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## Section 10. Fuel pin and fission product behaviors under accidental conditions High burnup fuel behavior related to fission gas effects under reactivity initiated accidents (RIA) conditions

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## Abstract

Specific aspects of irradiated fuel result from the increasing retention of gaseous and volatile fission products with burnup, which, under overpower conditions, can lead to solid fuel pressurization and swelling causing severe PCMI (pellet clad mechanical interaction). In order to assess the reliability of high burnup fuel under RIAs, experimental programs have been initiated which have provided important data concerning the transient fission gas behavior and the clad loading mechanisms. The importance of the rim zone is demonstrated based on three experiments resulting in clad failure at low enthalpy, which are explained by energetic considerations. High gas release in non-failure tests with low energy deposition underlines the importance of grain boundary and porosity gas. Measured final releases are strongly correlated to the microstructure evolution, depending on energy deposition, pulse width, initial and refabricated fuel rod design. Observed helium release can also increase internal pressure and gives hints to the gas behavior understanding. © 1997 Elsevier Science B.V.

## 1. Introduction

Fuel management optimization leads the utilities to increase the fuel burnup in operating LWR plants. Important effort has been made during the last years to evaluate the high burnup fuel behavior in accidental situations, and particularly under reactivity initiated accidents (RIAs) conditions. To enlarge the existing data base issued from earlier experiments (SPERT and PBF [1]) with low burnup fuel, experimental programs with high burnup fuel are in progress in the nuclear safety research reactor (NSRR) of JAERI (Japan) since 1989 [2], and in the CABRI test reactor of the IPSN (France) since 1993 [3].

The CABRI-REP program is devoted to the study of the high burnup  $UO_2$  fuel behavior under RIA conditions, and additionally of the MOX behavior. The current part of this programme (CABRI REPNa), with sodium as coolant liquid, is focused on the possible occurrence of an early clad loading by PCMI.

Recent experimental results have clearly shown that very high burnup fuel leads to a severe reduction in the clad failure enthalpy threshold comparatively to fresh fuel and to low burnup fuel. In this feature, increasing fuel loading and decreasing clad strength are involved and strongly correlated.

Specific aspect of highly irradiated fuel results from the important accumulation of gaseous and volatile fission products. At an average burnup of 60 GWd/t, the fission gas inventory (Xe, Kr) represents a physical volume equivalent to  $15 \times$  the fuel volume in normal conditions and of the same order as the fuel volume at 2500 K and 14 MPa. In fact, due to the low temperature in operating conditions of a PWR fuel, these fission gases are in a confined form, solution or micro bubbles with high internal pressures (in the range of 1 GPa depending on their size). Under fast overpower conditions, the retained gas can lead to solid fuel pressurization and swelling, and consequently to clad loading by PCMI. This effect is increased in the peripheral region of UO<sub>2</sub> pellets, where the retention of fission gases is multiplied by a factor two, due to the local increase of fissile Pu 239. Furthermore, a typical microstructural change occurs during irradiation in this 'rim' region, char-

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acterized by an extremely fine grained structure and an increasing porosity containing the major part of created fission gases. The change in fuel mechanical properties, connected to the high gas content, can increase strongly the fuel mechanical loading and the risk of an early clad failure, during the heating phase of a RIA. This view is supported by rough energetic considerations.

At the same time, the failure risk is also largely enhanced by the clad embrittlement due to the possibility of hydrogen accumulation and local hydrides spots. Correlations can be established on the basis of the experimental failures and the corrosion and hydriding levels (see Table 1). But it is not the purpose of this paper to enter in these global considerations, for which important effort is made in the French experimental program [15]. In the present study, we analyze the results of the REPNa experiments, and to some extent, the NSRR tests, in order to establish the fuel loading mechanisms, based on the general knowledge of fission gas induced phenomena: fuel cracking and swelling and its impact on PCMI loading, internal rod pressure increase due to fission gas release with further consequences on hot plastic clad after reaching critical heat flux and DNB.

Some specific features of high burnup  $UO_2$  PWR fuel are revealed, especially the importance of grain boundary and porosity gas and the influence of microstructural change on fuel thermal and mechanicals properties.

Finally, this analysis leads to the conclusion that in addition to global testing, investigations are necessary for quantification of the physical mechanisms.

## 2. Experimental results

In Table 1, we summarize the main experimental conditions and results of REPNa and NSRR tests using industrial PWR UO<sub>2</sub> fuel rods. Despite some unrepresentative aspects of these tests correlated mainly to cooling conditions, they provide important data which are characteristic for the high burnup fuel rod behavior under RIA conditions. Major results which are identified from on-line in pile diagnostics or from posttest examinations are

- fuel rod failure which can occur during the early phase of the transient, in fast pulse, and with the most corroded fuel segments,

- clad straining of unfailed rods which increases with fuel enthalpy level,

- final fission gas release, including not only xenon and krypton, but also helium.

Additionally, microstructual changes are observed (fuel fragmentation, microcracking, porosity changes, ...), but they must be analyzed carefully because not necessarily representative of the fuel in hot state and possibly occurring during the cooling phase. The knowledge of the kinetic aspects is one of the major and most difficult problems concerning the fuel behavior under accidental conditions.

# 3. Early phase of the transient: Role of fission gases in PCMI loading

## 3.1. Influence of initial state

A reactivity initiated accident in cold or hot stand-by conditions is characterized by a rapid increase of fuel temperatures in quasi-adiabatic conditions. So, during a short time scale, the radial temperature profile inside the fuel pellet is very near to the radial power profile. The transition from the end-of-life (EOL) to transient fuel temperatures induces mechanical stresses inside the fuel. The solicitation is the most important in the outermost regions of the pellet, and significant even with a relatively low energy deposition (Fig. 1).

The relative effect in outer zones increases with

- the decrease of the pulse width (test pulses are shorter than calculated reactor pulses),

- the burnup, due to the enhancement of Pu 239 production by resonance neutron capture of U 238 in the epithermal energy region.

Additionally, in high burnup fuel a particular microstructure is observed in the peripheral region of the  $UO_2$  pellet (above a local burnup of around 70 GWd/t). The so-called 'rim' structure is characterized by [5–8]

 an apparent loss of definable grain structure with classical optical microscopy, which is revealed as an extremely fine-grained structure with transmission electron microscopy (TEM),

- an increased porosity (more than 20% according to some authors [4,5]),

- a depletion of matrix fission gas deduced from microprobe measurements,

- a high local gas content.

Some mechanisms have been postulated for this restructuring [6-8], but in the present study, we are interested by the final microstructure and its impact on fuel rod behavior under RIA conditions. So, on the basis of the experimental observations presently available, we consider

- Small grains of 0.1  $\mu$ m in radius, corresponding to the average of the experimental observations (0.02-1  $\mu$ m).

- Small intergranular bubbles of 1 nm, coherently with the observations of nanometer size bubbles coating the grain boundaries [6,8]; these bubbles are assumed in equilibrium inside the fuel.

– Large micrometer size overpressurized pores (the pressure disequilibrium can result from the insufficient number of vacancies to allow pores to grow and reach their equilibrium size, coherency between the Xe depletion measured by microprobe and high local gas content implies that the major part of retained gas is present in these large bubbles).

The characteristics of this bimodal bubble repartition are summarized in Table 2, for the case of an average fuel burnup of 65 GWd/t. We also give an evaluation of the associated stored energy. The contributions of small intergranular bubbles and large pores are of the same order, the

| REPNa I         HBO1         HBO2         HBO2         HBO3         HBO3         HBO1         HBO1         HBO2         HBO3         HBO3 <th< th=""><th></th><th>CABRI-REP (flowing so</th><th>dium: 0.5 Mpa</th><th>; 280°C)</th><th></th><th></th><th>NSRR (</th><th>stagnant water: 0.1 MPa; 2</th><th>0°C)</th><th></th><th></th></th<>  |                                       | CABRI-REP (flowing so                     | dium: 0.5 Mpa | ; 280°C) |         |         | NSRR ( | stagnant water: 0.1 MPa; 2 | 0°C) |      |      |
|---|---------------------------------------|---|---------------|----------|---------|---------|--------|----------------------------|------|------|------|
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$   |                                       | REPNa 1                                   | REPNa 2       | REPNa 3  | REPNa 4 | REPNa 5 | GK 1   | HB01                       | HB02 | HB03 | HBO4 |
| Interal pressure (MPa)       0.1       0.01       0.31       0.301       0.302       3.3       3.2       5.1       0.1       0.1       0.1       0.1       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.3       0.4       50.4<   | Initial antichment (M)                |   |               |          |         |         |        |                            |      |      |      |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$   |                                       | C.4                                       | 6.85          | 4.5      | 4.5     | 4.5     | 3.4    | 3.2                        | 3.2  | 3.2  | 3.2  |
| Active length (mm)5691000440567.6563.5122135135135Max. burnup (GWd/t)63.83352.862.364.342.150.450.4Corrosion thickness ( $\mu$ m)80 (spallation)4408020-4850.4Corrosion thickness ( $\mu$ m)9.59.564.342.150.450.450.4Energy deposition <sup>b</sup> (1/g)460882502397439506389213397Plake width (ms)9.59.59.56496444450.4Plake width (ms)9.59.59.564964444Peak fuel enthalpy (1/g)481876522.5414481389305155309Max. mean boop strain (%)early failure (125 1/g)3.52.10.371.112.12.11151Fission gas release (%)5.5413.78.315.112.217.72.27Helium release (mm <sup>3</sup> /g)9.5335.738.22.46n.m.n.m.n.m.n.m = not measured.n.m = not measured.b.m.b.m.b.m.b.m.b.m.b.m.b.m.b.m.b.m.b.m.h.m.b.m.b.m.b.m.b.m.b.m.b.m.b.m.<  | Internal pressure (MPa)               | 0.1                                       | 0.101         | 0.31     | 0.301   | 0.302   | 3.9    | 0.1                        | 5 1  | 10   | 10   |
| Max. burnup (GWd/t) $63.8$ $33$ $52.8$ $62.3$ $64.3$ $42.1$ $50.4$ $50.5$ Corrosion thickness ( $\mu$ m) $80$ (spallation) $4$ $40$ $80$ $20$ $ 48$ $80.5$ Energy deposition <sup>b</sup> ( $J/g$ ) $460$ $882$ $502$ $397$ $439$ $506$ $389$ $213$ $397$ Energy deposition <sup>b</sup> ( $J/g$ ) $460$ $882$ $502$ $397$ $439$ $506$ $389$ $213$ $397$ Energy deposition <sup>b</sup> ( $J/g$ ) $481$ $876$ $522.5$ $414$ $481$ $389$ $305$ $213$ $397$ Pulse width (ms) $9.5$ $9.5$ $9.5$ $522.5$ $414$ $481$ $389$ $305$ $213$ $397$ Pack fuel enthalpy ( $J/g$ ) $481$ $876$ $522.5$ $414$ $481$ $389$ $305$ $177$ $513$ Max. mean hoop strain (%)carly failure ( $125 J/g$ ) $3.5$ $2.11$ $0.37$ $1.11$ $2.11$ $2.11$ $2.11$ $2.11$ $17.7$ $227$ Heilum release ( $mn^3/g$ ) $5.54$ $13.7$ $8.3$ $3.5.1$ $3.8.2$ $4.4$ $1.51$ Heilum release ( $mn^3/g$ ) $5.54$ $13.7$ $8.3$ $2.27$ $2.16$ $n.m.$ $n.m.$ $n.m.$ $n.m.$ Max. mean hoop strain (%) $max (max)^2$ $3.57$ $3.8.2$ $4.14$ $3.99$ $305$ $305$ $305$ Heilum release ( $mn^3/g$ ) $9.53$ $3.5.7$ $3.8.2$ $2.46$ $n.m.$ $n.m.$  | Active length (mm)                    | 569                                       | 1000          | 440      | 567.6   | 563.5   | 122    | 135                        | 135  | 135  | 1.0  |
| Corrosion thickness ( $\mu$ m) <sup>a</sup> 80 (spallation)40.00.0.30.0.450.450.4Energy deposition <sup>b</sup> (J/g)460882502397439506389213397Energy deposition <sup>b</sup> (J/g)9.59.59.564964.45954Pulse width (ms)9.59.564964.4694.4Rask fuel enthalpy (J/g)481876522.5414481389305155309Max. mean hoop strain (%)early failure (125 J/g)3.52.10.371.112.1early failure (251 J/g)0.411.51Fission gas release (%)5.5413.78.315.11.2217.722.7Helium release (mm <sup>3</sup> /g)9.5335.738.24.31n.m.n.m.n.m.Aanswinun value.0.532.272.46n.m.n.m.n.m.  | Max. burnup (GWd /t)                  | 63.8                                      | 33            | 57 8     | 613     | 643     | ç      |                            |      |      | 001  |
| Corrosolu uncorress (µm) 80 (spatiation) 4 40 80 20 - 48 38 25<br>Energy deposition $^{b}(J/g)$ 460 882 502 397 439 506 389 213 397<br>Pulse width (ms) 9.5 9.5 64 9 6 4.4 6.9 213 397<br>Pulse width (ms) 9.5 9.5 11.1 2.1 early failure (251 J/g) 0.41 1.51<br>Fission gas release (%) 5.54 13.7 8.3 15.1 1.2.1 early failure (251 J/g) 0.41 1.51<br>Heitum release (mm <sup>3</sup> /g) 9.53 35.7 38.2 43.1 n.m. n.m. n.m. n.m. n.m. n.m. n.m. n.  |                                       |   | ,<br>,        | 0.76     | C.20    | 04.0    | 47.1   | 50.4                       | 50.4 | 50.4 | 50.4 |
| Energy deposition $^{b}(J/g)$ 460882502397439506389213397Pulse width (ms)9.59.59.56.4964.46.94.4Peak fuel enthalpy (J/g)481876522.5414481389305155309Max. mean hoop strain (%)early failure (125 J/g)3.52.10.371.112.1early failure (251 J/g)0.411.51Fission gas release (%)5.5413.78.315.112.20.411.51Helium release (mm <sup>3</sup> /g)9.5335.738.24.31n.m.n.m.n.m.n.m.I. = not measured.n.m. = not measured.  | COLLOSION UNCKNESS (µm) -             | 80 (spallation)                           | 4             | 40       | 80      | 20      | I      | 48                         | 38   | 25   | 22   |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$   | Energy denosition <sup>b</sup> (I /a) | 160                                       | 600           | 003      |         |         |        |                            |      |      |      |
| Pulse width (ms)9.59.564964,46.94,4Peak fuel enthalpy $(J/g)$ 481876522.5414481389305155309Max. mean hoop strain (%)early failure (125 J/g)3.52.10.371.112.1early failure (251 J/g)0.411.51Fission gas release (%)5.5413.78.315.112.217.722.7Helium release (mm <sup>3</sup> /g)9.5335.738.243.1n.m.n.m.n.m.n.m.I. = not measured.* Maximum value.* Maximum value. </td <td>Envision (1/g)</td> <td></td> <td>202</td> <td>700</td> <td>165</td> <td>439</td> <td>506</td> <td>389</td> <td>213</td> <td>397</td> <td>280</td>  | Envision (1/g)                        |   | 202           | 700      | 165     | 439     | 506    | 389                        | 213  | 397  | 280  |
| Peak fuel enthalpy (J/g)       481       876       522.5       414       481       389       305       155       309         Max. mean hoop strain (%)       early failure (125 J/g)       3.5       2.1 $0.37$ $1.11$ 2.1       early failure (251 J/g) $0.41$ $1.51$ Fission gas release (%) $5.54$ $13.7$ $8.3$ $15.1$ $12.2$ $17.7$ $22.7$ Helium release (mm <sup>3</sup> /g) $9.53$ $35.7$ $38.2$ $43.1$ $n.m.$   | Fulse width (ms)                      | 5.6                                       | 9.5           | 9.5      | 64      | 6       | 9      | 4.4                        | 6.9  | 4.4  | 53   |
| Max. mean hoop strain (%)       carly failure (125 1/g)       3.5       2.1       0.37       1.11       2.1       early failure (251 1/g)       0.41       1.51         Fission gas release (%) $5.54$ $13.7$ $8.3$ $15.1$ $12.2$ $20.3$ $0.41$ $1.51$ Helium release (mm <sup>3</sup> /g) $9.53$ $35.7$ $8.3$ $15.1$ $12.2$ $0.41$ $1.77$ $22.7$ He release (mm <sup>3</sup> /g) $0.53$ $35.7$ $38.2$ $43.1$ $n.m.$ $n.m$   | Peak fuel enthalpy (J/g)              | 481                                       | 876           | 522.5    | 414     | 481     | 389    | 305                        | 155  | 300  | 000  |
| Fiscing as release (%) $(\%)$ | Max mean hoon strain (%)              | early failure (135 I $/\alpha$ )          | 35            | ,<br>,   |         |         |        |                            | CC1  | 600  | 502  |
| rission gas release $(\%)$ 5.54       13.7       8.3       15.1       12.2       17.7       22.7         Helium release $(mn^3/g)$ 9.53       35.7       38.2       43.1       n.m.   |                                       | carry rainer (12.) / g)                   | C.C           | 2.1      | 1.3/    | 1.11    | 2.1    | early failure (251 $J/g$ ) | 0.41 | 1.51 | 0.2  |
| Helium release (mm <sup>3</sup> /g)       9.53       35.7       38.2       43.1       n.m.  | rission gas release (%)               |   | 5.54          | 13.7     | 8.3     | 15.1    | 12.2   |                            | 17.7 | 7.7  | 110  |
| He release/(Xe + Kr) formation (%) 1.28 2.48 2.27 2.46 n.m. n.m. n.m. n.m. n.m. n.m. n.m. n.m   | Helium release (mm <sup>3</sup> /g)   |   | 9.53          | 35.7     | 38.2    | 43.1    | n.m.   | n.m.                       |      |      |      |
| n.m. = not measured.<br><sup>a</sup> Maximum value.   | He release $/(Xe + Kr)$ format        | ion (%)                                   | 1.28          | 2.48     | 2.27    | 2 46    | a<br>a |                            |      |      |      |
| n.m. = not measured.<br><sup>a</sup> Maximum value.   |                                       |   |               |          |         | 24-14   |        | п.ш.                       | п.п. | n.m. | n.m. |
| a Maximum value.  | n.m. = not measured.                  |   |               |          |         |         |        |                            |      |      |      |
|   | <sup>a</sup> Maximum value.           |   |               |          |         |         |        |                            |      |      |      |
| HOT V HUND tacks of 1/ a propert fam DIPDNI- 4 -1 1 O   | <sup>b</sup> Eor DEDNo tests of 0.4 c | $\frac{1}{1000} f_{000} DEDNI_{-1} = 1.0$ |               |          |         |         |        |                            |      |      |      |

Table 1 Characteristics and results of REPNa and NSRR tests with commercial PWR rods  $(\mathrm{UO}_2)$ 

lower gas concentration being compensated by the higher internal pressure linked to the smaller bubble size. We will see later that internal pressure and stored energy are probably the most important parameters involved in the loading mechanisms of the rim postulated in fast and energetic transients. The effective contribution of fission gas will depend on the possibility to liberate the stored energy. This feature is one of the key points for the basic understanding of fission gas induced phenomena under fast power transients.

## 3.2. Transient behavior

Under RIA conditions, the heating rate is very high: about  $10^5$  K/s. So, no sufficient time is available to

activate diffusion phenomena, even at high temperatures. This induces an important overpressurization inside grain boundary bubbles, and a high stress field between the grains in a still brittle fuel. A schematic view of the phenomena is represented on Fig. 2. Grain boundary rupture occurs if the fracture stress is exceeded. Clear evidence of this 'brittle' fuel fragmentation has been obtained from the SILENE experiments with LMR fuel [9], at a temperature threshold of 2300–2400 K, leading, in the absence of significant fuel restraint, to energetic dispersal of fine-grained fuel at large velocities (about 10 m/s). A criterium for solid fuel breack-up by cracking of the grain boundaries has been developed by WORLDLEGE [16] involving stress, energy and mass transfer conditions. Stress



Fig. 1. Radial temperature profile of high burnup fuel (62 GWd/t) in RIA transient [maximum fuel enthalpy 335 J/g (80 cal/g)].

considerations, depending on the excess pressure inside bubbles and on the grains surface coverage could be sufficient to explain most of the NSRR and CABRI experimental observations.

In a first approximation, the excess pressure can be expressed by

$$P_{\rm bexc} = P_{\rm bo}(T/T_{\rm o} - 1), \tag{1}$$

where  $P_{bo}$  is the pressure at EOL and  $T_o$ , T are the fuel temperature respectively at EOL and during the transient. So, the threshold for grain fragmentation is lower in PWR fuel than in LMR fuel due to the lower temperature under irradiation. A value of 1300 to 1500 K can be estimated, assuming similar fuel mechanical properties. This result is coherent with experimental observations: grain boundary separation has been observed in the outer zones of NSRR and REPNa tests (see example on Fig. 3), with relatively low fuel enthalpy levels, and the width of this zone increases with energy deposition.

Nevertheless, we must keep in mind that this phenomenon is not necessarily perfectly visible on the posttest examinations. Indeed, the final structure is a complex mixing of this initial grain boundary fragmentation, intergranular gas flowing and stress field during the postfragmentation phase.

In the rim region, the high porosity induces a lower fracture stress. Indeed, an increase of porosity of  $UO_2$ 

from 5 to 20% causes a 80% reduction in failure strength [10]. So, it is possible to obtain a grain boundary cracking in the rim region at a low temperature (around 900 to 1000 K).

This fragmentation mechanism is the first condition for a fast liberation of the energy stored in intergranular bubbles. But we must regard now if this phenomenon can lead to an effective contribution of fission gases to the PCMI loading during the heating phase.

After grain boundary fragmentation, the work of expanding gas is transformed into clad deformation (Fig. 2). If  $\sigma_r$  is the rupture stress of the clad, equivalent to an internal hydrostatic pressure  $P_r = (2e_g/d_g) \sigma_r$ , and  $\varepsilon_r$ , the corresponding rupture strain, the condition for clad rupture, expressed in linear energy, is roughly given by

$$m_{\rm L} \times \frac{1}{\rho} \sum_{i=b, p} P_{\rm bi} g_i \log \frac{P_{\rm bi}}{P_{\rm r}} > \frac{2e_{\rm g}}{d_{\rm g}} \times 2 \prod r_{\rm g}^2 \int_{\varepsilon_{\rm th}}^{\varepsilon_{\rm r}} \sigma \, \mathrm{d}\varepsilon,$$
(2)

where  $m_{\rm L}$  is the linear mass of the fragmented zone,  $e_{\rm th}$  is the contribution of differential thermal dilatation (fuel-clad) to clad deformation,  $P_{\rm bi}$  is the pressure of the bubble group *i* (b = intergranular bubbles, p = large pores), at fragmentation temperature (or at a later time during the heat-up phase),  $e_{\rm g}$ ,  $d_{\rm g}$  are the thickness and the mean diameter of the clad.

Table 2

Characteristics of grain boundary gas in fuel outer region and evaluation of expansion work after grain fragmentation

|                                      | Average burnup        |             |                       |             |  |  |
|--------------------------------------|-----------------------|-------------|-----------------------|-------------|--|--|
|                                      | 40 GWd/t              |             | 65 GWd/t              |             |  |  |
| End of life state                    |                       |             |                       |             |  |  |
| Local burnup (GWd/t)                 | 57                    |             | 120                   |             |  |  |
| Fission gas content $(mm^3/g)$       | 1767                  |             | 2795                  |             |  |  |
| Fuel temperature (K)                 | 673                   |             | 673                   |             |  |  |
|                                      | Bubble population     |             |                       |             |  |  |
|                                      | intergranular bubbles | large pores | intergranular bubbles | large pores |  |  |
| End of life state                    |                       |             |                       |             |  |  |
| Bubble radius (nm)                   | 2.5                   |             | 1                     |             |  |  |
| Relative volume (%)                  | 0.019                 | 4.5         | 0.5                   | 19.5        |  |  |
| Gas content $(mm^3/g)$               | 5.83                  | 157         | 191                   | 2185        |  |  |
| Internal pressure ((Mpa)             | 496.6                 | 10          | 1228.4                | 40.16       |  |  |
| Stored energy $(J/g)$                | 0.0135                | 0.0643      | 0.877                 | 1.119       |  |  |
| Transient behaviour ( $P_r = 80$ Mpa | .)                    |             |                       |             |  |  |
| Fuel temperature (K)                 | 2900                  | 2900        | 900                   | 900         |  |  |
| Internal pressure (Mpa)              | 2140                  | 43.09       | 1642.7                | 53.7        |  |  |
| Expansion work $(J/g)$               | 0.127                 | -0.114      | 2.364                 | - 0.398     |  |  |
| Total <sup>a</sup>                   | 0.013                 |             | 1.966                 |             |  |  |
| Fragmented zone (vol.%)              | 0.5                   | 0.5         | 0.05                  | 0.05        |  |  |
| Expansion work (J/m)                 | 34.7                  | -31.2       | 64.2                  | - 10.8      |  |  |
| Total <sup>a</sup>                   | 3.55                  |             | 53.4                  |             |  |  |

<sup>a</sup> Sum of expansion work from intergranular bubbles and large pores.



Fig. 2. Fuel and clad behaviour in RIA transients.

The integral on the right hand side introduces the concept of strain energy density, recently used in the analysis of RIA experiments [11]. It represents normally the mechanical work performed by the combined states of stress imposed to the cladding. In this approach, the three stress and incremental strain components (radial, hoop and axial) are theoretically included. Nevertheless, the radial stress is compressive, and so is not taken into account. Besides, major contribution comes from the hoop stress and strain, so the axial component can be neglected. Finally, we obtain a result coherent with the present approach, which is justified in the case of a biaxial loading.

In case of fully elastic behavior of the clad (no plastic deformation), the second term of Eq. (2) has the simplified expression

$$\Pi e_{\rm g} r_{\rm g} \Big[ \sigma_{\rm r} \varepsilon_{\rm r} - E \varepsilon_{\rm th}^2 \Big], \tag{3}$$

where  $r_{\rm g}$  is the clad mean radius and E is Young's modulus.

On another side, from Eq. (2), it is clear that a necessary condition for an effective contribution of fission gases to clad loading is that the left term must be positive.

Two cases have been considered: the case of a fuel just before the rim formation, with an average burnup of about 40 GWd/t, and the case of a higher burnup fuel of 65 GWd/t, with a relatively well established rim microstructure as described previously. For the lower burnup, we assume intergranular bubbles of 2.5 nm in radius, consistent with experimental observations of Ref. [6], a grain coverage fraction of 0.5, and large porosities in equilibrium with plenum (see 4). The evaluation of expansion work is done at a high temperature (2900 K), near the melting point, and assuming no gas escape after the grain boundary fragmentation (which increases the effect of fission gases). Furthermore, we consider a fragmented zone of 50% of fuel volume. Concerning the higher burnup fuel, the evaluation is done at the grain boundary fragmentation temperature (900 K), and only the rim zone is taken into account (about 5% of fuel volume).

As rupture conditions, we have taken a value of  $P_r = 80$  MPa, corresponding to usual irradiated clad laws.

The results are indicated in Table 2. There is roughly a factor 10 between the potential action from fission gases of medium burnup and high burnup fuel, even with the same conditions for clad rupture stress.

From this evaluation, two important conclusions are obtained.

(1) The contribution of fission gases to PCMI loading during the fast heating phase of the RIA is negligible as far as medium burnup is concerned ( $\leq 40$  GWd/t). So, the risk of clad failure during this phase seems unlikely with standard LWR fuel. But clad failure can occur later with additional energy coming from intragranular bubble migration or due to swelling, with contribution of the most inner zones. That implies however high temperatures and thermal gradients, resulting from high energy deposition. Most

of the clad failures observed in the fast RIA tests with low and medium burnups (JMTR rods [12], SPERT-CDC, PBF [1]) occur during this phase from an enthalpy level around 150 cal/g (or 627 J/g).

(2) In higher burnup fuel, with the rim formation, important energy can be liberated from fission gas bubbles after grain boundary fragmentation, increasing strongly the risk of clad failure during the heating phase, at a low enthalpy level. But clearly, this occurrence depends also on the clad mechanical properties and particularly its brittleness. In REPNa 1 (Table 1), there is a clear evidence of the brittle character of the first failure for a fuel temperature of about 900 K in the rim zone. Assuming clad failure at yield level, which gives an upper value for the clad rupture strain (0.2%), and so for the energy needed, we obtain a value of 9.77 J/m for the linear energy required for clad failure ( $\varepsilon_{th} = 6 \times 10^{-3}$ ). Comparison with energy available from expanding gas: 53.4 J/m shows that the eventuality of an early clad failure is true, in case of degradation of clad mechanical properties (brittle behavior without residual ductility). Failure in REPNa 1 (see Table 1) is to be explained by such results. The



Fig. 3. Micrograph of high burnup fuel after RIA test (REPNa 5).

non-failure in REPNa 5 with similar high burn-up fuel but less corroded cladding is also consistent with this approach (higher linear energy for clad failure due to higher clad ductility).

## 4. Fission gas release

In the NSRR and CABRI tests, high fission gas release has been observed with commercial fuel rods refabricated or not, increasing with fuel burnup (Table 1). Releases up to 20% are obtained with low energy deposition (see for example HBO4). At these enthalpy levels, diffusion phenomena are theoretically not or only slightly activated. This is confirmed by the microprobe examinations on REPNa 1 which show no significant change before and after test [13]. So we can conclude that the gas release is issued in major part from porosity and grain boundary bubbles.

The intergranular gas concentration is limited by the specific area of the grains. In a previous paper [14], an upper value has been obtained assuming lenticular form and equilibrium conditions inside the fuel. The maximum intergranular gas content decreases with increasing grain radius and temperature, and increases with bubble radius. But, in the range of the bubble sizes observed in a PWR fuel (about 50 nm in the inner zones, 1 to 10 nm elsewhere [6]), this content is low and represents less than 1% of retained gas in high burnup fuel outside the rim region (see also Table 2).

The rim zone can contribute significantly to the gas release, but only for very high burnup fuel (about 65 GWd/t). In the case of medium burnup like GK1 fuel (42 GWd/t), the rim is just incipient and cannot explain a released gas of 12%.

So, the most probable conclusion is that the gas release results mainly from the gas accumulated in porosities in EOL: as fabricated porosities in which some fission gases have accumulated or corner pores formed during irradiation. In order to estimate the fission gas quantity in porosities, we have assumed in Ref. [14], that the porosities were in equilibrium with the plenum pressure. This leads to a significant quantity of gas (until about 10% of retained gases, see for example Table 2) and could explain the high gas release measured in medium burnup and moderately high burnup fuel with low energy deposition (see GK1 and REPNa 3 in Table 1). Consequently, even if this assumption is difficult to justify theoretically, considering the low diffusivity of fission gases at operating temperatures of a PWR fuel, we have to keep this assumption. Indeed, it leads to satisfactory agreements with experimental results for high burnup fuel, and we can assume that, even if it is not fully true locally, it represents the mean state of the porosities in the fuel.

In higher burnup fuel, a significant contribution of the rim region to the observed gas release can be postulated. Table 3

Correlation between fission gas release and structural pellet change in transient

| REPNa test                                 | REPNa 4         | REPNa 5         |
|--|-----------------|-----------------|
| Free volume before test (mm <sup>3</sup> ) | 2261            | 2426            |
| Clad deformation after test                | 0.17 (16.5 µm)  | 0.754           |
| (mean) (%)                                 |                 |                 |
| Free volume after test: calculation        | 2400            | 3026            |
| Free volume after test + closed            | ~ 3790          | ~ 4550          |
| Free volume after test: measure            | $2300 \pm 1600$ | $4710 \pm 1500$ |
| Fission gas release (%)                    | 8.3             | 15.1            |

From Table 2, we can see that intergranular and porosity gas of the rim region represent 85% of the local gas content, that means about 7% of the average retention in the pellet. After grain boundary fragmentation and dynamic loading of the clad, these fission gases escape from the fuel. This view might be supported by posttest examinations which show a relatively compact rim pressed against the clad, in case of non-failure test (REPNa 5, Fig. 3); this is a significant change of morphology compared to the state before the test.

Finally, if we add up the gas content in porosities and intergranular bubbles in and out the rim region, we obtain a value of about 15%, so fully coherent with the released gas measured in the fast pulse REPNa 5.

With the slower pulse REPNa 4, a significantly lower gas release is observed. So we have to investigate in more detail the influence of the pulse width on the fission gas release.

Furthermore, correlations can be established between the maximum clad strain, the free volume evolution and the final gas release (Table 3). Even if the uncertainty on the free volume measurements is high, it seems clear that:

(1) after the tests, most of the closed porosities are opened in REPNa 5 and still closed in REPNa 4 (but that does not exclude a partial temporary opening during the transient),

(2) this phenomenon is correlated to the clad deformation and to the fission gas release.

These results underline the complexity of the gas release mechanisms. In fast transients, the determinant step for fission gas release to the plenum, is the gas transfer inside the more or less interconnected porosity network (including fuel-clad gap). So it depends on the fuel permeability, itself correlated to the open porosity. In RIAs, the state of porosity (opened or closed), changes all along the transient, due firstly to the mechanical stresses induced by the thermal profile evolution, and secondly to the grain boundary decohesion (fragmentation and/or fission gas induced swelling). The global porosity evolution is correlated to the available free volume and so to clad strain. In case of low energy deposition (< 500 J/g) the major part



Fig. 4. Influence of fuel thermal history on clad strain (fast pulse REPNa 3).

of the clad strain occurs during the early phase of the transient due to the highest fuel temperatures obtained in outer zones during this step (Fig. 4). Significant contribution of fission gas induced swelling from inner zones during the late phase is significant only with high energy deposition (REPNa 2 case).

In REPNa 4 the slower pulse leads to a decrease of maximum fuel temperature (Fig. 1) and so to lower clad strain. Finally, even if the fission gas release is not correlated to the temperature by classical diffusion processes inside grains, it depends nevertheless on the fuel thermal level by its impact on microstructural changes. Pulse width seems a more directly important parameter than power level (see results for HBO tests in Table 1), probably due to the influence of the maximum temperature location (Fig. 1) and its consequences on micro-cracking in outer zones during the cooling phase.

Furthermore, results of Table 1 show that, under fast pulse conditions, the gas release is lower in REPNa than in HBO tests. Several explanations can be postulated:

- The pulse width: In HBO tests, the gas release increases when pulse width decreases; but it is also correlated to the energy deposition increase, so it is difficult to conclude. – The height of the refabricated fuel rod: There is a factor of about 4 between REPNa and HBO; the characteristic time for gas flow is proportional to  $L^2/D$  where L represents the mean flow length and D is equivalent to a diffusion coefficient (depending on pressure, permeability and gas viscosity).

- The fuel state at EOL: Gas release is issued in major part from porosities, considered in equilibrium with plenum pressure; the gas content will depend therefore on the fuel fabrication characteristics, on the densification or other porosity changes under irradiation, and on the EOL pressure.

Besides, if it is necessary to evaluate the final gas release, more important is the knowledge of the kinetics which influences the thermal and mechanical rod behavior. Specially, the delays between local gas release, gas release in the gap, and finally the escape to the plenum are of importance:

- The gas released from the grains, but still present inside the fuel can contribute to PCMI; if it leads to porosity increase, it can degradate the fuel thermal conductivity, this is already the case in the porous rim zone at the beginning of the transient, but it can lead to an opposite effect in case of rim compaction.

- The fission gases accumulated in the gap degradates the fuel-clad thermal exchanges with further consequences on fuel and thermal evolutions; they can induce also locally high gap pressure which reduces the gas transfer from the fuel and can affect the mechanical loading.

– The plenum pressure increase resulting from the final gas release causes clad ballooning and subsequent burst failure in case of significant increase of clad temperature ( $\geq 800^{\circ}$ C); local effects could be also envisaged: these phenomena require a sufficient energy deposition, but anyway, cannot be studied from the present experimental conditions of the NSRR and CABRI tests.

So additional efforts must be made to obtain experimental data on the gas release kinetics in realistic conditions. Presently, only qualitative assessments can be made. If it can be assumed that some gas release takes place during the heating phase from the porosities, the major release occurs probably during the cooling phase, by fuel cracking and stress relaxation. Quick cool-down in outer zones induces radial micro-cracks, making easier the gas transfer to the closed gap from the fragmented zones. But finally, the overall gas release is obtained during the later phase of cooling by fuel cracking and/or gap reopening.

#### 5. Helium behavior

In the REPNa tests, a characteristic release of helium has been observed (Table 1), deduced from the difference between the final content measured after puncturing and the initial content estimated from filling conditions (free volume, temperature and pressure). It is to be concluded that a significant quantity of helium is occluded in PWR fuel. This result is confirmed by some results of overall gas retention measurements by fuel sublimation, plotted on Fig. 5.

Helium in nuclear fuel results from two sources: the filling gas, and the production by ternary fissions, alphadecay of heavy nuclides and  $(n, \alpha)$  reactions. The range of production is generally considered as 1 to 2% of the fission gas production. In UO<sub>2</sub>, a major part of the production comes from ternary fission, the percentage increases with burnup, because the probability of a ternary fission is more important with Pu 239 than with U 235 [17]. The amount of helium produced by alpha-decay is negligible at low burnup in UO<sub>2</sub>, but it increases with burnup due to the buildup of Pu. The main source comes from the decay of Cm 242 produced from Pu 241. Production by alpha-decay goes on during periods at zero power, so also during the cooling time between EOL and RIA test or analysis. The production from  $(n, \alpha)$  reaction (with O16), is negligible. Rough estimation of the total helium production has been done in the case of REPNa 1, leading to a value of 1.2% of the fission gas production (1% by ternary fission and 0.2% by alpha-decay). Even if this result is preliminary, this value is less than the helium release measured in REPNa 4 and REPNa 5 (Fig. 5).

We can also envisage, as other origin of helium occluded in the fuel, some part of filling gas. That could be due to implantation and diffusion processes via gap and cracks, and to the relatively important mobility of helium during reactor operating conditions. This assumption is consistent with the quasi systematic lower amount of helium measured after puncturing than the initial amount estimated from the filling conditions. Though the uncertainty on the helium filling is high (about 15%) and of the order of helium production in pressurized rod until burnup of about 55 GWd/t, it becomes lower in higher burnup fuel, and helium production should be considered in this case.

The state of knowledge on helium is illustrated on Fig. 5. The volume of helium occluded in the fuel is in the range of fission gas release volume occurring during normal reactor operating conditions.

Under RIA conditions, the helium release can contribute significantly to the internal rod pressure increase (about 15% taking into account the results of REPNa 4 and REPNa 5). It could also have some influence on the thermal fuel rod behavior, due to its better conductivity than fission gases xenon and krypton.

## 6. Conclusion

The increasing amount of gaseous and volatile fission products with burnup generates a potential for fuel swelling and expansion work under rapid heating conditions. This feature characterizes the high burnup fuel behavior under power excursion accidents by comparison to fresh fuel.

The analysis of results obtained from the global RIA tests in CABRI and NSRR, leads to a better understanding of the transient loading and of some specific observations.

The assessment of an early clad failure can be done in terms of energy balance between the transient fuel loading by fission gas and the reduced clad strength due to hydriding. Nevertheless, with regards to these aspects, no dynamic effect is presently considered. Highly overpressurized bubbles can behave like hot spots, leading to additional mechanical loading.



Fig. 5. Helium balance in UO<sub>2</sub> (EOL and RIA transient).

The high gas release after tests is the proof that a significant part of the retained gas is located inside the high burnup fuel porosities. Under overpower conditions, porosity gas can easily escape from the fuel by microcracking. However, this implies open flow pathes and/or significant time for gas transfer inside the fuel rod. So, the fission gas release is strongly correlated to the fuel microstructure evolution, induced by the thermomechanical behavior, and depends on energy deposition, pulse width and fuel rod design (height/radius ratio).

Helium production could become significant in high burnup fuel, and contributes to the inner rod pressure increase under transient heating.

We must state that for some aspects, precise knowledge is missing and must be improved:

- The mode and form of gas retention inside fuel, and in particular inside porosities.

- The production and the behavior of helium under irradiation conditions.

- The grain boundary fragmentation threshold and the influence of the microstructure change in the rim region on thermal and mechanical fuel properties.

- The kinetics of fission gas release and the impact of the fuel rod design and of the pulse width.

- The influence of representative reactor conditions (pressure and coolant), during the RIA experiments.

Finally, results from application oriented tests with great importance for reactor licensing reveal the insufficient state in the basic knowledge of the high burnup fuel: Fission gas behavior under irradiation conditions and fuel mechanical properties. Fast transient heating and high system pressures are additional parameters to be considered for the correct understanding of fission product effects under accident conditions.

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