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Estimation of Pore Pressure in the Rim Region of High Burnup UO_2 Fuel

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Abstract

An attempt has been made to estimate the pore pressure in the rim region of high burnup UO_2 fuel as a function of rim burnup using the measured rim width, average porosity and pore density in the rim region. First, a linear relationship is developed based on measured rim burnup and rim width. Second, fraction of fission gas retained in the grain boundary of rim region is estimated. Third, total pores in the rim is calculated from the measured pore density in the rim region. Finally using the assumption that all the pores in the rim have the same size of 1.2μm, pore pressure is calculated from the equation of state for ideal gas. An estimated pore pressure of about 60 to 80 MPa for the rim burnup of 90 GWd/tU appears to be in reasonable agreement with other value given in a literature that pore pressure at 800 K become 90-210 MPa for pellet average burnup of 80 GWd/tU.

1. Introduction

In order to accomodate longer refueling cycles and to extend fuel burnup per unit energy production, the assembly average discharge burnup of LWR fuel has been increased in recent years from typical values of 40 GWd/tU to typical values of 50 to 60 GWd/tU in some countries. In other countries, similar applications are being made by utilities or currently under review by regulatory authorities.

Up to now, analysis and operating experience show that the performance of high burnup fuel seem to be acceptable under normal operating conditions. However, the behavior of the fuel under transient conditions, in particular during reactivity initiated accidents (RIA), requires careful consideration. For example, experimental programs performed in the CABRI test reactor (France) and in the Nuclear Safety Research Reactor (NSRR) (Japan) appear to indicate that cladding failures and fuel dispersion of high burnup fuel may occur at enthalpy values lower than previously estimated.

Fission gas release at high burnup fuel is one of the most important factors that can limit the performance of high burnup fuel in the reactor through the increased gas release and rod internal gas pressure during operational transients. In addition, the presence of rim structure in high burnup fuel, whose characteristics are 1) development of a subgrain microstructure with a typical grain size of 0.2 to 0.3μm, 2) development of fission gas bubbles with a typical diameter of 1-2μm, and 3) Xe depletion of the matrix [1], requires that special attention be given to this region when fuel behavior is analyzed in terms of gas release because large number of gas atoms are available for release during RIA. Fuel temperature in the rim region increases rapidly from coolant temperature to about 600^{o}C in a typical RIA in LWR that might occur during hot standby condition [2]. This accident might produce gas bubble pressure that is high enough to cause crack propagation along subgrain boundaries and consequently lead to ventilation of gas atoms in the bubbles. This could ultimately increase rod internal pressure and cause fuel failure in the cladding area where its mechanical properties might have been degraded due to corrosion or hydride. Therefore, based on the experimental data accumulated up to now, an attempt has been made in this paper to estimate the pressure of rim pores as a function of rim burnup under RIA conditions.

2. Thickness of the rim region

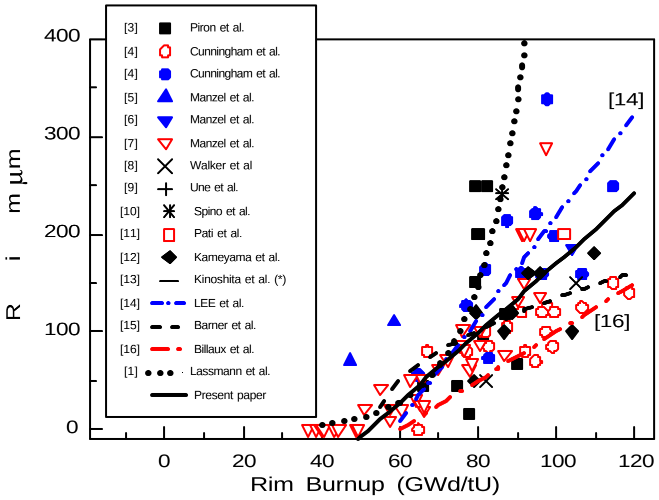
The data on the thickness of the rim region, which is usually defined by the radius at which the relative Nd concentration representing Xe production and the Xe concentration begin to diverge, has been measured by numerous investigators both by EPMA and optical microscopy [3-13]. In addition, several correlations are available which express the rim width as a function of pellet average burnup or rim burnup [1,14-16]. Using all the relevant informations, a new correlation which describes the rim width in terms of rim burnup has been developed as follows:

$$R_{rim} = 3.55\, BU_{rim} - 185 \tag{1}$$

where R_{rim} is the rim width in μm and BU_{rim} is the rim burnup in GWd/tU. Fig.1 shows that a linear formula of Eq.(1) represents a best-estimate relationship between rim width and rim burnup up to 120 GWd/tU. Eq.(1) indicates that the threshold local burnup for rim formation is about 52 GWd/tU. This value is slightly lower than the threshold burnup of 56 GWd/tU that was used by Lee et al. [14]. On the other hand, a linear conservative correlation that covers all the data shown in Fig.1 is

$$R_{rim} = 5.28\, BU_{rim} - 178. \tag{2}$$

At the moment there is some controversy over whether the rim width would increase linearly or exponentially with fuel burnup. While some data show that rim width increases linearly with burnup, others support the exponential growth of the rim region with burnup. However, Fig.1 reveals that the data accumulated so far are appropriately fitted with a linear relationship up to 120 GWd/tU. The filled symbols in Fig.1 represent the data measured by EPMA and the closed ones show the data measured by optical microscopy. There is a general trend that rim width measured by EPMA is higher than that obtained by optical microscopy.



(*) [13] Kinoshita et al.: 7.9% FIMA (99.6 GWd/tU), 1650µm

Fig.1. Rim width as a function of rim burnup.

It is to be noted that Kinoshita et al. [13] reported a very wide rim width of 1650μm for 7.9% FIMA (99.6 GWd/tU) pellet. At present it is not clear how the wide rim width of this value was achieved for a local fuel temperature of 1200°C.

3. Fraction of fission gas in the rim region

Lassmann et al. [1] expressed the Xe concentration in the matrix of rim region Xe_m (wt%) as follows:

$$Xe_m = \dot{c}\left[\frac{1}{a} + \left(bu_{rim}^{0} - \frac{1}{a}\right) e^{-a(bu_{rim} - bu_{rim}^{0})}\right] \tag{3}$$

where $\dot{c}$ is the Xe production rate in wt% per unit burnup (1.46 x10^{-2} wt% per GWd/tU), a is a fitting constant of 0.0584, bu_{rim} is the rim burnup in GWd/tU and bu_{rim}^{0} is the threshold burnup for rim formation in GWd/tU. Therefore, the Xe concentration in the grain boundary of a newly created subgrain Xe_g (wt%) is

$$Xe_g = \dot{c} \cdot bu_{rim} - \dot{c}\left[\frac{1}{a} + \left(bu_{rim}^{0} - \frac{1}{a}\right) e^{-a(bu_{rim} - bu_{rim}^{0})}\right] \tag{4}$$

Using Eqs.(3) and (4), Xe concentration in the matrix and in the grain boundary of the rim region was calculated as a function of rim burnup for 4 different threshold burnups for rim formation (52, 60, 65, and 70 GWd/tU) and they are shown in Fig.2 and Fig.3, respectively. The Xe atoms in the grain boundary of the rim region exist as pores with diameter as shown in Fig.7. The lower the threshold burnup for rim formation, the more Xe atoms are available in the grain boundary of the rim region as expected.

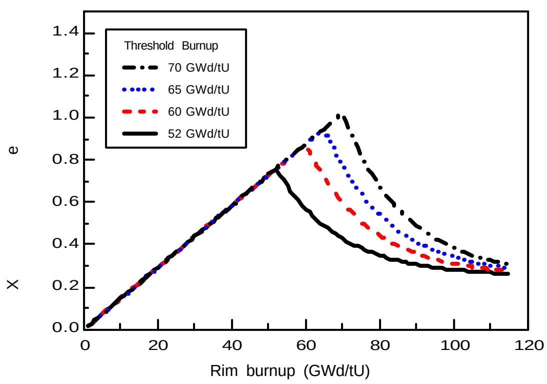


Fig.2. Calculated Xe concentration in the matrix of the rim region for various threshold burnup for rim formation.

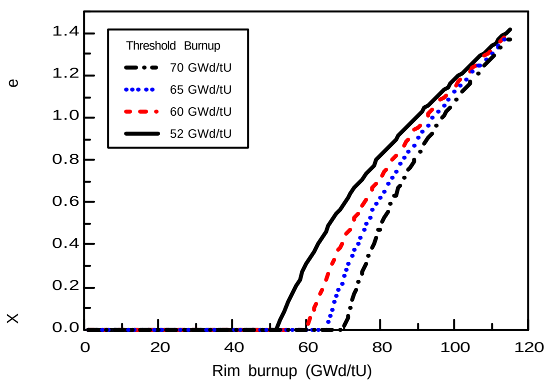


Fig.3. Calculated Xe concentration in the grain boundary of the rim region for various threshold burnup for rim formation.

Fractional gas retention in the rim pores is calculated as follows:

$$f_{rim} = \frac{Xe_g}{Xe_m + Xe_g} \cdot \frac{Rim\ volume \cdot \rho_{TD}}{Pellet\ volume \cdot \rho_r} \cdot (1 - P_{rim}^{avg}/100) \cdot B_r \tag{5}$$

where ρ_{TD} is the theoretical density of fuel pellet, ρ_r is the real density of pellet, P_{rim}^{avg} is the average porosity in the rim (Fig.6) and B_r is the ratio of rim burnup to pellet average burnup. B_r is assumed to be 1.43 [14]. Fig.4 shows the calculated fraction of Xe retained in the rim region for the total Xe produced in a pellet using the best-estimate and conservative correlation for rim width, respectively. For a pellet average burnup of 80 GWd/tU (corresponding rim burnup is about 114 GWd/tU), Xe fraction in the rim available for release under RIA conditions is 10 to 20%. The calculated maximum value of 20% for pellet burnup of 80 GWd/tU is comparable to the fission gas retention in coarsened rim pores estimated from Xe profiles of EPMA [6,8-9].

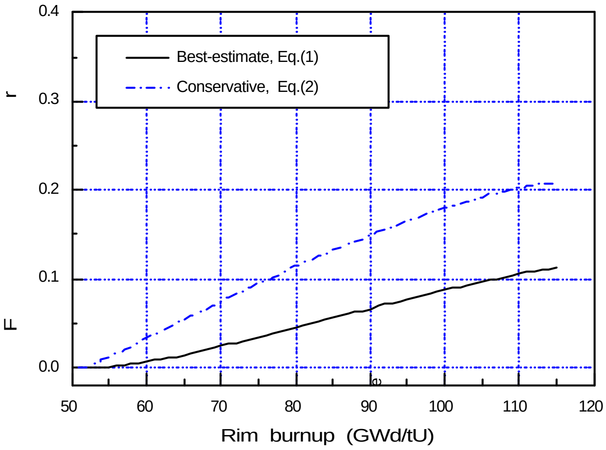


Fig.4. Fraction of Xe atoms at the grain boundary for the total Xe produced in a pellet.

4. Pore pressure in the rim region

Spino et al. [17] measured fuel porosity, volume pore density and pore size distributions as a function of the radial position for specimens with average burnup between 40.3 and 66.6 GWd/tU. Fig.5 shows the measured fuel porosity at several radial locations for three specimens. Measured porosity in Fig.5 was fitted as follows as a function of radial position and pellet average burnup: first, porosity is expressed by a function of $a_1 + \exp[-a_2 + a_3 \cdot (r/r_o)]$. Second, three constants in the function are determined for each set of measured values. Finally each corresponding constants are expressed as a function of pellet average burnup. For example, there are three a_1 for respective burnup of 40.3, 56.9 and 66.6 GWd/tU. Then these three constants for a_1 are expressed by one formula as a function of pellet average burnup.

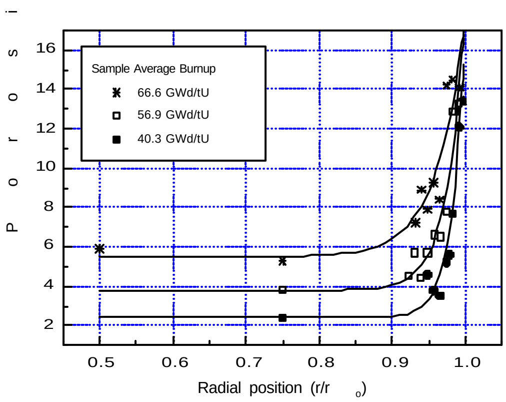


Fig.5. Measured porosity across fuel pellet [19].

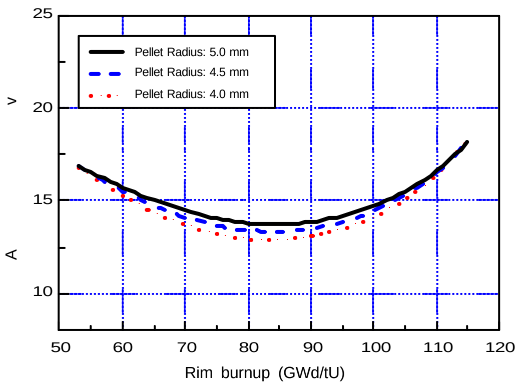


Fig.6. Calculated average porosity in the rim region.

Finally, this expression is integrated over the rim region and then divided by the rim volume, giving the average porosity as shown in Fig.6 for three different pellet radius.

The reason that average porosity in the rim decreases with burnup between 52 to 85 GWd/tU is that increase rate of porosity in the rim region with burnup is slower than the increase rate of rim width. After 85 GWd/tU, the trend is reversed and the average porosity increases with burnup.

Fig.7 shows the measured distribution of volume pore density in the rim region as a function of pore diameter. It is interesting to note that while the volume pore density increases with sample average burnup, the most probable pore diameter almost remains at the same value of about 1.2μm. A fitting function for the measured volume pore density was derived as a function of pellet average burnup and radial position using the same way as used for Fig.5 and this function was integrated over the rim region to get the total pores in the rim. The difference is that the fitting function here is of the type of $a_1 \exp[-0.5(\ln r_p - a_2)^2/a_3]$, where r_p is the pore diameter in μm. Fig.8 displays the total number of pores for three pellet radii, which increases linearly with rim burnup. However, since the calculated total pore for average burnup greater than 66.6 GWd/tU (corresponding rim burnup is 94 GWd/tU) is extrapolated one, total pore above rim burnup of 94 GWd/tU is uncertain.

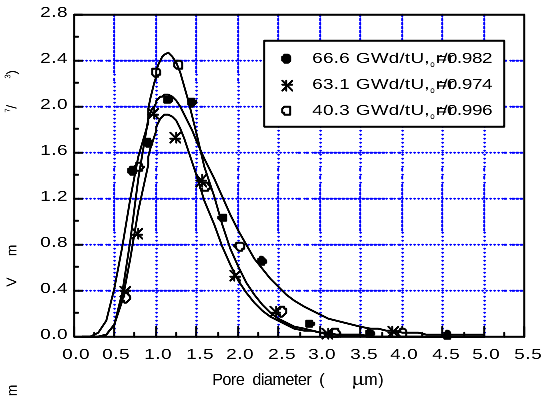


Fig.7. Measured volume pore density versus pore diameter [19].

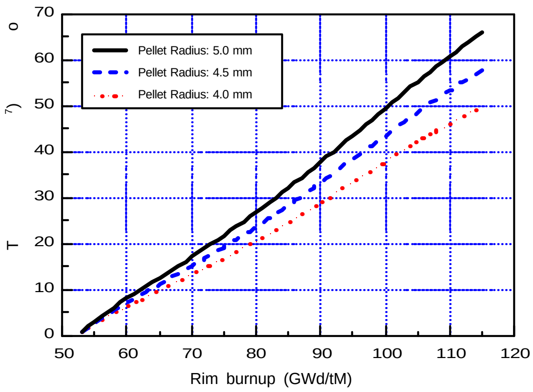


Fig.8. Calculated total pores in the rim region.

Using the calculated Xe concentration in the grain boundary Xe_g and the average porosity in the rim (Fig.6), the average number of Xe atoms per pore Xe_g^p is calculated as follows:

$$Xe_g^p = Total\ Xe\ in\ the\ rim \cdot \frac{V_p}{Rim\ volume \cdot Rim\ average\ porosity/100} \tag{6}$$

where the volume of a pore V_p is calculated using the assumption that the size of all the pores in the rim region is 1.2μm, which is the most probable size as shown in Fig.7. In Eq.(6) total Xe in the rim can be calculated from Xe_g (Eq.(4)) and the rim volume obtained from the rim width (Eq.(1) or Eq.(2)). In addition, since the rim average porosity is available from Fig.6, the average number of Xe atoms per pore Xe_g^p can be obtained by Eq.(6). Then the moles of Xe atoms per pore n_{Xe} is obtained by dividing Xe_g^p by the Avogadro's number.

Fig.9 shows the moles of Xe per pore in the grain boundary of the rim region. Both the Xe concentration and total pores increases with burnup as can be seen in Fig.3 and Fig.8. However, while the rim volume increases parabolically with rim burnup, the increase rate of total Xe in the rim reduces with burnup (see Fig.3). This is why the Xe moles per pore decrease for the rim burnup greater than about 100 GWd/tU. That is, it seems that for rim burnup beyond about 95 GWd/tU the Xe moles per pore decrease with burnup because the total pore used in this paper is a extrapolated one and this value may be larger than the real one. This argument can also be applied to explain the decrease in pore pressure for rim burnup beyond 105 GWd/tU as shown in Fig.10.

Using the value in Fig.9 and the equation of state for ideal gas, the pressure in the rim pore is calculated for a typical rim temperature of around 600°C under RIA conditions [2]. The assumption introduced to get the results of Fig.10 is that all the Xe atoms in the subgrain boundary of the rim region are evenly distributed among the pores with the same size of 1.2 μm. Therefore, depending on the pore size and pore number, there would be some variation in the pore pressure given in Fig.10. Gas pressure in the rim pore is calculated to be about 60 to 80 MPa for the rim burnup of 90 GWd/tU (corresponding pellet average burnup is about 63 GWd/tU) depending on whether the best-estimate or conservative rim width is used. In addition, fractional gas retention of 6 to 15% in the rim pores (Fig.4) and a pore density of $6 \times 10^7/mm^3$, which was obtained by dividing the total pores in the rim (Fig.8) by the rim volume, was used. On the other hand, Une et al. [18] showed that pore pressures at 800 K become 90-210 MPa when an equation of state for rare gases by Ronchi [19] is used for the rim pore size of 0.5 μm, pore density of $6 \times 10^7/mm^3$ and fractional gas retention of 30 to 50% in the rim pores for pellet average burnup of 80 GWd/tU. Considering the differences in parameters used, the present method yields reasonable pore pressure up to rim burnup of 100 GWd/tU.

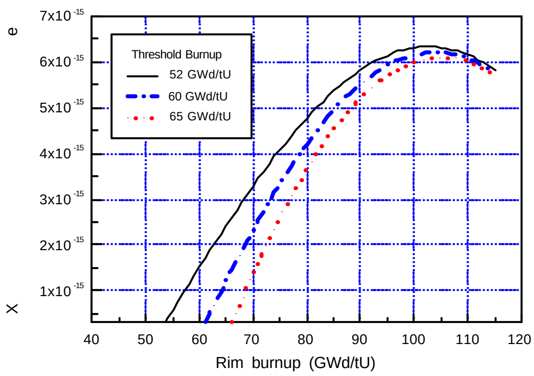


Fig.9. Calculated Xe in the grain boundary of the rim region.

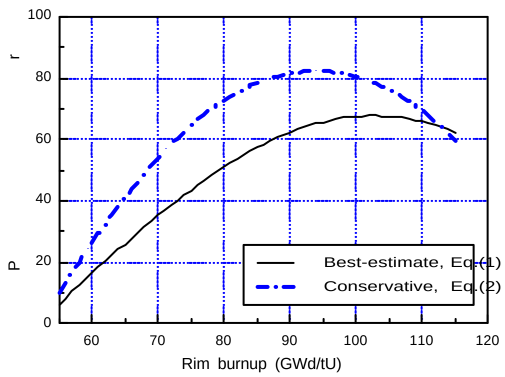


Fig.10. Calculated gas pressure in a rim pore for fuel temperature of 600 °C.

5. Conclusion

Using the measured rim width, average porosity and pore density in the rim region, an attempt has been made to estimate the pore pressure as a function of rim burnup, which is required for the modelling of fission gas release from the rim region under RIA conditions.

Estimated pore pressure in the rim is calculated to be about 60 to 80 MPa for the rim burnup of 90 GWd/tU depending on whether the best-estimate or conservative rim width is used. This result is consistent with other value given in a literature that pore pressures at 800 K become 90-210 MPa for the rim pore size of 0.5 μm, pore density of $6 \times 10^7/mm^3$ and fractional gas retention of 30-50% in the rim pores for pellet average burnup of 80 GWd/tU.

Considering the differences in parameters used, the present method yields reasonable pore pressure up to rim burnup of 100 GWd/tU.

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