Degradation of Irradiated UO₂ Fuel Thermal Conductivity Calculated by FRAPCON Model Due to Porosity Evolution at High Burn-Up

B. Roostaii, H. Kazeminejad, S. Khakhshournia

Abstract—The evolution of volume porosity previously obtained by using the existing low temperature high burn-up gaseous swelling model with progressive recrystallization for UO₂ fuel is utilized to study the degradation of irradiated UO₂ 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 within UO₂ 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, UO₂ 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 UO₂ 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, UO₂ 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 et al. [6] as well as the Rest model [2], [4], [5] for UO₂ matrix swelling with progressive recrystallization, the present authors [7] quantified the influence of porosity evolution on the thermal conductivity of UO₂ 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 UO₂ fuel thermal conductivity due to porosity evolution at high burn-up. But in the present work the FRAPCON model [9] for UO₂ 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].

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 UO₂ Pellets related to the Nuclear Fuels Industries (NFI) model. This model applied to UO₂ thermal conductivity at 95% of theoretical density (TD) is expressed as (1) [9], [11].

\[
\frac{W}{\text{mk}} = \frac{1}{A + Bf(Bu) + (1 - 0.94 \exp(-0.04Bu))g(Bu)h(T)} + \frac{1}{E \exp \left( \frac{F}{T} \right)}
\]

\[
A = 4.52 \times 10^{-2} \text{ (mK/W)}
\]

\[
B = 2.46 \times 10^{-4} \text{ (mK/W/K)}
\]

\[
E = 3.5 \times 10^{9} \text{ (WK/m)}
\]

\[
F = 16361 \text{ (K)}
\]

\[
f(Bu) = 1.87 \times 10^{-3} \text{Bu (mk/W)}
\]

\[
g(Bu) = 0.039Bu^{0.28} \text{ (mk/W)}
\]

\[
h(T) = \frac{1}{1 + 396 \exp(-\frac{T}{Q})}
\]

\[
Q = 6380 \text{ (K)}
\]

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 UO₂ 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.

$$P_v = 1 - (1 + \Delta V_{\text{matrix}}/V_0^{\text{matrix}})(\rho/\rho_{\text{th}})$$

where $\rho$ is the bulk fuel density and $\rho_{\text{th}}$ is the theoretical density of UO$_2$, and $\Delta V_{\text{matrix}}/V_0^{\text{matrix}}$ presents total fractional fuel matrix swelling caused by both solid ($\Delta V_{\text{solid}}/V_0^{\text{matrix}}$) and gaseous ($\Delta V_{\text{gaseous}}/V_0^{\text{matrix}}$) fission products [7].

$$P_s = (\Delta V_{\text{gaseous}}/V_0^{\text{matrix}})/(1 + \Delta V_{\text{gaseous}}/V_0^{\text{matrix}})$$

where $V_0^{\text{matrix}}$ denotes the initial matrix volume, $\Delta V_{\text{gaseous}}$ is the increase of matrix volume due to bubble formation and $\Delta V_{\text{gaseous}}/V_0^{\text{matrix}}$ 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, material [10], can be proposed a model for porosity factor. 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).

$$k_{\text{eff}}^{V} = 1 - \left[\pi \frac{3^2}{4n} P_v^{\frac{2}{3}}\right] \left[1 - \frac{k_g}{2k_0 (\frac{3}{4n} P_v^{\frac{1}{3}})}\right]$$

where $k_{\text{eff}}^{V}$ 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 $P_v$ 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 ($\kappa_{P}$) to form of (6).

$$k_{\text{eff}}^{s} = 1 - \left[\pi \frac{3^2}{4n} P_s^{\frac{2}{3}}\right] \left[1 - \frac{k_g}{2k_0 (\frac{3}{4n} P_s^{\frac{1}{3}})}\right]$$

$$\kappa_{P} = \frac{k_{\text{eff}}^{s}}{k_0} = 1 - \left[\pi \frac{3^2}{4n} P_s^{\frac{2}{3}}\right] \left[1 - \frac{\pi (\frac{3}{4n} P_s^{\frac{1}{3}})}{2k_0 (\frac{3}{4n} P_s^{\frac{1}{3}})}\right] (6)$$

where $k_{\text{eff}}^{s}$ 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]:

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

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

$$k_{g} = \sum_{i=1}^{n} k_i \left[\frac{k_{0}}{1 + \sum_{j=1}^{n} M_{j} / M_{i}}\right]$$

$$\Phi_{ij} = \left[1 + 2.41 \frac{(M_{i} - M_{j})}{(M_{i} - M_{j})} \right]^{\frac{1}{2}}$$

where $k_i$ is the thermal conductivity of i gas, $X_i$ is the mole fraction of the i gas ($\frac{X_i}{\sum_i X_i} \approx 10.8$) [13], $M_i$ is the molecular weight of i gas ($M_{Xe} = 131.3, M_{Kr} = 83.8$) [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):

$$k_0 = k_{95} \times \left[1 - \pi \left(\frac{3}{4n} P_0^{\frac{1}{3}}\right)^{\frac{2}{3}}\right]^{-1}$$

$$k_d = k_0 \times \left[1 - \pi \left(\frac{3}{4n} P_s^{\frac{1}{3}}\right)^{\frac{2}{3}}\right]$$

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 $\left[1 - \pi \left(\frac{3}{4n} \right)^{\frac{2}{3}}\right]^{-1}$ 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 $P_v$, 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.

**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


