Development of a Fuel Performance Model of (Th,Pu)O_2

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A PhD Level Submission

Summary

A fuel performance model is being produced for (Th,Pu)O_2 ceramic fuel for the ongoing development of a design analysis tool for the Canadian SCWR. Work on the development of the model's grain growth and single grain fission gas diffusion capabilities will be presented, with the modelling results for grain size and fission gas release compared to Post Irradiation Examination (PIE) data from an irradiation experiment conducted at Chalk River Laboratories.Further development of the single grain fission gas diffusion behaviour is required.

1. Introduction

Canada's participation in the Generation IV International Forum (GIF) is being led by Canadian Nuclear Laboratories (CNL) in the development of a pressure tube type Super Critical Water cooled Reactor (SCWR). The proposed fuel is comprised of (Th,Pu)O_2 pellets (with ~13 wt% Pu in total heavy elements) [1]. By expanding the physical properties within an existing model for UO_2 (FAST), to include the properties of Th-mixed oxide ceramic fuel, a design analysis tool can be produced for SCWR (Th,Pu)O_2 fuel elements [2]. Initial modeled fuel centerline temperature results and a comparison of fission gas release results to PIE data from BDL-422 were reported in Prudil et al. [3]. BDL-422 was an irradiation experiment that examined (Th,Pu)O_2 fuel with 1.53 wt% Pu, with initial grain size between 3-4µm and assembled into National Research Universal (NRU) reactor36-element test bundles, labelled: ADA, ADC, ADD, ADE, and ADF [4,5]. The evaluation of two separate fuel grain growth models and the ongoing work examining the single grain fission gas diffusion in (Th,Pu)O_2 is presented in this work.

2. Grain Growth

Two separate grain growth models were examined to determine which provides more physically realistic results in comparison to the BDL-422 PIE data. The first model was presented in Goldberg et al. for the grain growth of ThO_2-based ceramic fuel given in equation (1)based on the assumption that grains can be approximated as spheres [6].

$$(g_d)^3 - \left(g_{di}\right)^3 = K_f \cdot t \cdot \exp\left(\frac{-Q}{RT}\right) \tag{1}$$

Where g_d, g_{di} are the grain diameter and initial grain radius of the ceramic fuel (cm), t is time in hours, R is the universal gas constant (8.314 J $mol^{-1}K^{-1}$), K_f is a fitting coefficient with a value of 800 $cm^3 \cdot h^{-1}$ maintained from Nichols' work on UO_2, T is the temperature in K, and Q is the vapour activation constant for $(Th,U)O_2$, with a value of 594×10^3 $J\cdot mol^{-1}$.Equation(1) is the steady state solution to Nichol's formulation for grain growth rate given by equation (2) [7].

$$\frac{dg_d}{dt} = \frac{k_g}{g_d^2} \tag{2}$$

The second grain growth model examined isgiven byKhoruzhii et al. for the grain growth of UO_2 during irradiation presented in equation (3) [8].

$$\frac{dg_d}{dt} = k_g\left(\frac{1}{g_d} - \frac{1}{g_{max}} - \frac{1}{g_{ir}}\right) \tag{3}$$

Here, k_g is the grain growth rate in m^2 s^{-1}, with its value found using equation (4), $g_{max}$is the maximum stable grain size (m) as a function of temperature given in equation (5), and g_{ir} is a function of temperature and fission rate that accounts for the irradiation effects on the grain size given in equation (6).

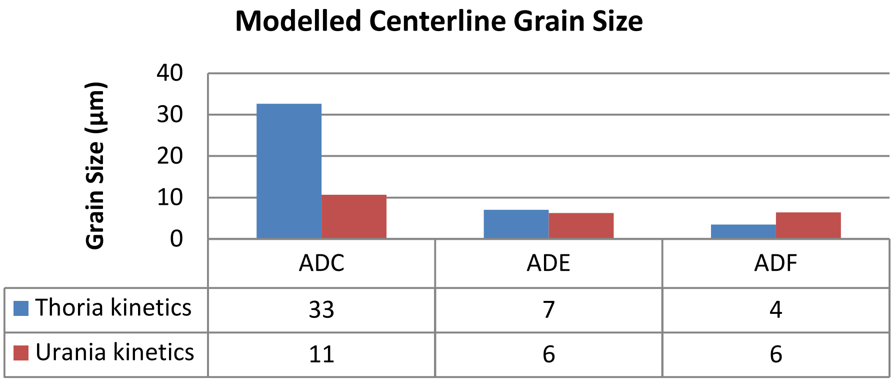
$$k_g = 1.46\times10^{-8}\exp\left(\frac{-32100}{T}\right) \tag{4}$$

$$g_{max} = 2.23\times10^{-3}\exp\left(\frac{-7620}{T}\right) \tag{5}$$

$$g_{ir} = \frac{6.71\times10^{18}\exp\left(\frac{-5620}{T}\right)}{F_{rate}\,T} \tag{6}$$

In equation (6),$F_{rate}$is the rateof fission in the fuel.

Figure 1 presents the modeled results for the centerline grain size of the elements from ADC, ADE and ADF from the BDL-422 experiment using both formulations of grain growth. Karam et al. reported that the centerline grain size from these elements is ~ 10 µm [5]. Of the three power histories, ADC experiences the highest linear power (67 kW m^{-1}) while ADF's maximum linear power is significantly lower (52 kW m^{-1}). This indicates that the temperature at which the Goldberg model begins to generate appreciable grain growth is higher than those achieved in the model of the element from ADF, and also demonstrates that it is not physically representative of BDL-422 fuel since the PIE results show there is grain growth. In the case of the modelled element from ADC, the grain growth from the Goldberg model exceeds the PIE measured value (33µm compared to ~10 µm). The grain growth model developed for UO_2 is currently recommended for the use of modelling $(Th,Pu)O_2$.



	ADC	ADE	ADF
🟦 Thoria kinetics	33	7	4
🟥 Urania kinetics	11	6	6

Figure 1 Comparison of modeled centerline grain sizes, using two different grain growth models.

3. Fission Gas Diffusion

The model used to calculate the fission gas diffusion through a single grain of UO_2 (D_0) used in FAST is given in equation (7). It was developed based on the work of Turnbull et al. [8-10], and is the weighted sum of diffusion coefficients that describe the contribution of three separate diffusion mechanism.

$$D_0 = D_{thrm} + 4D_{irr} + 4D_{athrm} \tag{7}$$

Here D_{thrm} is the diffusion coefficient due to thermally activated processes, D_{irr} is the diffusion coefficient due to irradiation induced vacancies, and D_{athrm} is the diffusion coefficient due to athermal effects.D_{thrm}, D_{irr}, and $D_{athrm}$are given in equations(8) – (10), respectively; all are in units of $m^2\ s^{-1}$. A full description of the development of equation (7) can be found in the thesis work of Morgan [8].

As this is a preliminary attempt at modelling $(Th,Pu)O_2$ fuel irradiation behaviour, changing the weighting of the three components of D_0 was performed to approximate the fuel's fission gas behaviour. At this point in time, there is limited data available on fission gas diffusion behaviour in Th-based fuel. Limiting the ability to derive a model of the fission gas diffusion in thorium, this is why this approach was taken. So far, sixteen different combinations of weighting factors have been examined in an attempt to replicate the fission gas release behaviour of the outer elements from BDL-422 that have undergone PIE. Three of the cases are shown in equations (8)-(10).

$$D_{0,ThPu,1} = 0.1D_{thrm} + 0.1D_{irr} + 0.1D_{athrm} \tag{8}$$

$$D_{0,ThPu,15} = D_{thrm} + 0.1D_{irr} + 0.1D_{athrm} \tag{9}$$

$$D_{0,ThPu,16} = 0.1D_{thrm} + 0.5D_{irr} + D_{athrm} \tag{10}$$

All fission gas diffusion through a single grain of $(Th,Pu)O_2$ permutations examined are of the form demonstrated in equations (8)-(10), $D_{0,ThPu,N}$,where N at the end of each subscript is an index number. Figures 2-4 display the modelled percent fission gas release results using each of the different diffusion coefficients for fuel elements from ADD, ADC and ADF, with the x-axis representing the index number N. The PIE measurement of the fission gas within the element is marked by a blue line (or lines, bundle ADD achieved high burnup> 1000 MWh $kgHE^{-1}$; the experimenters requested multiple elements to be measured for fission gas release, hence the range in Figure 2) [4, 11].

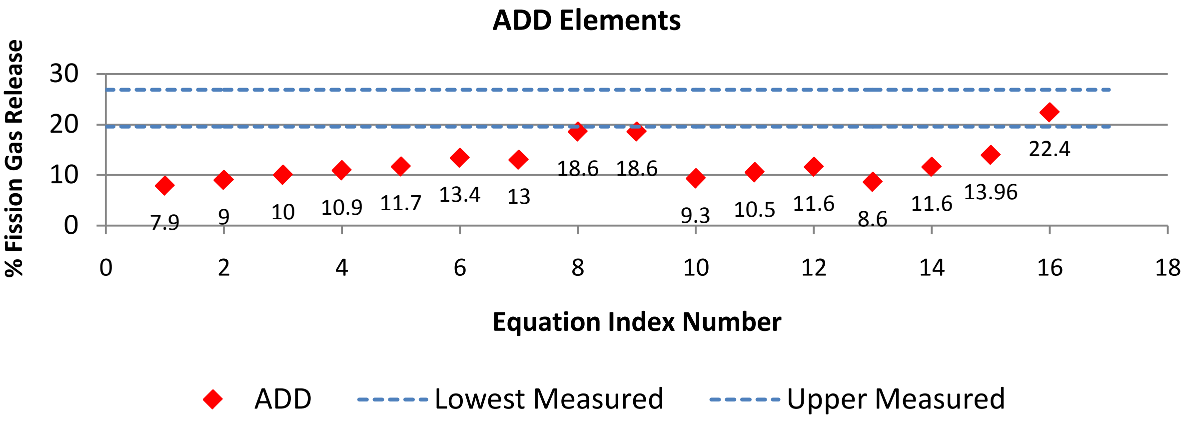


Figure 2 Fission gas release model results compared to PIE measurements of ADD elements.

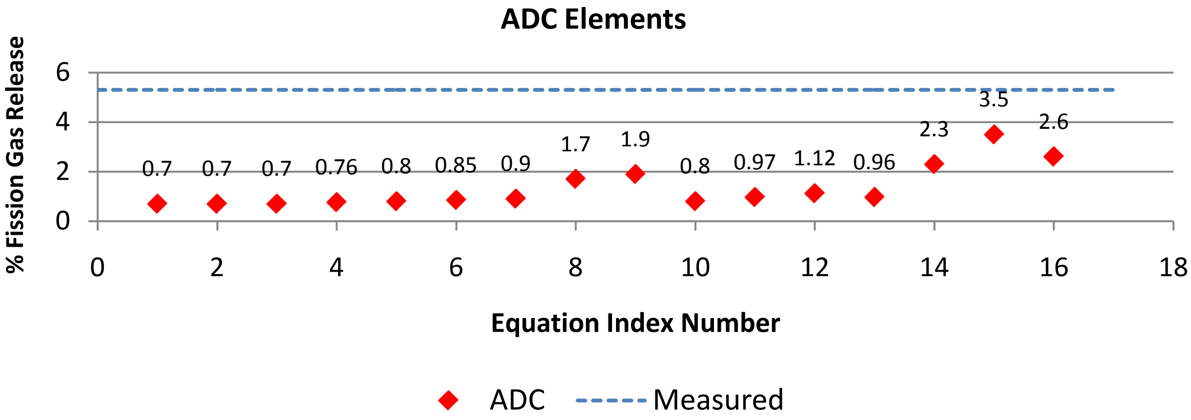


Figure 3Fission gas release model results compared to PIE measurements of ADC elements.

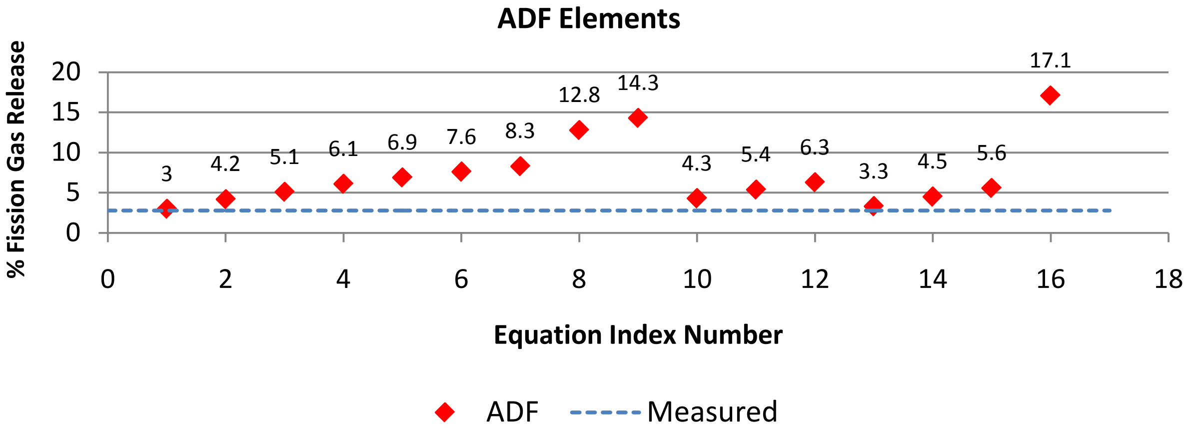


Figure 4 Fission gas release model results compared to PIE measurements of ADF elements.

$D_{0,ThPu,1}$ was the initial attempt, as most of the fission gas diffusion data on $(Th,U)O_2$ suggested that Th-based fuel has a diffusion rate approximately an order of magnitude lower than U-based fuel [2]. As seen in Figures 2-4, $D_{0,ThPu,1}$ produces good results for ADF, but drastically under predicts ADC and ADD. $D_{0,ThPu,16}$produced the best agreement with ADD, under predicts ADC, but greatly over predicts ADF. $D_{0,ThPu,15}$ generated the closest agreement to ADC than any of the other sixteencombinations, showed significant improvement in replicating ADD's measurements, while not drastically over predicting the release of ADF,as did $D_{0,ThPu,16}$. At this point in time, $D_{0,ThPu,15}$ is the recommended single atom diffusion coefficient.

The next step in the development of this model will be to examine the response of the fission gas release results based on the variations in D_0 that the sixteendifferent cases represent. From this data, it is envisionedthat an optimized series of weightings can be determinedand applied to each component of D_0 in order to replicate the behaviour of the BDL-422 gas release.

4. Conclusions

The development of a $(Th,Pu)O_2$ fuel performance model is underway, that will be validated against PIE data from the BDL-422 irradiation experiment. To date, the grain growth kinetics developed for UO_2appear to best represent $(Th,Pu)O_2$ grain growth; fission gas diffusion is behaviour is still being investigated.

5. Acknowledgements

Funding to the Canada Gen-IV National Program was provided by Natural Resources Canada through the Office of Energy Research and Development, Atomic Energy of Canada Limited, and Natural Sciences and Engineering Research Council of Canada.

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