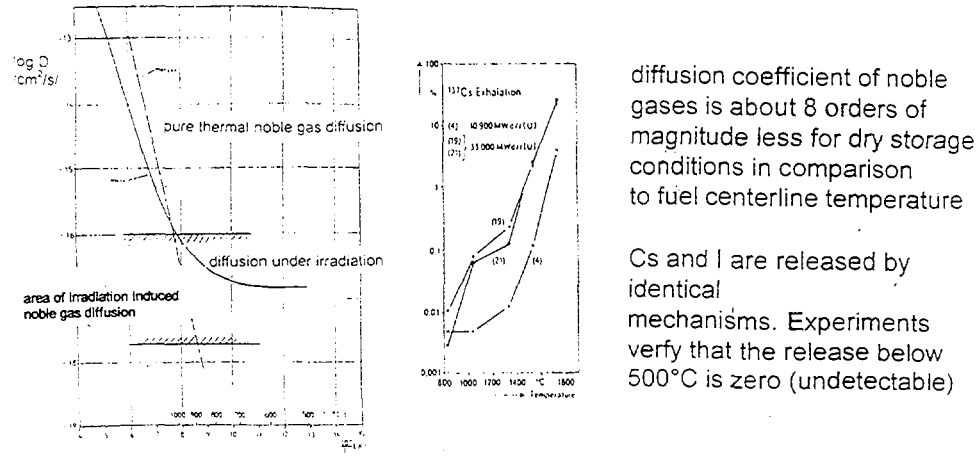

FIG. 7. Thermal release of noble gases, Cs and I

2.3.3. Delayed hydrogen cracking

If the hydrogen content of a Zry-cladding exceeds the solubility - which is 160 ppm at 400°C - the hydrogen is precipitated in form of Zr-hydride platelets. Fuel rods with a burnup exceeding 50 GW·d/tHM may have a greater hydrogen content. The hydrogen precipitates reduce the ductility of the cladding if orientated orthogonal to the applied stress.

Crack initiation: Incipient cracks at the inner cladding surface may initiate delayed hydrogen cracking if hydrogen hydrogen precipitates at the crack tip decreasing the critical stress intensity factor. Radially-orientated hydrogen precipitates may serve as a incipient crack only if the temperature of the cladding is less than 160°C which represents the ductile-brittle transition for the precipitated Zr-hydride platelets.

Hydride re-orientation: The cladding tube manufacturing process generates a texture in the cladding causing the hydride-platelets to precipitate mostly tangentially. Re-orientation of hydride platelets may occur if the texture of the cladding changes under specific stress conditions and hydrogen is precipitated afterwards. At the grain sizes typical for LWR cladding those stress conditions are [8]: δ = 400°C ; σ = 120 -180 N/mm^2 and δ = 250°C ; σ = 250 - 350 N/mm^2 . Those stresses in the cladding are precluded under dry storage conditions in Germany by limitations in the storage license.

Critical crack size: Based on Canadian results [9,10], the critical crack size can be calculated by the following equation:

$A = C \times (K_{IH} / \sigma)^2$ ...../ m/

with C=0,4

resulting in the data in Table I.

TABLE I. CRITICAL CRACK DEPTH FOR DELAYED HYDROGEN CRACKING

T /°C/	K_{IH}/ MNm$^{-3/2}$/	σ/ MN/m^2/	Critical crack depth
150	10 (unirradiated)	120	695 μm
150	5 (irradiated)	120	348 μm

The stress intensity factor for the unirradiated material results from experimental investigations. The value for the irradiated material status is assumed to be only half of that from unirradiated material. As a result the critical crack depth is 348 μm (Fig. 8). Neither incipient cracks from such depth nor re-oriented hydride-platelets had been observed in spent fuel in the EOL condition or after dry storage. In Germany the maximum hoop stress in the cladding during storage is limited by the licenses to 120 MN/m^2, the likelihood that hydride re-orientation might occur during storage can therefore be excluded. Altogether it can be concluded, that delayed hydrogen cracking will not occur in dry storage, even if the hydrogen contents rises in the cladding with the burnup. This conclusion is in agreement with the experience that this defect mechanism has never been observed world-wide.

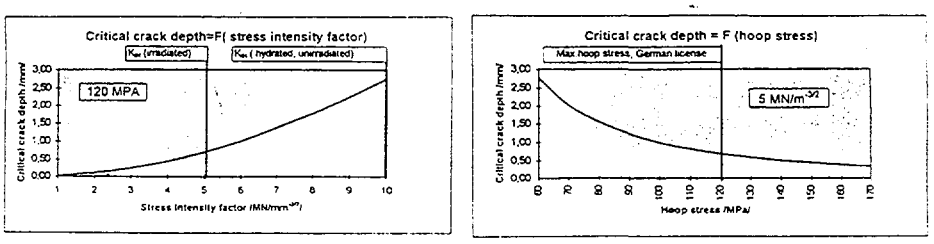

FIG. 8. Critical crack depth in relation to stress intensity factor and hoop stress

2.3.4. Creep

Creep is the enveloping criterion for consideration of cladding integrity during storage. At the temperatures of between 300 and 400°C (storage mode I), which prevail at the start of dry storage, the cladding undergoes strain. Its numerical value is largely determined by the fuel rod internal pressure and the temperature-time history in the course of dry storage. At maximum temperatures of less than 400°C, a total cladding strain of approximately 2 to 3% has no negative effect on cladding integrity, and is therefore used as a basis for current licenses [11,12].With higher burnup the dry storage temperature increases due to the higher decay heat from fuel assemblies. The also increased fission gas release results in a higher internal gas pressure within a spent fuel rod. This will generate higher stresses and strain in the fuel rod cladding. Since the longer residence time of a fuel assembly in the core to achieve the higher burnup tends to reduce the residual wall thickness through in-reactor corrosion, stress and strain is furthermore increased. MOX fuel exhibits, already at the same burnup as U fuel, increased decay heat and fuel rod internal EOL-pressure. MOX fuel with increased burnup will therefore have the highest stress and strain while in dry storage.

3. PREDICTION OF SPENT LWR FUEL ASSEMBLY DRY STORAGE PERFORMANCE

3.1. Method and procedure

The results from Section 2.3.4 show that cladding creep from inner gas over pressure is the rate determining degradation and the failure mechanism for setting maximum allowable storage temperature limits in dry inert storage. If creep were allowed to proceed to rupture, the fracture mode would most likely be of a pinhole type. Nevertheless, it is desirable to avoid any degradation. This can be accomplished by confining the creep degradation mechanism to its primary and early secondary stages. If it can be shown that the creep strain never exceeds that critical strain domain during inert dry storage, the tertiary creep with its subsequent fuel rod defection can be excluded. This approach is in compliance with standard creep engineering practices. This methodology relies on the availability of:

- a date base of cladding creep test results under internal pressurisation conditions [13]. The data base was published for the traditional type of Zry cladding at the spent fuel storage symposium in Seattle in the year 1985 [14] and on the occasion of the SMIRT-conference in Brussels in the same year [15]. The data on cladding especially developed for higher burnup fuel was published recently[19] (Fig. 9);
- a correlation which allows the prediction of post-pile creep from creep of unirradiated material. As could be shown by comparison of the creep of unirradiated and irradiated Zry the creep for unirradiated material always describes the post-pile creep conservatively (Fig. 10);
- a date base of the potential total strain of fuel rod cladding under dry storage conditions. For a burnup less than 40 GW·d/tHM the data from the post-pile burst test indicate that all cladding will reach, at minimum, 1% uniform strain before tertiary creep starts (Fig. 11). Since the straining capability decreases with burnup the very conservative measurement of the allowable strain by the burst test with its very high strain rates was replaced by a creep burst test with much lower strain rates to verify that Zry will have more than 1% strain under dry storage conditions. Siemens performed such a test programme under contract to GNB providing the spent fuel from a Siemens high performance programme having a burnup up to 64 GW·d/tHM. W. Sowa et al. reported from that programme that its result provided the basis for the recent German licence to store spent LWR fuel with increased burnup allowing 1% strain for U FA and MOX FA with an average batch burnup as high as 55 GW·d/tHM [16];
- a numerical model to predict creep strain during dry storage. This model was first published in Seattle [17,18] and was revised for cladding developed specially for increasing burnup in 1997 [19].

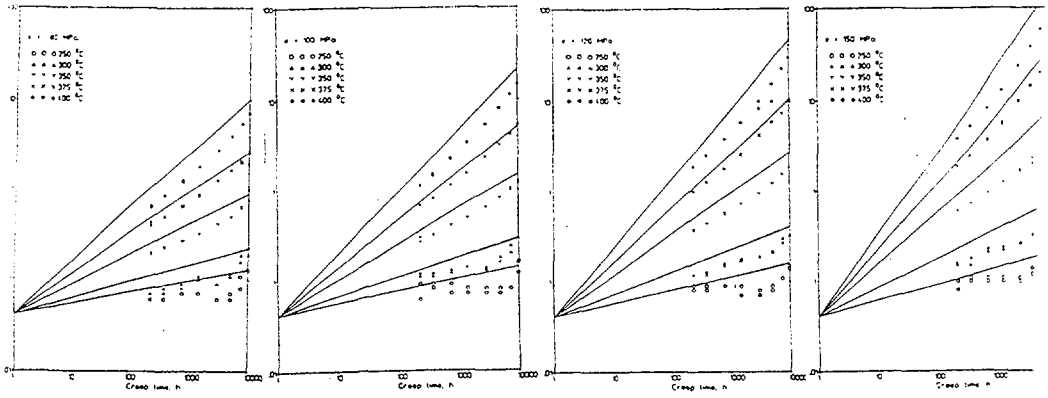

FIG. 9. Experimental creep data and calculated values using the creep equation derived from the experimental data base

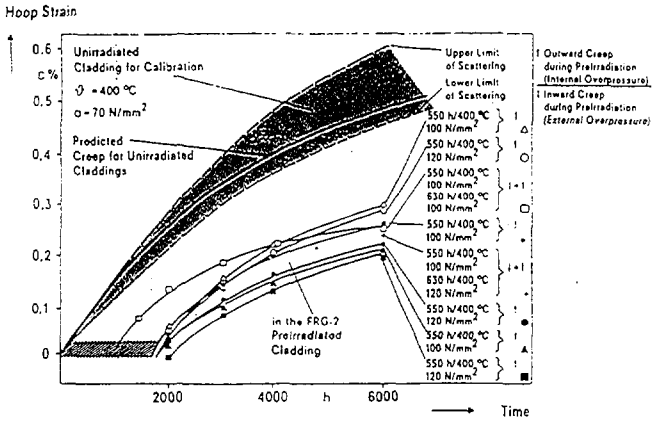

FIG. 10. Comparison of creep strain of unirradiated and irradiated Zry-4 cladding

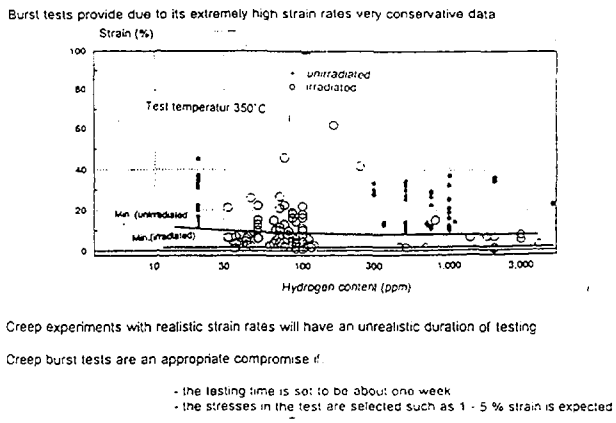

FIG. 11. Burst strain data of Zry cladding in relation to its H content

3.2. Dry storage predictions for spent fuel with increased burnup

As an example, a CASTOR V/19 is considered loaded with:

- 19 UO_2 LWR FAs having a maximum rod burnup of 55 GW·d/tHM; or
- 15 UO_2 LWR FAs **and** 4 MOX LWR FAs both having a maximum rod burnup of 55 GW·d/tHM.

Additionally, 2 types of fuel rod cladding is looked at to assess the influence of different cladding material. Table II shows the scope of the assessment.

Table II. SCOPE OF STRAIN CALCULATION FOR CASKS LOADED WITH SPENT LWR FA

Case	1	2	3	4	units
Cladding	fast creeping clad		less fast creeping clad		
Burnup (rod)	55	55	55	55	GW·d/tHM
Fuel	UO_2	UO_2/PuO_2	UO_2	UO_2/PuO_2	
Pool storage	5.6	6.5	5.6	6.5	years
Decay heat/cask	32.3	37.5	32.3	37.5	kW at start of storage
FR hot spot temp.	348	357	348	357	°C
Storage period	40	40	40	40	years

Siemens-PCA-2 is an example of a relatively fast creeping cladding, whereas the Siemens-DUPLEX cladding represents a relatively strong cladding. In calculating the end-of-life conditions for reactor operation, the different in-service behaviour of both cladding types has been considered. Fast creeping cladding contacts, due to the creep down under the external coolant, pressure the oxide fuel earlier than strong cladding. As a result of the earlier closing fuel cladding gap, the fuel temperature and consequently the fission gas release is less than for the stronger cladding. Therefore, at the start of dry storage the internal fuel rod pressure in the cases 1 and 2 is less than for cases 3 and 4. The starting conditions and the hoop strain after 40 years for dry storage of the 4 considered cases are compiled in the Tables III and IV respectively. The results of the calculation for case 1 through 4 are given in Figs. 12 through 13.

TABLE III. BEGIN OF DRY STORAGE CONDITIONS FOR CASE 1 THROUGH 4

type of cladding	lower temperature	higher temperature
fast creeping cladding	348°C $\sigma_{hoop} = 60$ N/mm^2 case 1	357°C $\sigma_{hoop} = 60$ N/mm^2 case 2
less fast (stronger) creeping cladding	348°C $\sigma_{hoop} = 80$ N/mm^2 case 3	357°C $\sigma_{hoop} = 80$ N/mm^2 case 4

TABLE IV. HOOP STRAIN AFTER 40 YEARS DRY STORAGE FOR CASE 1 THROUGH 4

Type of cladding	lower temperature	higher temperature
fast creeping cladding	0.37%	0.51%
stronger cladding	0.52%	0.77%

Since it is generally accepted that spent Zry-type cladding can withstand, under dry storage conditions, at least 1% hoop strain, the results presented indicate that under the considered conditions spent LWR fuel up to an average fuel rod burnup of 55 GW·d/tHM can be stored free of systematic

defects for periods up to 40 years. Since follow-on storage up to 100 years will be mostly within the mode 3 an extension of the storage period will not result in defects in such spent LWR fuel.

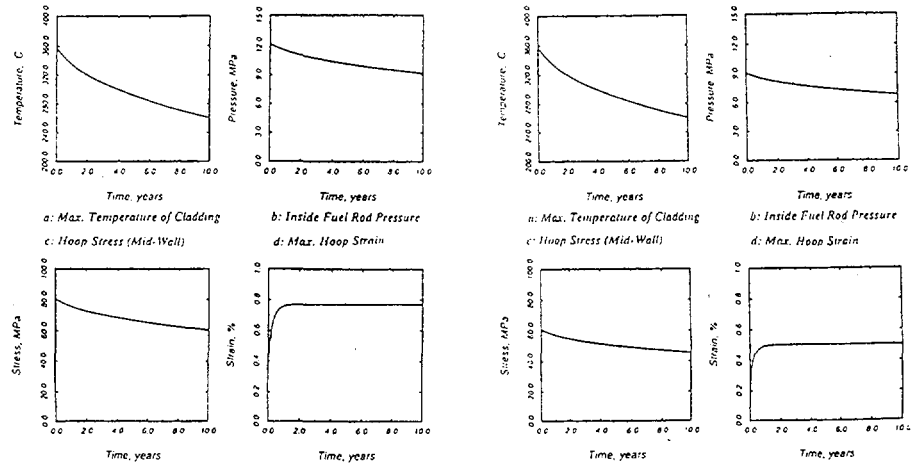

FIG. 12. Spent PWR MOX FA, predicted strain in dry storage, 55 GW·d/tHM (left strong cladding, right fast creeping cladding)

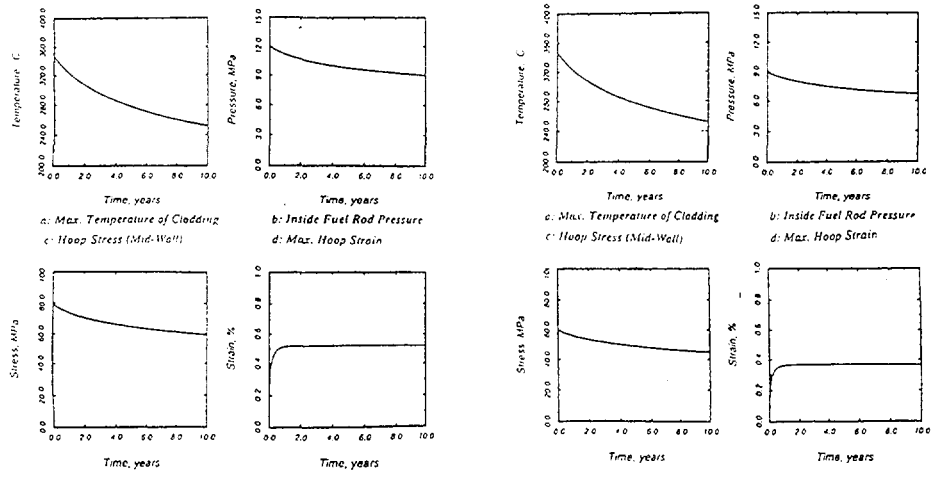

FIG. 13. Spent PWR UO_2 FA, predicted strain in dry storage, 55 GW·d/tHM (left strong cladding, right fast creeping cladding)

4. CONCLUSIONS

The leading defect mechanism for spent fuel rods in dry storage - also for fuel rods with increased burnup - remains creep due to the hoop strain resulting from the fuel rod internal fission gas pressure. Limiting the creep to its primary and secondary stages prevents fuel rod degradation. Post-pile creep of fuel rod cladding can be described conservatively by the creep of unirradiated cladding. The allowable uniform strain of the cladding in its typical post-pile condition preventing tertiary creep under dry spent fuel storage conditions is 1 -2%. Calculation of the predicted dry storage performance of fuel assemblies with a burnup $\leq$55 GW·d/tHM was based on the fuel assemblies' end-of-life data and on a representative curve T = f(t). The maximum hot spot temperature assumed for a fuel rod in the CASTOR V cask was between 348°C (U FA) and 358°C (MOX FA) at the beginning of storage. The highest hoop strain predicted after 40 years of storage is 0.77% and 0.52% respectively. This result proves that dry storage is safe for LWR fuel of such burnup.

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