Proceedings of the Korean Nuclear Society Spring Meeting
Seoul, Korea, May 1998

Threshold burnup for recrystallization and model for rim porosity in the high burnup UO_2 fuel

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Abstract

Applicability of the threshold burnup for rim formation was investigated as a function of temperature by Rest's model. The threshold burnup was the lowest in the intermediate temperature region, while on the other temperature regions the threshold burnup is higher. The rim porosity was predicted by the van der Waals equation based of the rim pore radius of 0.75μm and the overpressurization model on rim pores. The calculated centerline temperature is in good agreement with the measured temperature. However, more efforts seem to be necessary for the mechanistic model of the rim effect including rim growth with the fuel burnup.

1. Introduction

To obtain high burnup of UO_2 fuel, it is essential to predict the accurate performance of high burnup fuel such as thermal and fission gas behaviors. These fuel behaviors are significantly affected by fuel temperature distribution, which is related to the change of fuel microstructure due to the irradiation. One of the changes of fuel microstructure is the formation of porous rim in the periphery of the high burnup fuel. The rim structure is characterized by the very high porosity, the subdivided grain and coarsened bubbles. The grain subdivision causes 10^4 to 10^5 new small subgrain [1].

Especially, Spino [2] has reported that the porosity and the pore density in the rim region increase with burnup, while on the other hand the mean pore size remains in the range of 1.2 to 1.6 μm. Understanding the rim structure is prerequisite to obtain the thermal conductivity of the porous rim, which is the essential physical parameter for the thermal behavior of high burnup UO_2 fuel.

We have recently proposed the thermal conductivity model to predict an accurate temperature distribution considering the thermal conductivity degradation with burnup and the rim effect of thermal barrier [3].

In this paper we have modified the model for rim porosity based on the experimental results [2]. And then we applied the rim porosity model to one of the HALDEN results for validation.

On the other hand, Rest [4] has developed a mechanistic description on the recrystallization in the rim region. So one of the purpose of this paper is to show the feasibility for the application on the threshold burnup for rim formation.

2. Threshold burnup for rim formation

An increase of the plutonium concentration and fissioning due to the resonance absorption of epithermal neutrons by ^{238}U results in the formation of very porous rim region in the periphery of pellet [2].

Threshold burnup for rim formation is assumed about the pellet average burnup of 40MWD/kgU even though the experimental results [5] show the variation (scattering) of the threshold burnup. Generally, the higher threshold burnup can be expected at the higher power density and the higher temperature.

Analytical threshold burnup can be expressed by Rest's model [4] for the formation of rim region instead of assuming the pellet average burnup of 40 MWD/kgU.

$$THBU(T)=\frac{CF\cdot E_{sf}\left[9\cdot\Omega\cdot\dot{f}+7\cdot a\cdot\sqrt{\pi\cdot B\cdot r_{iv}\cdot\xi\cdot\nu_v\cdot\Omega\cdot\dot{f}}\cdot EXP\left(\frac{-\varepsilon_{vI}-\frac{\varepsilon_v}{2}}{k\cdot T}\right)\right]}{168\cdot\pi\cdot r_{sm}\cdot k\cdot T\cdot c_I\cdot\left(\xi_{vi}\cdot a^2\cdot\nu_v\cdot EXP\left(-\frac{\varepsilon_{vI}}{k\cdot T}\right)+X_2\cdot\dot{f}\right)}$$

where *CF* is the conversion factor from fission density (fission/m^3) to MWD/kgU. Table 1 lists the other parameters for the definition and values.

Fig. 1 shows the threshold burnup for recrystallization as a function of temperature. At the lower temperature and the other higher temperature, the threshold burnup is very high compared with the intermediate temperature region. The high burnup at the low temperature results from the higher activation energy for the grain subdivision, while at the high temperature the fast fission gas release by thermal process retarded the formation of tangled dislocation structure.

This relationship will be implemented into KAERI's fuel performance code, *COSMOS[6]*.

3. Prediction for rim porosity

The rim porosity is prerequisite to obtain the thermal conductivity of the porous rim, which is the essential physical parameter for the thermal behavior of high burnup UO_2 fuel. Recently Spino [2] has reported that the rim porosity and the pore density in the rim region increase with burnup, while on the other hand the mean pore size remains in the range of 1.2 to 1.6 μm. In order to estimate the rim porosity, van der Waals equation can be applied after obtaining the pressure on the pores.

Van der Waals equation of state is most commonly used to describe the thermodynamic state of fission xenon in the bubble. It can be written as [7]

$$p(\frac{1}{\rho_g}-B)=kT$$

where p is the pressure of a gas of molecular density ρ_g at temperature T. The constant

B can be regarded as expressing the volume occupied by the atoms proper, or more precisely, it is a reflection of the short-range repulsive forces in the interatomic between xenon atoms. In most analysis, B is taken to be a constant equal to 85 A^3/atom.

The pressure p consists of the hydrostatic, surface tension, and dislocation punching terms as follow [1],

$$p=\frac{2\gamma}{R}+\frac{\mu b}{R}+\sigma$$

The molecular density, ρ_g can yields the number of gas atoms contained in a bubble of radius $\overline{R}$

$$m=\frac{4\pi\overline{R}^3}{3}\rho_g=\frac{4\pi\overline{R}^3}{3}\left[\frac{1}{B+\dfrac{kT}{\dfrac{2\gamma}{\overline{R}}+\dfrac{\mu b}{\overline{R}}+\sigma}}\right]$$

Assuming 30 stable fission gas atoms are generated from every 100 nuclear fission, the overall balance for the fission gas can be written as

$$n_{RIM}N_A=mN_{POR}V_{RIM}$$

where N_A is Avogadro number, N_{POR} the rim pore density, and V_{RIM} is the rim volume. Using the assumption that energy released per fission is 200 MeV, the generated fission gas, n_{RIM} is calculated to be 1.343×10^{-3} mol per MWD.

Then, the rim pore density is obtained by

$$N_{POR}=\frac{n_{RIM}}{V_{RIM}}.\frac{N_A}{m}$$

Finally, the porosity at the rim can be evaluated by

$$\overline{P}=\frac{n_{Rim}N_A}{V_{Rim}}\left(B+\frac{kT_{RIM}}{\dfrac{2\gamma}{\overline{R}}+\dfrac{\mu b}{\overline{R}}+\sigma}\right)$$

The rim temperature, T_{RIM} is assumed to be constant at 500°C in this analysis. The pore radius is taken to be a constant equal to 0.75μm by experimental measurement [1,2].

Fig. 2 shows the rim porosity as a function of pellet average burnup with the measured data [2,8]. The rim porosity increases with the pellet average burnup, which is caused by the increase of the total amount of fission gas in the rim pores.

4. Validation by experimental results, IFA526

The thermal conductivity decreases as a function of the rim porosity. In the analysis fuel rod temperature was calculated by MATPRO, SIMFUEL and HALDEN model [9]. The HALDEN model is expressed by

$$k_{95} = \frac{1}{0.1148 + 0.0035 \cdot BU + 2.475 \times 10^{-4} \cdot (1 - 0.00333 \cdot BU) \cdot T} + 0.0132 \cdot e^{0.00188 \cdot T} \quad (4)$$

where T in °C, Burnup BU in MWD/kgUO$_2$, k_{95} in W/m-K. And burnup limit is 75MWD/kgUO$_2$. This thermal conductivity degradation causes the fuel temperature increase during fuel performance analysis.

Fig. 3 presents a comparison with HALDEN data normalized at 25 kW/m from the measured data in a fuel rod with small diametrical gap operating at temperatures below 1000 °C [9]. The calculated surface temperature of the pellet is also shown as a function of burnup. The measured centerline temperature increases continuously with burnup mainly due to the degradation of thermal conductivity. Since it was assumed that the threshold pellet average burnup for the formation of the rim region is 40 MWD/kgU, the slight temperature increase appears in the present calculation at burnups higher than 40MWD/kgU. When the HALDEN model is used, the calculated temperature, without considering rim effect, increases linearly, while the calculated temperature with rim effect considered shows a slight increase after the threshold burnup. The other two correlations of MATPRO and SIMFUEL underpredict fuel centerline temperature by not considering the burnup degradation and fission product effect on the thermal conductivity, respectively.

Although correlating pellet average burnup with rim burnup shows very wide statistical scattering, the thermal conductivity model developed in this paper seems to well predict the temperature variations even in the burnup higher than 40 MWD/kgU.

5. Conclusion

The threshold burnup for rim formation was obtained by Rest's model. The threshold burnup was the lowest in the intermediate temperature region as expected. The rim porosity was predicted using the van der Waals equation based on the rim pore radius of 0.75μm and the overpressurization model on rim pores. The calculated centerline temperature is good agreement with the measured temperature. However, it needs more efforts to develop the mechanistic rim model including rim growth with the fuel burnup.

Reference

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Table 1. Values of various properties used in the calculation of rim threshold burnup [4]

Property	Definition	Value
E_{sf}	$E_{sf} = E_s + E_f\,[eV]$	0.62
ε_{vI}	Migration energy for a vacancy – solute pair $[eV]$	2.8
ε_v	Migration energy for a vacancy $[eV]$	2.4
k	Boltzmann constant $[eV/K]$	8.617×10^{-5}
B	Conversion factor : $\dot{f} = Bk\ [1/m^3\cdot s]$	6×10^{23}
c_I	Solute concentration	1×10^{-8}
r_{iv}	Radius of recombination volume $[m]$	2×10^{-10}
r_{sm}	Annihilation radius of a nucleus/vacancy – solute pair $[m]$	3×10^{-10}
Ω	Atomic volume $[m^3]$	39.3×10^{-30}
$\dot{f}$	Fission rate per unit volume $[fission/m^3\cdot s]$	10^{20}
X_2	$X_2 = D_{red}/\dot{f}\ [m^5]$	2×10^{-39}
a	Atomic radius $[m]=\sqrt[3]{\Omega}$	
ξ	Pre-exponential factor for deviation from diffusion for v	0.1
ξ_{vi}	Pre-exponential factor for deviation from diffusion for v-i	0.01
ν_v	Vibration frequency factor for vacancy $[s^{-1}]$	5×10^{13}

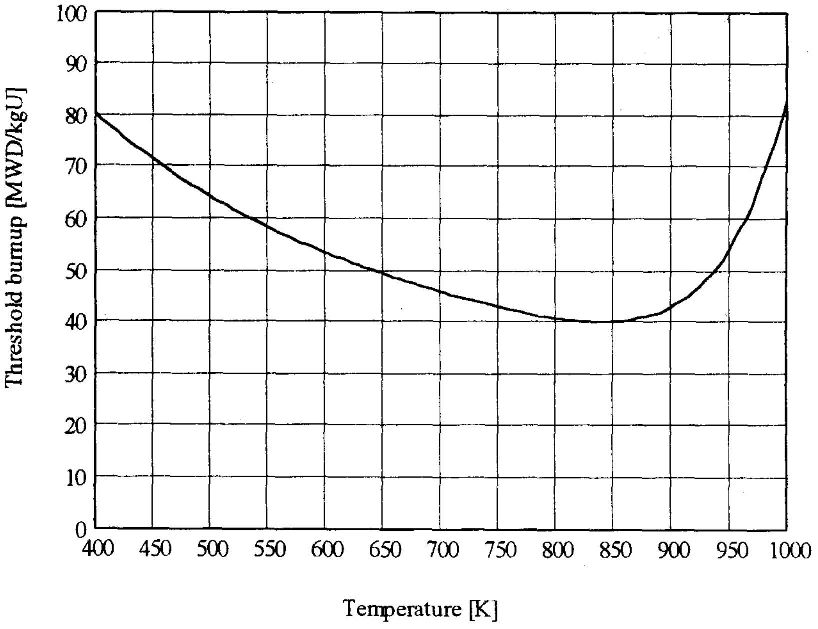

Fig. 1 Threshold burnup as a function of temperature.

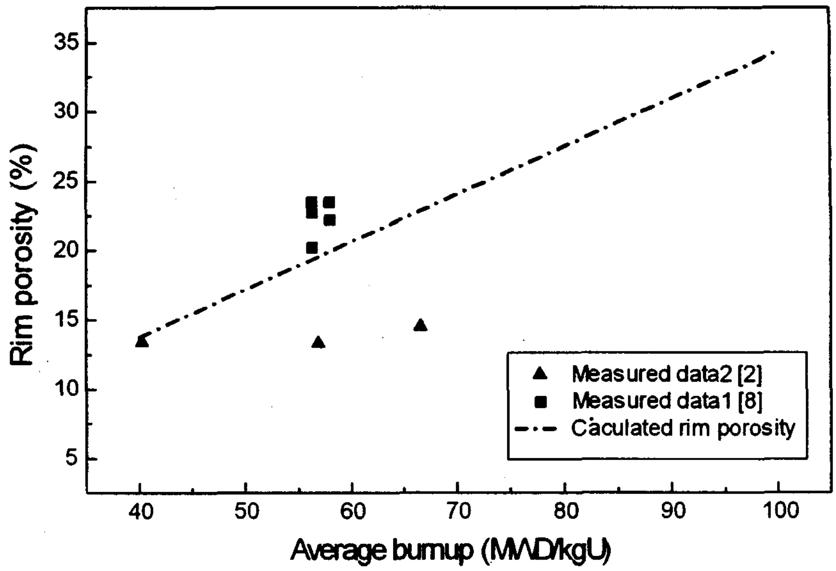

Fig. 2 Comparison of calculated rim porosity with the measured data

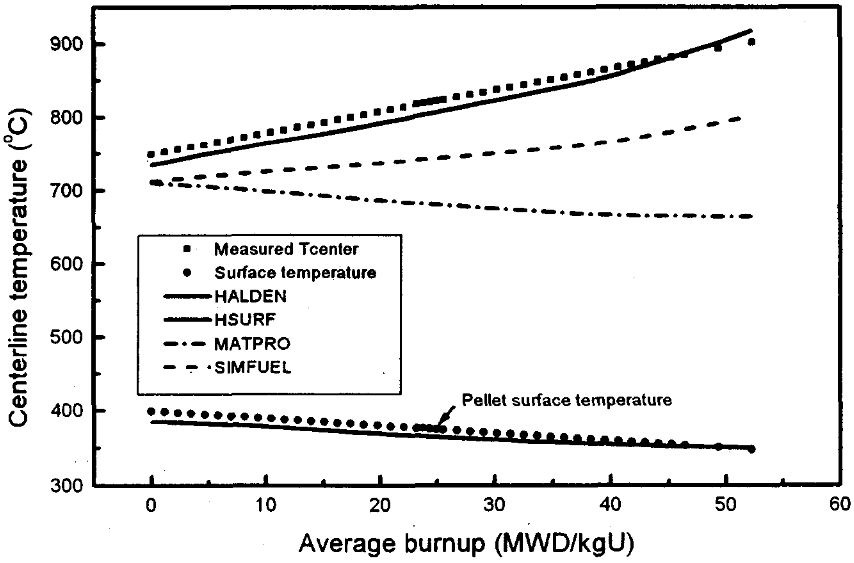

Fig. 3 Comparison of measured and calculated centerline temperature of UO_2 fuel at linear heat rating of 25 kW/m.