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Effect of overpressurization on rim porosity in the high burnup UO_2 fuel

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Abstract

By introducing the concept of overpressurization of rim pores due to dislocation punching, the total pressure exerted on the rim pores is estimated. Then this concept is combined with the assumption that all the fission gases produced in the rim region are retained in the rim region to calculate the rim porosity. Rim porosities calculated in this way are compared with measured data, which produces reasonable agreement. Finally a correlation for the thermal conductivity of the rim region is obtained using the hypothesis that the rim region consists of pores and fully dense material of UO_2.

1. Introduction

To extend fuel burnup, it is important to predict the accurate performance for high burnup fuel. The fuel behaviors are significantly affected by fuel temperature distribution which is related to the change of fuel microstructure by irradiation. So, the understanding of fuel microstructure during irradiation is prerequisite to predict the thermal behaviors.

One of the distinguished microstructural changes is the formation of porous rim in the periphery in the high burnup fuel. This rim structure characterized by the subdivided grain and coarsened bubbles is very porous so that it acts as thermal barrier[1]. Furthermore, it is reported that the pressure of pores in the rim region exceeds the equilibrium pressure. This overpressurization results in the rim porosity that can estimate the temperature distribution by ideal gas law and thermal conductivity correlation.

In the present study, the rim is characterized such as rim burnup, width, and porosity in order to obtain the accurate thermal conductivity considering the degradation with burnup. This developed model will be incorporated into a computer subroutine capable of eventually interfacing with a general fuel performance code.

2. Analysis of rim overpressurization

2.1. Rim burnup and rim width

The rim effect by an increase of plutonium concentration and fissioning in a low temperature occurs in the periphery of pellet[2]. This region, which could be formed in high-burnup fuel, can be characterized by very high porosity, subdivision and recrystallization of UO_2 grain, and depletion of xenon from UO_2 matrix as measured by EPMA.

To characterize the rim region, rim burnups (pellet-edge burnups) are correlated from measured experimental results [3,4] by least square method. A rim-to-average burnup ratio of 1.43 is consistent with the value measured in RISO project, as well as with transport theory [5]. In this analysis the threshold pellet average burnup for the formation of rim region is assumed to be 40 MWD/kgU.

Another characteristic of rim region, rim width is estimated by using the measured data [6,7]. EPMA data for PWR are chosen for the analysis due to larger fluctuation of OM data even though the width could be determined by both OM and EPMA. Fig. 1 shows the fitted rim width as a function of rim burnup with experimental data. The least square method using linear relationship yields the following formula between rim width and rim burnup :

$$R_{Rim} = -285.3 + 4.987 \cdot BU_{Rim} \qquad (1)$$

where R_{Rim} is the rim width in μm and BU_{Rim} rim burnup. Fig. 1 also shows the Barner's [7] and Sierra's relation [8]. Compared with Barner's and Sierra's relation, linear relationship of Eq. (1) can be considered more reasonable for PWR because the rim width could be larger than 200 μm at very high burnup[9].

2.2. Porosity in the rim region

In LWR fuel the pellet rim starts to become very porous at the local burnup of about 60 MWD/kgU. It was proposed that the rim structure is caused by recrystallizatoin of the heavily radiation damaged UO_2 grains, by nucleation and growth of high angle subgrain boundary. Sweeping of fission gas bubbles should occur during subgrain growth, giving rise to the new micron sized porosity in the fuel by coalescence of bubbles at given preferential sites with the original matrix grains [9].

For the simplified analysis of porosity in the rim region it is assumed that 3 stable fission gas atoms (Xe and Kr) are generated from every 100 nuclear fission. Although it was reported that the xenon in the rim region was released to free volume of fuel, XRF and X-ray mapping results indicate that nearly all of the xenon produced in the rim region do not escape to free volume but are retained in the matrix. This also implies that the lost matrix xenon could be retained in the porosity of the rim region [3]. Furthermore, as yet no microcracks have been detected in the structure and there is no evidence to suggest that the pores are interconnected. Therefore it is a reasonable assumption that the fraction of released fission gas is negligible so that the produced fission gas is retained in the newly formed pores [10].

The mass of heavy metal in the rim region in a pellet, M_{Rim} is given by

$$M_{Rim} = V_{Rim} \cdot \rho \tag{2}$$

where the volume of rim region, V_{Rim} is obtained from the rim width and ρ is the density of a pellet. Energy produced in the rim region, E_{Rim} is given by

$$E_{Rim} = M_{Rim} \cdot BU_{Rim} \tag{3}$$

Using the assumption that energy released per fission is 200 MeV, the generated fission gas is calculated to be 1.437×10^{-3} mol per MWD. Then the total fission gas produced at the pellet rim is expressed by

$$n_{Rim} = 1.437 \times 10^{-3} \cdot E_{Rim} \tag{4}$$

where n_{Rim} is produced total fission gas in mol. To get the total fission gas volume which results in the rim porosity, the total fission gas volume under reactor operating condition is approximated by simple ideal gas law that requires the pressure on the rim pores.

The equilibrium pressure at the coarsened pores is given by

$$P_{eq} = \frac{2\gamma}{r_{pore}} + \sigma \tag{5}$$

where γ is surface tension (1 J/m for UO_2), σ hydrostatic pressure on the pore, and r_{pore} the mean pore radius with a typical diameter of 0.5 ~ 1.0 μm. The hydrostatic pressure is determined by the gap pressure between cladding and pellet. Furthermore, the pressure exerted on the pores in the rim region exceeds the equilibrium pressure. Due to the surface tension forces implied by dislocation punching[2], fission product gas pressure required to form dislocation can be obtained by,

$$P_{ex} = \frac{\mu b}{r_{pore}} \tag{6}$$

where μ is the shear modulus and b is burgers vector (0.39 nm for UO_2). The shear modulus which is dependent on the temperature and porosity can be calculated as follows

$$\mu = \frac{\left(22.43 \times 10^{4} - 31.19 \cdot T_{Rim}\right)\cdot\left(1 - 2.6 \cdot Por\right)10^{6}}{1+\nu} \tag{7}$$

where ν is Poisson ratio (0.32 for UO_2) and Por is the pore volume fraction of a pellet.

Finally, the pore volume in the rim region can be calculated by ideal gas law

$$V_{Rim} = \frac{n_{Rim} \cdot R \cdot T_{Rim}}{P_{Rim}} \tag{8}$$

where R is gas constant and P_{Rim} is obtained from summation of equilibrium and excess pressure.

The mass of heavy metal in the rim region in a pellet, M_{Rim} is given by

$$M_{Rim} = V_{Rim} \cdot \rho \tag{2}$$

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$$V_{Rim} = \frac{n_{Rim} \cdot R \cdot T_{Rim}}{P_{Rim}} \tag{8}$$

where R is gas constant and P_{Rim} is obtained from summation of equilibrium and excess pressure.

Using the porosity of the rim region by eq. (8), the porosity under typical PWR operating condition is correlated with rim burnup as shown in Fig. 2. In the case of pore radius of 1μm comparison shows that the developed model for rim porosity results in the higher prediction than Sierra correlation.

3. Thermal conductivity degradation

Thermal conductivity during irradiation is degraded by the introduction of defects to the previously almost perfect UO_2 lattice. Furthermore, thermal conductivity steeply decreases across the rim with burnup due to the porous microstructure and higher local burnup. Consequently the thermal conductivity of irradiated UO_2 fuel is dependent on porosity, stoichiometry and burnup as well as temperature [11]. Models for three thermal conductivities of MATPRO [12], SIMFUEL [11], and HALDEN [13] are chosen for the present analysis. The obtained thermal conductivity is given in Fig. 3 as a function of burnup. It is noted that the thermal conductivity of MATPRO is independent of burnup, while the others decrease with burnup.

In the rim region, the thermal conductivity degradation is obtained under the assumption that the rim region consists of pores and fully dense material. The dependence of thermal conductivity on porosity is given as follows [14]:

$$k_{Rim} = k_0 \cdot \left\{ 1 - a \cdot Por_{Rim}^{\frac{2}{3}} \cdot \left[1 - \frac{1}{1 + \frac{1}{a} \cdot Por_{Rim}^{\frac{1}{3}} \cdot \left(\frac{k_0}{k_p} - 1 \right)} \right] \right\} \tag{9}$$

where k_{Rim} = thermal conductivity of the porous rim (W/m·K)

k_o = thermal conductivity of the fully dense material (W/m·K)

= $k \big/ \left[1 - \left(2.58 - 0.58 \times 10^{-3} T \right) P_j \right]$ [15]

k_p = thermal conductivity of the pore

Por_{Rim} = porosity (volume fraction of the porous phase)

a = anisotropy factor (a=1 means isotropic pore distribution)

For the analysis of the thermal conductivity of pores in the rim region, it is assumed that all the xenon gas produced in the rim region are retained in the porosity. Then the thermal conductivity of porosity in Eq. (9) can be replaced by thermal conductivity of xenon [14] expressed by

$$k_{Xe} = 0.72 \times 10^{-4} \cdot T^{0.79} \qquad (10)$$

Under the assumption that all the pores are isotropically distributed, thermal conductivity of the rim is shown in Fig. 4 as a function of burnup at 500°C. The thermal conductivity decreases abruptly around the pellet average burnup of 40 MWD/kgU. Fig. 4 shows that the thermal conductivity calculated by the HALDEN correlation is the lowest among three both in the rim and in the pellet interior.

4. Conclusion

A correlation has been developed that can estimate the porosity of rim region as a function of rim burnup under the assumptions that all the fission gases produced in the rim region are retained in the rim porosity and threshold pellet average burnup required for the formation of rim region is 40 MWD/kgU. Rim width is correlated to rim burnup using measured data. And a model for the rim porosity was proposed based on the overpressurization of rim pore. Thermal conductivity was estimated in highly porous pellet rim.

References

1. J.O.Barner, et al., *International Topical Meeting on LWR Fuel Performance*, Avignon, France, April 21-24, 1991.
2. Hj. Matzke, *Journal of Nuclear Materials*, **189**,141 (1992).
3. M.E.Cunningham, et al., *Journal of Nuclear Materials*, **188**, 19 (1992).
4. S.R.Pati, et al., *International Topical Meeting on LWR Fuel Performance*, Williamsburg, Virginia, April 17-20, 1982.
5. N.Kjaer-Pederson, Proceedings of a technical committee meeting, Ontario, Canada, April 28, p.111, 1992.
6. J.O.Barner, et al., *Nuclear Technology*, **102**, 210 (1993).
7. R.Manzel and R.Eberle, *International Topical Meeting on LWR Fuel Performance*, Avignon, France, April 21-24, 1991.
8. M.R. Billiaux, et al, *International Topical Meeting on LWR Fuel Performance*, March 2-6, 1997
9. K.Nogita and K.Une, *Journal of Nuclear Materials*, **226**, 302 (1995).
10. K.Lassmann, et al., *Journal of Nuclear Materials*, **226**, 1 (1995).
11. P.G Lucuta, Hj. Hatzke, R.A. Verrall, *Journal of Nuclear Materials*, **217**, 279 (1994).
12. MATPRO-VERSION 11 (Revision 2) NUREG/CR-0497, EG&G Idaho, August, 1981.

13. W. Wiesenack, *International Topical Meeting on LWR Fuel Performance*, March 2-6, 1997

14. H. Kampf and G. Karsten, *Nuclear Application and Technology*, **9**, 288 (1970).

15. D.G.Martin, *Journal of Nuclear Materials*, **110**, 73 (1982).

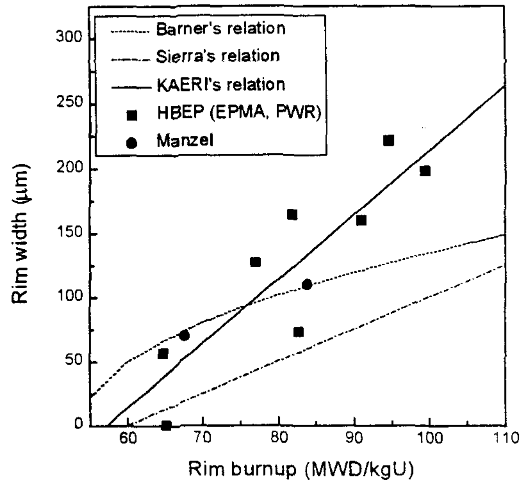

Fig. 1. Rim width with rim burnup.

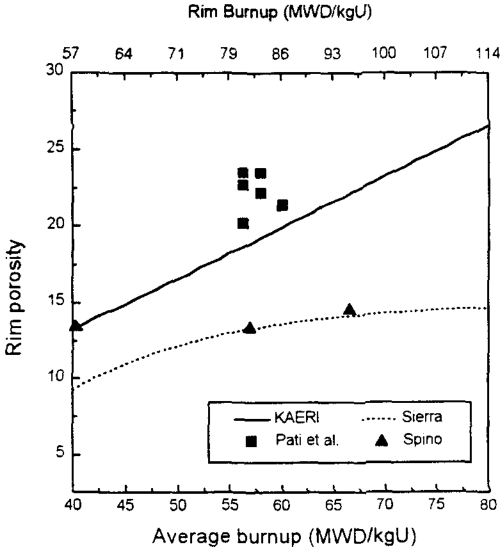

Fig. 2. Rim porosity derived from rim overpressurization

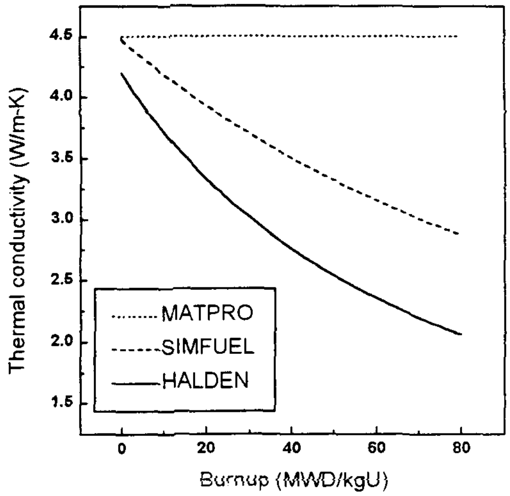

Fig. 3. Thermal conductivity as a function of burnup in UO_2 fuel.

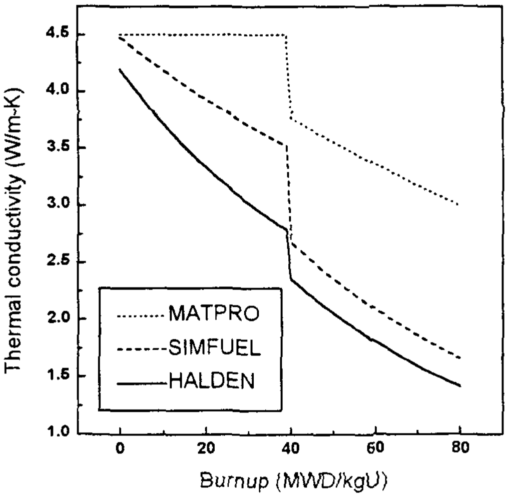

Fig. 4. Thermal conductivity as a function of burnup in the rim region.