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Degradation of Irradiated UO_2 Fuel Thermal Conductivity Calculated by FRAPCON Model Due to Porosity Evolution at High Burn-Up

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***Abstract*—**The evolution of volume porosity previously obtained by using the existing low temperature high burn-up gaseous swelling model with progressive recrystallization for UO_2 fuel is utilized to study the degradation of irradiated UO_2 thermal conductivity calculated by the FRAPCON model of thermal conductivity. A porosity correction factor is developed based on the assumption that the fuel morphology is a three-phase type, consisting of the as-fabricated pores and pores due to intergranular bubbles whitin UO_2 matrix and solid fission products. The predicted thermal conductivity demonstrates an additional degradation of 27% due to porosity formation at burn-up levels around 120 MWd/kgU which would cause an increase in the fuel temperature accordingly. Results of the calculations are compared with available data.

***Keywords*—**Irradiation-induced recrystallization, matrix swelling, porosity evolution, UO_2 thermal conductivity.

I. Introduction

THE porosity impact on thermal conductivity, among other factors, is one of the most difficult properties to assess. In fact, the oxide fuel is generally fabricated by sintering pressed powdered UO_2 at high temperature. By controlling the sintering conditions, material of any desired density, usually around 90% of the maximum possible or theoretical density of the solid, can be produced. As a result, as-fabricated porosity is already present in unirradiated fuel. The fission gas atoms resulted from the fission process are known to be insoluble in the fuel matrix and to precipitate into intragranular and intergranular bubbles, which contribute to fuel swelling and porosity generation. With increasing burn-up under low-temperature condition, UO_2 fuel is known to undergo irradiation-induced recrystallization process by which more grain boundaries are formed to accommodate fission gas atoms. Recrystallization initiates from the boundary of the grain and proceeds toward its center until the grain is totally consumed. Large grains turn into small fine grains so as to absorb gas atoms in the grain boundary effectively, thus large intergranular bubbles will be produced and make a major contribution to the gas bubble swelling and corresponding porosity generation [1]-[5].

Very recently, using an approach based on the work of Spino at al. [6] as well as the Rest model [2], [4], [5] for UO_2 matrix swelling with progressive recrystallization, the present authors [7] quantified the influence of porosity evolution on the thermal conductivity of UO_2 matrix determined by the Lucuta et al. [8] model of thermal conductivity having the Maxwell-Eucken correlation as porosity factor.

The aim of this paper is to use the same approach as followed in [7] to estimate the degradation of irradiated UO_2 fuel thermal conductivity due to porosity evolution at high burn-up. But in the present work the FRAPCON model [9] for UO_2 thermal conductivity is selected and combined with a proposed porosity factor by this paper that developed through the analytical approach adopted in the DART code [10].

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II. Models and Methods

A. FRAPCON Model of Thermal Conductivity

The fuel thermal conductivity model as implemented in the FRAPCON-3 code comes from works of Ohira et.al. [11] that is based on the thermal conductivity measurements of high burn-up UO_2 Pellets related to the Nuclear Fuels Industries (NFI) model. This model applied to UO_2 thermal conductivity at 95% of theoretical density (TD) is expressed as (1) [9], [11].

$$\begin{cases} k_{95}\left(\frac{W}{mK}\right) = \frac{1}{A+BT+f(Bu)+(1-0.9\exp(-0.04Bu))g(Bu)h(T)} \\ \qquad + \frac{E}{T^2}\exp\left(\frac{-F}{T}\right) \\ A = 4.52\times10^{-2} \quad (mK/W) \\ B = 2.46\times10^{-4} \quad (mK/W/K) \\ E = 3.5\times10^{9} \quad (WK/m) \\ F = 16361 \quad (K) \\ f(Bu) = 1.87\times10^{-3}Bu \quad \left(\frac{mK}{W}\right) \\ g(Bu) = 0.039Bu^{0.28} \quad \left(\frac{mK}{W}\right) \\ h(T) = \frac{1}{1+396\exp\left(-\frac{Q}{T}\right)} \\ Q = 6380 \quad (K) \end{cases} \tag{1}$$

where $f(Bu)$ is the effect of fission products in crystal matrix (solution), $g(Bu)$ is the effect of irradiation defects, $h(T)$ is temperature dependence of annealing on irradiation defects, Q is temperature dependence parameter, Bu is burn-up in MWd/kgU and T is the temperature in Kelvin.

B. Porosity Factor

To include the effect of total porosity P on the thermal conductivity of irradiated UO_2 described by the FRAPCON

model, we assume a fuel with morphology of three-phase type. The phases consist of the as-fabricated pores P_v in (2) with no contribution to the matrix swelling and large pores of intergranular bubbles P_S in (3) with a contribution to matrix swelling that are derived in the author's previous paper [7] and dispersed uniformly in the fully dense material composed of UO_2 matrix and solid fission products.

$$P_v = 1 - \left(1 + \Delta V^{matrix}/V_0^{matrix}\right)(\rho/\rho_{th}) \quad (2)$$

where ρ is the bulk fuel density and ρ_{th} is the theoretical density of UO_2, and $\Delta V^{matrix}/V_0^{matrix}$ presents total fractional fuel matrix swelling caused by both solid ($\Delta V^{solid}/V_0^{matrix}$) and gaseous ($\Delta V^{gaseous}/V_0^{matrix}$) fission products [7].

$$P_s = \left(\Delta V^{gas\ bubble}/V_0^{matrix}\right)/\left(1 + \Delta V^{gas\ bubble}/V_0^{matrix}\right) \quad (3)$$

where V_0^{matrix} denotes the initial matrix volume, $\Delta V^{gas\ bubble}$ is the increase of matrix volume due to bubble formation and $\Delta V^{gas\ bubble}/V_0^{matrix}$ represents the fractional fuel matrix swelling due to fission gas bubbles [7].

To use DART code thermal conductivity model for porous material [10], can be proposed a model for porosity factor. The model depends to fuel initial thermal conductivity, evolution of as-fabricated pores, P_v, swelling porosity, P_S, and thermal conductivity of gas within swelling porosities. The model includes a solid material surrounding a spherical pore and forms a cube unit cell of fuel material. According the model when solid material and spherical pore is considered fuel and pore P_v, respectively then effective cell thermal conductivity is given by (4).

$$\frac{k_e^V}{k_0} = 1 - \left[\pi\left(\frac{3}{4\pi}P_v\right)^{\frac{2}{3}}\right]\left[1 - \frac{k_g}{2k_0\left(\frac{3}{4\pi}P_v\right)^{\frac{1}{3}}}\right] \quad (4)$$

where k_e^V is effective thermal conductivity of fuel material contain of pore P_v in W/mK, k_0 is thermal conductivity of fully dense UO_2 material and k_g is thermal conductivity of gas within pore. For the pore P_v is considered as a cavity without any gas atom, so $k_g = 0$.

To include the effect of swelling porosity P_S on fuel thermal conductivity, once again the unit cell is applied as solid material is fuel contain of pore P_v and spherical pore is swelling porosity P_S. So can be obtained a porosity factor (κ_P) to form of (6).

$$\frac{k_{eff}}{k_e^V} = 1 - \left[\pi\left(\frac{3}{4\pi}P_s\right)^{\frac{2}{3}}\right]\left[1 - \frac{k_g}{2k_e^V\left(\frac{3}{4\pi}P_s\right)^{\frac{1}{3}}}\right] \quad (5)$$

$$\kappa_P = \frac{k_{eff}}{k_0} = \left[1 - \pi\left(\frac{3}{4\pi}P_v\right)^{\frac{2}{3}}\right]\left\{1 - \left[\pi\left(\frac{3}{4\pi}P_s\right)^{\frac{2}{3}}\right]\left[1 - \frac{k_g}{2k_e^V\left(\frac{3}{4\pi}P_s\right)^{\frac{1}{3}}}\right]\right\} \quad (6)$$

where k_{eff} is effective thermal conductivity of porous fuel material including P_v and P_S, and k_g is thermal conductivity of the Xe and Kr gas given by (7)-(11) [12]:

$$k_{Xe} = (4.0288 \times 10^{-5})T^{0.872}(W/mK) \quad (7)$$

$$k_{Kr} = (4.726 \times 10^{-5})T^{0.923}(W/mK) \quad (8)$$

$$k_g = \sum_{i=1}^{n} \frac{k_i}{1 + \sum_{j=1, j\neq i}^{n} \Psi_{ij}\frac{X_j}{X_i}} \quad (9)$$

$$\Psi_{ij} = \phi_{ij}\left[1 + 2.41\frac{(M_i - M_j)(M_i - 0.142M_j)}{(M_i - M_j)^2}\right] \quad (10)$$

$$\phi_{ij} = \frac{\left[1 + \left(\frac{k_j}{k_j}\right)^{1/2}\left(\frac{M_i}{M_j}\right)^{1/4}\right]^2}{2^{3/2}\left(1 + \frac{M_i}{M_j}\right)^{1/2}} \quad (11)$$

where k_i is the thermal conductivity of i gas, X_i is the mole fraction of the i gas $\left(\frac{X_{Xe}}{X_{Kr}} \sim 10.8\right)$ [13], M_i is the molecular weight of i gas $(M_{Xe} = 131.3, M_{Kr} = 83.80)$ [14], and n is number of components in the gas mixture (n=2).

The proposed model for porosity factor is adjusted for as-fabricated fuel density d using the porosity factor of (6) with no bubble porosity ($P_s = 0$), as (12) and (13):

$$k_0 = k_{95} * \left[1 - \pi\left(\frac{3}{4\pi}0.05\right)^{\frac{2}{3}}\right]^{-1} \quad (12)$$

$$k_d = k_0 * \left[1 - \pi\left(\frac{3}{4\pi}P_v\right)^{\frac{2}{3}}\right], \quad (13)$$

where k_0 and k_{95} are the thermal conductivity at 100% and 95% theoretical density, respectively, k_d is the thermal conductivity at the *d* percentage of theoretical density. The factor of $\left[1 - \pi\left(\frac{3}{4\pi}0.05\right)^{\frac{2}{3}}\right]^{-1}$ adjusts the thermal conductivity from 95% to 100% theoretical density.

Note that in this paper, the impact of intragranular bubbles on phonon scattering is neglected before grain recrystallization. Because of the paper scope is determination of thermal conductivity in rim region where no exist nanometer intragranular bubbles (that have size comparable with phonon mean free path) but instead exist large bubbles from scale of micrometer.

III. Results and Discussion

A. Porosity Evolution

The evolution of the swelling porosity P_s, pore contribution P_v, and total porosity P as a function of burn-up previously calculated in [7] are shown in Fig. 1. It is found that at the high burn-ups due to the increase of gas bubble swelling contributed by the intergranular bubbles gathered in the recrystallized grain boundary, the swelling porosity increases,

leading to an increase in the total porosity while the porosity due to as-fabricated pores goes down with the onset of recrystallization at high burn-ups. As seen in Fig. 1, the calculated total porosity values follow the trend of the porosity data [15] as a function of burn-up.

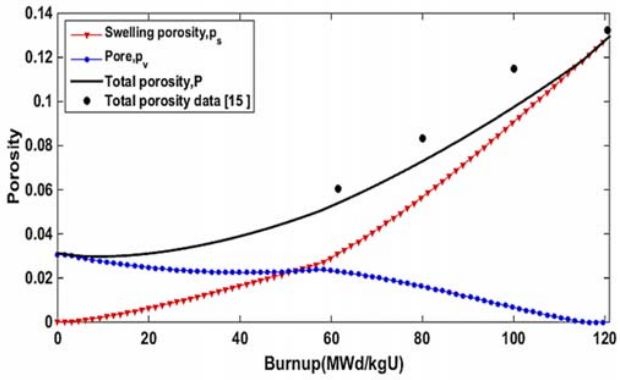

Fig. 1 Pore P_v, swelling porosity P_s, and total porosity P as a function of burn-up at 800K [7] compared with experimental data [15]

B. Porosity Factor

The porosity factor proposed by this paper is compared with some common porosity factors [16], [17] in Fig. 2. As can be observed, this work porosity factor is more conservative than others that can cause further reduction in thermal conductivity. So the effective thermal conductivity predicted for fuel as can be observed in section C, gets close to experimental data.

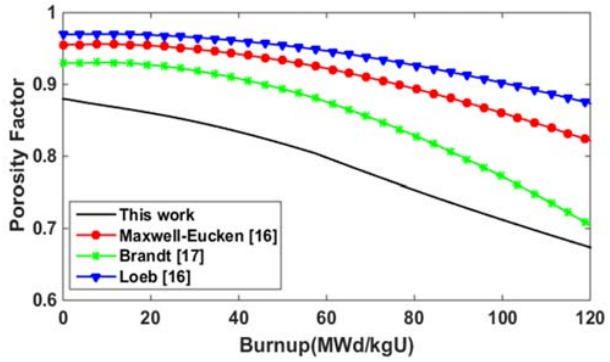

Fig. 2 Comparing the proposed porosity factor in (6) with others at 800K

C. Thermal Conductivity

Fig. 3 compares the evolution of calculated UO_2 thermal conductivity based on the FRAPCON code model as a function of local burn-up for two cases, including a constant volume porosity 3% and an evolving one as given above. It can be seen that taking into account the evolution of porosity with burn-up leads to a decrease in the thermal conductivity of about 27% at a local burn-up of 120MWd/kgU at 490 K. Fig. 3 also shows that the model satisfactorily predicts the Walker et.al experimental data [18] for 92.2, 99.8 and 120 MWd/kgU burn-ups obtained at the temperature of 490 K. The experimental data were normalized from the 95% theoretical density to 97% using (12) and (13), assuming spherical pore shape.

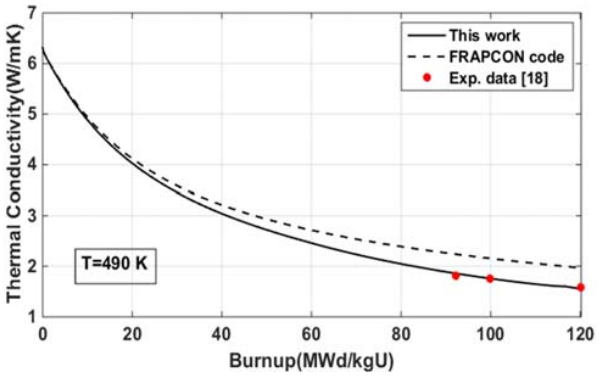

Fig. 3 Calculated UO_2 fuel thermal conductivity based on FRAPCON code model vs. burn-up at 490K with porosity evolution included in comparison with the case with constant porosity and experimental data [18]

Fig. 4 shows the UO_2 thermal conductivity calculated by the FRAPCON code model as a function of the temperature for different burn-ups while the computed porosity evolution in the porosity factor is taken into account. The thermal conductivity degradation due to high burn-up and temperature is seen in this figure.

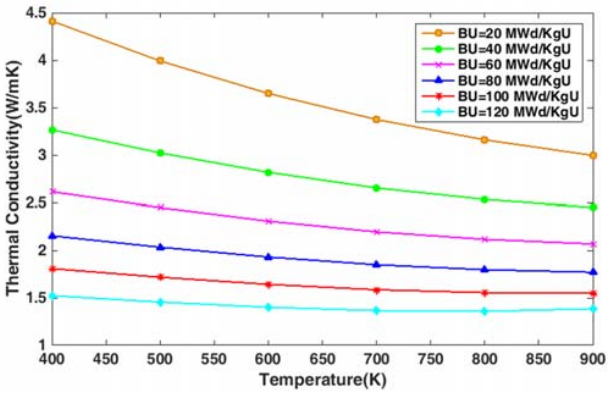

Fig. 4 Calculated UO_2 Thermal conductivity as a function of temperature for different burn-ups

IV. Conclusion

In this paper the volume porosity evolution of irradiated UO_2 fuel with burn-up previously estimated using the Rest's gaseous swelling model with progressive recrystallization [7] was used to study the degradation of irradiated UO_2 thermal conductivity with burn-up. To do that, the empirical thermal conductivity model adopted by the nuclear fuel performance code, FRAPCON-3 was selected. Then we combined it with a porosity correction factor developed on the basis of three-phase type morphology of irradiated fuel. The phases are the as-fabricated pores not contributing to the matrix swelling and large pores of intergranular bubbles with a contribution to matrix swelling dispersed uniformly in the fully dense

material. A decrease in the UO_2 thermal conductivity up to 27% at local burn-up levels around 120MWd/kgU compared to the case with as-fabricated porosity during irradiation time, was shown. A very good agreement was seen from the comparison of predicted thermal conductivity with the available experimental data at high burn-ups.

The present results show the importance of taking into account the effect of porosity evolution on UO_2 fuel thermal conductivity model in fuel performance code of FRAPCON-3.

REFERENCES

[1] J. Rest, An alternative explanation for evidence that xenon depletion, pore formation, and grain subdivision begin at different local burnups, J. Nucl. Mater., vol. 277, 2000, pp. 231-238.
[2] J. Rest, A model for the effect of the progression of irradiation-induced recrystallization from initiation to completion on swelling of UO2 and U–10Mo nuclear fuels, J. Nucl. Mater., vol. 346, 2005, pp. 226–232.
[3] J. Rest, Derivation of analytical expressions for the network dislocation density, change in lattice parameter, and for the recrystallized grain size in nuclear fuels ,J. Nucl. Mater., vol. 349, 2006, pp. 150–159.
[4] J. Rest, G. Kagana, A Physical description of fission product behavior in fuels for advanced power reactors, ANL-07/24, Argonne National Lab., 2007, pp. 21-26.
[5] J. Rest, editor: Rudy J. M. Konings, Comprehen. Nucl. Mater., vol.3, 2012, pp. 579-627.
[6] J. Spino, J. Rest, W. Goll, C. T. Walker, Matrix swelling rate and cavity volume balance of UO2 fuels at high burnup, J. Nucl. Mater., vol. 346 2005, pp. 131-144.
[7] B. Roostaii, H. Kazeminejad, S. Khakshournia, Influence of porosity formation on irradiated UO2 fuel thermal conductivity at high burnup , J. Nucl. Mater., vol. 479, 2016, pp. 374-381.
[8] P.G. Lucuta, Hj. Matzke, I.J. Hastings, A pragmatic approach to modelling thermal conductivity of irradiated UO_2 fuel: review and recommendations, J. Nucl. Mater., vol. 232, 1996, pp. 166-180.
[9] K. J. Geelhood, W.G. Luscher, FRAPCON-3.5: A Computer Code for the Calculation of Steady-State, Thermal-Mechanical Behavior of Oxide Fuel Rods for High Burnup, NUREG/CR-7022, vol. 1, 2014, pp. 84.
[10] J. Rest, The DART Dispersion Analysis Research Tool: A Mechanistic Model for Predicting Fission-Product-Induced Swelling of Aluminum Dispersion Fuels, AN L-95/36, 1995, pp. 16-20.
[11] K. Ohira and N. Itagaki, Thermal Conductivity Measurementsc of High Burnup UO2 Pellet and a Benchmark Calculation of Fuel Center Temperature, proceedings pp. 541-549. Applies to UO2., In Proc. of the ANS Topical Meet. on Light Water Reactor Fuel Performance, Portland, Oregon, 1997, pp. 541–549.
[12] DL. Hagrman, GA. Reymann, MATPRO version 11-A, Handbook of materials properties for use in the analysis of light water reactor fuel rod behavior, TREENUREC-1280, Revision 3, Adv. Inorg. Chem., 1979, pp. 485-486.
[13] Martín Lemes, Alejandro Soba, Alicia Denis, An empirical formulation to describe the evolution of the high burnup structure, J. Nucl. Engin. Tech., vol. 456, 2015, pp. 174-181.
[14] Yi Cui, Shurong Ding, Zengtao Chen, Yongzhong Huo, Modifications and applications of the mechanistic gaseous swelling model for UMo fuel, J. Nucl. Mater., vol. 457, 2015, pp. 157-164.
[15] J. Spino, A. D. Stalios, H. Santa Cruz, and D. Baron, Stereological evolution of the rim structure in PWR-fuels at prolonged irradiation: Dependencies with burnup and temperature, J. Nucl. Mater., vol. 354, 2006, pp. 66-84.
[16] C.B. Lee, .G. Bang, D.H. Kim, Y.H. Jung, Development of irradiation UO_2 thermal conductivity model, IAEA-TECDOC-1233, Session 6, 2001, pp. 363-371.
[17] R. Brandt, J. Neuer, Thermal conductivity and thermal radiation properties of UO_2, J. Non-Equilib. Thermodyn., vol. 1, 1976, pp. 3-23.
[18] C.T. Walker, D. Staicu, M. Sheindlin, D. Papaioannou, W. Goll, F. Sontheimer, On the thermal conductivity of UO2 nuclear fuel at a high burnup of around 100 MWd/kgHM, J. Nucl. Mater., vol. 350, 2006, pp. 19-39.