POTENTIAL STRESS ON CLADDING IMPOSED BY THE MATRIX SWELLING FROM ALPHA DECAY IN HIGH BURNUP SPENT NUCLEAR FUEL

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Studies using literature data were conducted on potential swelling of the spent nuclear fuel (SNF) matrix due to alpha decays. Swelling of the matrix may impose stress on cladding during long-term storage of SNF. The literature data were obtained for UO_2 having undergone accelerated irradiation conditions. The representation of the simulated accelerated tests for the SNF is discussed. The stress on cladding, and its potential implications in terms of long-term degradation, would depend on SNF matrix swelling, gap size, crack pattern and crack inter-locking, and friction of the SNF matrix and cladding. Assessments and reviews are made for the observed SNF matrix swelling, stress generated on cladding, and gap size. Applying this information, the stress on cladding due to alpha decays is semi-quantitatively assessed using stress analysis induced by thermal expansion.

I. INTRODUCTION

During long-term storage of spent nuclear fuel (SNF), the UO_2 matrix may undergo swelling due to alpha decays. The alpha decays introduce microstructure defects and He in the matrix.[1,2] This swelling may impose stress on cladding, especially if the gap between the cladding and the matrix is closed, as occurring for high burnup SNF (e.g., ~ 60 GWd/MTU). If this stress is sufficiently high it may impact cladding integrity. This paper assesses the potential for matrix swelling and gap closure in SNF, and assesses potential stresses imposed on the cladding. Various short-term test methods and data of accelerated irradiation are discussed, in simulating long-term swelling without acceleration. The use of simulated unirradiated UO_2 representing SNF is also discussed. From literature reviews, the stress analysis accounting for thermal expansion is presented to estimate the potential stress on the cladding from swelling due to alpha decays. The gap size is measured following irradiation after cooldown in the SNF pool and heat-up in dry storage to estimate the time when the maximum stress would be applied to the cladding.

II. EXPERIMENTAL TEST METHODS AND DATA ON UO_2 MATRIX SWELLING BY ACCELERATED IRRADIATION

Rondinella et al.[1] measured the lattice parameter changes by alpha decays in UO_2 doped with short-lived alpha emitter. The primary alpha-dopants are short-lived actinides such as Pu-238 or U-233. Alpha decays in SNF at lower density in a longer time. To simulate in a shorter time the effect of the alpha decays in SNF, higher density of alpha emitter is doped in UO_2. In Fig. 1 the changes in lattice parameter over time are due to the alpha-decay events in terms of both radiation damage and He accumulation. The displacement per atom (dpa) was used as an indicator of the cumulative concentration of defects and He. The rate of the increase of the lattice parameter decreases with time and, eventually, the lattice parameter becomes saturated at a maximum level of 0.4-0.5 %.[3] Complementary measurements support Fig. 1: (i) similar trends to the lattice parameter are observed by monitoring hardness[4] and thermal diffusivity;[5] and (ii) alpha-doped samples with 100 times lower specific alpha-activity shows similar behavior. Thermal annealing of the lattice swelling and of other properties was investigated.[6] The first annealing stage starts at ~500 ºK (227 ºC, 441 ºF) and peaks around 650 ºK (377 ºC, 711 ºF).[6,7] The temperature regimes expected under current storage conditions fall below the annealing range during most of the storage time. Possible annealing effects associated with peak temperature and related storage times experienced by SNF shall be investigated in dedicated experimental campaigns.

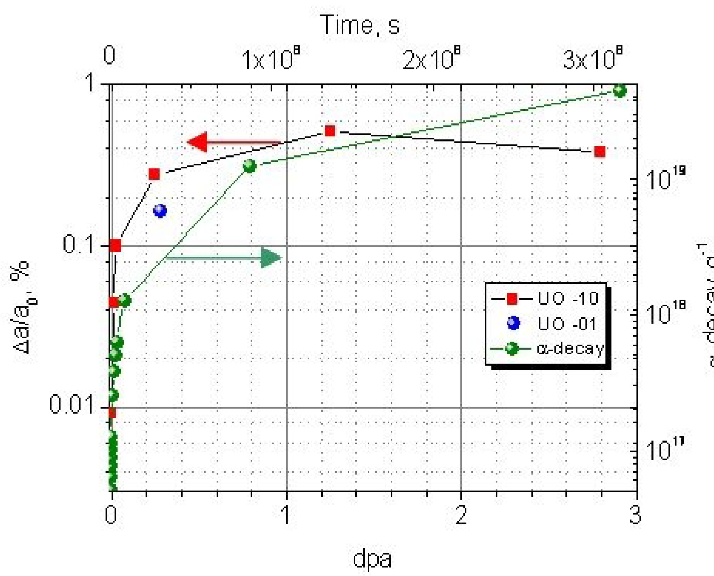

Fig. 1. Lattice parameter "a_o" and its fractional change,."$\Delta a/a_0$" as a function of time and accumulated damage (displacement per atom, dpa); UO – 10 is UO_2 with 10 wt % of Pu-238; UO – 0.1 is UO_2 with 0.1 wt % of Pu-238; testing time is 6.3 years. Also shown is the cumulative alpha-decay curve for Pu-238 (right axis).[1]

Fig. 2 shows a summary of literature data on the swelling of UO_2 and some other actinide oxides, including the present data on UO_2 doped with alpha emitter and alpha-implanted UO_2.[8] The swelling curves are in contrast with the lattice parameter contraction expected for hyper stoichiometric UO_{2+x} (dashed diagonal line), indicating that the swelling is not caused by oxidation during storage. The doped cases with alpha emitter show up to 0.2 - ~0.5% swelling in the range 0.1 – 1.0 dpa corresponding to tens or hundred years of SNF storage, (see Fig. 3). The alpha particle implantation cases show more pronounced swelling, up to ~ 0.8 % at ~ 0.07 dpa.

The lattice swelling measured for doped UO_2 with alpha emitter is caused by the defects induced in the lattice by the decay damage, and also by the accumulation of He. The latter, in particular, may be responsible for the maximum (or "saturation") fractional swelling, observed at an accumulated dose of ~1.2 dpa.[3]

Long-lived actinides such as Pu series, Am series, Cm series and Np are present in SNF and determine the alpha-dose during long-term storage and disposal. Fig. 3 shows the storage and disposal time required for SNF to reach the dpa level corresponding to the saturated lattice parameter in Figs. 1 and 2, and the accompanying generation of He.

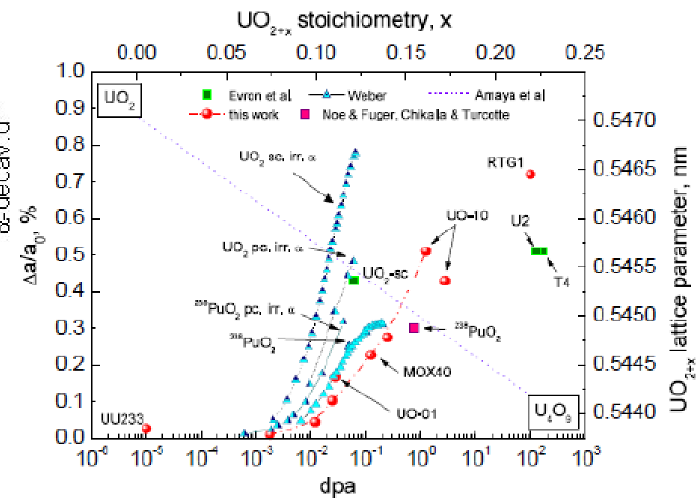

Fig. 2. Summary of lattice parameter swelling as a function of accumulated alpha particle damage from different authors[1] [pc=polycrystal; sc=single crystal]. The alpha particle damage was obtained by doping with alpha emitter,[6] alpha-particle implantation,[8] recoil atom bombardment[8] of UO_2, or by direct measurement of lattice parameter in actinide oxide samples (PuO_2, $^{238}PuO_2$ - P^3B; see also Eyal's review[9]), or natural analogues.[10] UU233: UO_2 doped with 10 w% U-233, P3B: $^{238}PuO_2$ for radio-isotopic thermal generator, U2: urinate, T4: thorianite, MOX40: sol-gel MOX with 40% Pu. Amaya, et al.[11] Noe and Fuger[12] Chikalla and Turcottee[13]

For 40 – 80 GWd/MTU , doses are 0.2 – 0.4 dpa in 100 years as shown in Fig. 3, which would correspond to 0.2 – 0.5 % lattice expansion for doped UO_2 with alpha emitter in Figs. 1 and 2. This expansion value is likely to result in an appreciable stress condition, as described in the next section.

The doped UO_2 with alpha emitter used in the cited experiments provides a homogeneous and isotropic distribution of alpha-decay events in the UO_2 matrix, due to the intimate mixing of the alpha-emitter in the UO_2 obtained by sol-gel routes sample fabrication methods. However, there are uncertainties on the applicability of results obtained by using doped unirradiated UO_2 with alpha emitter (and even more so for alpha particle implantation) to represent SNF. The complex micro-structure of SNF includes fission products, different phases, fission gas bubbles, and cracks, as a result of the

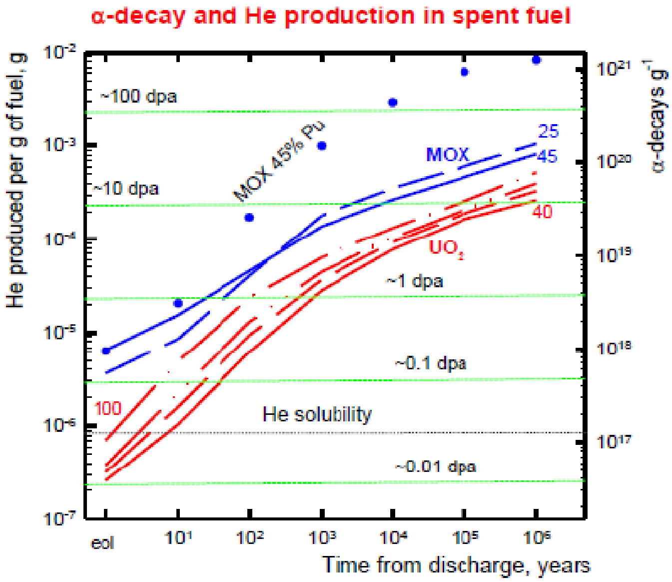

Fig. 3. Alpha particle damage accumulated during SNF storage.[3] Burnup range is 40-80 GWd/MTU for UO_2 and 25-60 GWd/MTU for MOX.

in-pile operation of the fuel. These features are not present in the doped UO_2 with alpha emitter. A comparison of alpha-decay damage and He accumulation effects in doped UO_2 with alpha emitter and SNF has been performed with respect to thermal diffusivity, microstructure alteration and He release.[1,2,5,6] It showed doped UO_2 with alpha emitter represented SNF well. However, no bench marking with SNF has been done yet concerning the matrix swelling. This is the object of ongoing studies aiming at comprehensively assessing the transferability of data obtained from model materials to the case of actual SNF.

III. STRESS ANALYSIS ON CLADDING BY THERMAL EXPANSION OF UO_2 MATRIX AS AN INDICATOR OF IRRADIATION-INDUCED UO_2 MATRIX SWELLING

The stress that is generated from solid matrix swelling[14,15] has a global simple form of:

$$\sigma = E/(1 - 2\,\nu)\,[\Delta V/(3\,V)] \qquad (1)$$

σ: stress developed
E: Young's modulus
ν: Poisson's ratio
V: volume
Δ: volume change

Retel et al.[16] presented a more rigorous formula and a numerical simulation of the stress imposed on cladding surface by the thermal expansion of the UO_2 matrix under reactor ramping conditions. This analysis of the thermal expansion is appropriate to assess the stress imposed by the irradiation-induced swelling. This is because the models used by Retel. et al.[16] to calculate the stress generated are primarily dependent on the magnitude of swelling. The schematic of fuel fragments and cladding used in the model is given in Fig. 4.

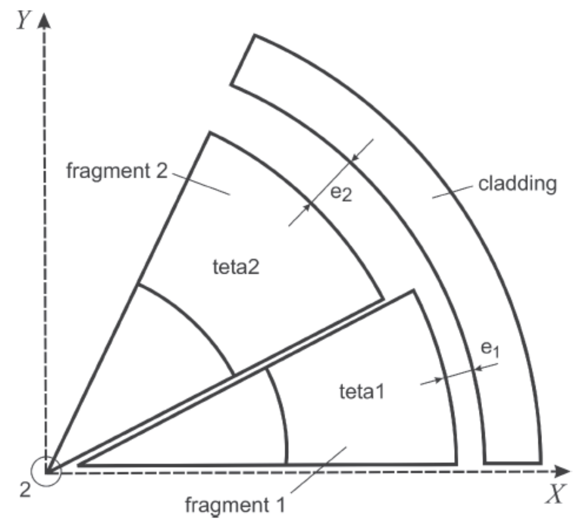

Fig. 4. Model used in the calculation of thermal expansion of SNF by Retel, et al.[16]

Table I shows the hoop stress on cladding and the UO_2 matrix displacement from thermal expansion deduced from Retel, et al.[16] Initially, this displacement closes the gap or removes compressive stress (minus stress) on cladding from back water pressure. It imposes further tensile stress on cladding. The different displacements in Table I reflect the hourglass phenomena with different thermal behavior along the longitudinal direction. At a displacement of around 14.0 μm (5.5 μ inch), hoop stress disappears with 0.17 to 0.30% displacement. Therefore, the thermal expansion could be from a value between 0.17 and 0.30 to 0.68% for this specific reference configuration. The hoop stress, 434 MPa (62.9 psi) is from [0.68 minus (0.17 to 0.30) %]; 318 MPa (46.1 ksi) is from [0.50 minus (0.17 to 0.30) %] (e.g., Ahn and Rondinella[17]).

TABLE I. Cladding hoop stress and the UO_2 matrix displacement from thermal expansion deduced from Retel, et al.[16]

Maximum Radial Displacement of teta1 and teta2, μm (μ inch)	Percentage of Displacement with respect to Rod Radius, 0.4096 cm (0.1613 inch)	Cladding Hoop Stress, MPa (ksi)
27.9 (11.0), 0 (0)	0.68	434 (62.9)
21.0 (8.3), 7 (2.8)	0.50	318 (46.1)
14.0 (5.5), 14.0 (5.5)	0.30	-34.8 (-5.1)
7.0 (2.8), 21.0 (8.3)	0.17	82.7 (12.0)

The gap depicted in Fig. 4 represents combined crevice sizes of all cracks. In reality cladding can be bonded with the matrix. Under thermal shrinking, this bonding allows some compressive stress on cladding, if no additional cracking occurs in the matrix. At high burnup SNF, this compressive stress will not exist with tensile stress as a result of permanent swelling of the matrix. For example, in reactor there will be about 1 % linear swelling by irradiation in reactor at burnup of ~ 60 GWd/MTU.[18] Therefore, no gap will exist after cooling without residual compressive stress on the cladding. This implies that any swelling during storage will impose the stress on cladding directly without the need for prior gap filling.

Considering these two examples of existing bonding or gap, Table I shows a maximum thermal expansion, which will produce gap upon cooling after reactor discharge. Some minor filling of the gap if any will also occur with a rise of temperature during storage. Other factors associated with gap and cladding stress before storage include: (i) ridge formation of the matrix imposing localized stress on cladding: (ii) crack interlocking, and (iii) friction coefficients for (cladding and fragment) and (fragment and fragment).[16] The friction effect may be more pronounced when there is bonding of cladding with the matrix and the UO_2 matrix cracking forms ridges to impose concentrated stress on cladding.[19,20] Fig. 5 shows examples of crack pattern, showing bonding, and various crack directions and interlocking. Detailed analyses of gap size from thermal shrinking will be examined in future work.

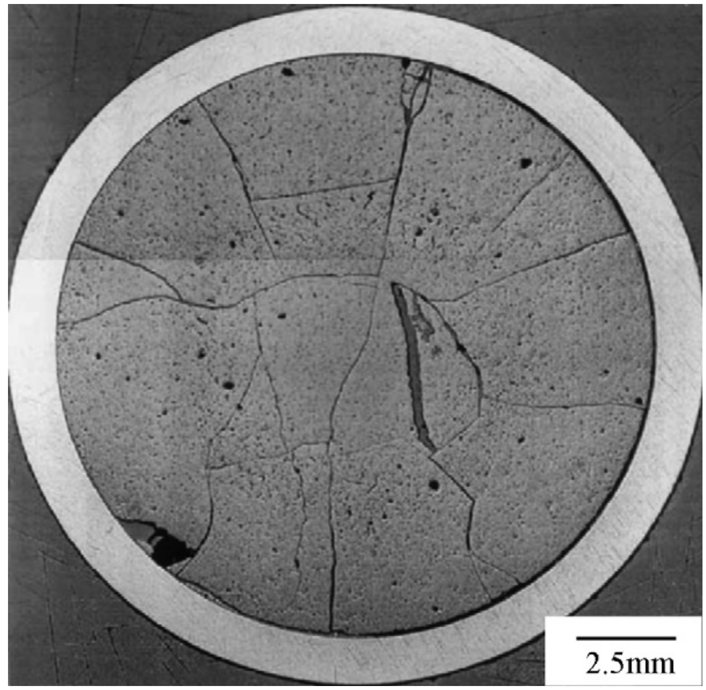

Fig. 5. Examples of crack pattern by Sasahara and Matsumura.[21]

IV. INFORMATION AND ASSESSMENT OF GAP SIZE AND STRESS ON CLADDING

As discussed above, the magnitude of stress imposed on cladding by the matrix swelling depends on the initial gap and the matrix swelling during storage. Currently, available information on the gap size from Post Irradiation Examination is limited and qualitative. The gap closure is routinely observed in the higher axial burnup regions of LWR SNF rods starting from above 40-45 GWd/MTU average rod burnup. Medium-high burnup rods in commercial reactors are characterized by axial burnup gradients which allow within a single rod, for the presence within a single rod of regions where the fuel-cladding gap is fully closed and regions where a sizable gap is still present. In reactor operation the gap will be closed after a relatively short period of operation. Normal reactor operation induces thermal expansion and irradiation-induced swelling simultaneously. Whereas the thermal expansion reaches a steady-state value, essentially set by the temperature regimes in-pile, the radiation-induced swelling, associated with the fission damage and the production of fission gases, will continue. However, the total swelling should not exceed the required creep limit of cladding. The pellet cladding mechanical interaction at medium-high burnup, after closure of the gap, is positively affected by the formation of the high burnup structure at the outer rim of the fuel pellet.[2] The high burnup structure is characterized by reduced hardness compared to the unrestructured SNF matrix.[22]

As discussed above, the thermal stress will be relieved in various ways after reactor discharge., High burnup SNF may develop more cracks in the matrix upon cool down due to the bonding of cladding with the matrix, in addition to the matrix cracks developed during reactor operation. As mentioned earlier, however, the gap by thermal shrinking may not occur in some cases when the bonding does not create further matrix cracking. The FRAPCON[18] exercise is currently under way to estimate the gap size after discharge. However, FRAPCON does not consider bonding or other geometric complications. Also, the temperature of SNF storage will reach up to 400 °C (752 °F) initially, which reduces the thermal shrinking of the matrix. SNF rods were examined after 20 year of storage in Japan.[21] Fig. 6 shows that for PWR SNF at a burnup of 58 GWd/MTU, the gap was fully closed (less than 2 μm) and bonding was observed. It is not known if irradiation-induced swelling processes during storage affected the bonding. The gap decreased after storage for MOX SNF.

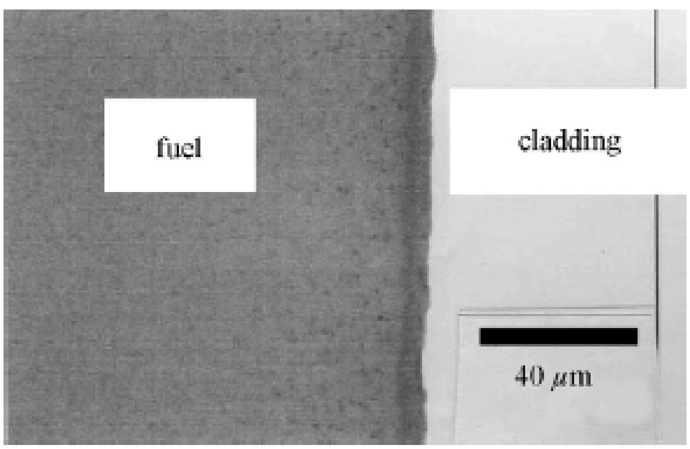

Fig. 5. Inner oxide layer of PWR SNF cladding with burnup of 58 GWd/MTU after dry storage for 20 Years in Japan.[21]

The data from 20 year storage and the analysis of the above mentioned results of thermal expansion and alpha-decay swelling indicate that the gap size appears to fall in the range where the subsequent net swelling by alpha decays will likely cause stresses on the magnitude of those predicted by the thermal model described above in Table I. The likelihood of generating cladding stresses to be generated in cladding during storage may become higher when irradiation-induced swelling on the order of magnitude shown in Fig. 2 is expected or when the gap is closed and bonded to the SNF-cladding. Finally as mentioned previously, the stress is very sensitive to other factors such as the aforementioned ridge formation, crack inter-locking, and friction coefficients. The stress on cladding may vary significantly, depending on the magnitude of these factors.

Since the gap/bonding region and crack pattern could significantly affect the stress on cladding, the estimated stresses presented in Fig. 4 serve only as a case example. More case studies need to be conducted with additional information on gap and crack pattern for a better understanding of the stress exerted on cladding by the SNF matrix during dry storage. Currently, there are no easy methods to experimentally bench mark these stress assessments experimentally.

V. SUMMARY

Potential swelling of the SNF matrix due to alpha decays is reviewed and assessed from literature data. Swelling may impose stresses on cladding during long-term storage of SNF. The following summarizes the review:

(i) The literature test data were obtained using doped UO_2 with alpha emitter, alpha-particle implantation or recoil atom bombardment on simulated unirradiated UO_2. The representativeness of the simulated accelerated tests for the SNF is addressed.

(ii) The potential for stresses applied on cladding depends on the swelling of the SNF matrix, the gap between cladding and the SNF matrix, and the crack pattern in SNF matrix. Limited data and information on gaps and cracks are presented, including observations after long-term storage for 20 years.

(iii) Based on a review and analysis of the stress induced on cladding by thermal expansion of the SNF matrix, an estimate of the stress associated with the irradiation-induced swelling of the SNF matrix during long term storage is presented. The estimated cladding stress from irradiation-induced swelling is sensitive to gap closure as a result of high burnup, crack pattern, and frictions between the matrix and the cladding. Based on this stress estimate, it is recognized that non negligible swelling from alpha decays is observed on doped UO_2 with alpha emitter at accumulated dose levels corresponding to tens and hundred years of SNF storage.

(iv) More information may be needed to confirm this observation through bench-marking with SNF, stress analysis for the storage system, and more Post Irradiation Examination data.

DISCLAIMER

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

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