Continuing Development of a Fuel Performance Model for Th-Based Ceramic Fuel

J.S. Bell[1]and P.K. Chan[1]
[1]Royal Military College of Canada, Kingston, Ontario, Canada
(john.bell@rmc.ca)

Abstract

A finite element analysis based fuel performance model for Th-based ceramic fuel is being developed. The development of a $(Th,Pu)O_2$ model is novel, and will be capable of providing estimates of fission gas release and sheath/clad strains. Once this model is complete, it will be used to perform a design analysis for the conceptual fuel for the Canadian SCWR pellets and cladding.Results obtained for $(Th,U)O_2$ compared with Post Irradiation Examination(PIE) data from the DME-221 irradiation test are encouraging. The current state of the model, as well as modelling results compared to data from a $(Th,Pu)O_2$ fuel irradiation experiment, will be presented.

1. Introduction

The Generation IV International Forum (GIF) was established to undertake the research required to determine the feasibility and performance capabilities of the next generation of nuclear energy systems [1]. The GIF identified six reactor designs to focus research upon. One of these reactor designs is the Super Critical Water cooled Reactor (SCWR). Canada's participation in GIF is led by Canadian Nuclear Laboratories (CNL), with the design of a pressure-tube-based SCWR [2]. The reactor is in the design concept phase. The proposed fuel is comprised of ceramic thorium-plutonium mixed-oxide ($(Th,Pu)O_2$, 13 wt% Pu content) pellets. The fuel will operate in supercritical water coolant, pressurized to 25 MPa and temperatures ranging from 315-625^{0}C. Linear element rating limits and target exit burnup goals have been established for the fuel [3]. With coolant and fuel conditions provided, a fuel performance model is being developed to support a feasibility study on fuel designs [4].

Work to develop a fuel performance model for the Canadian SCWR is ongoing at the Royal Military College of Canada (RMCC).Results from a test model (modelling UO_2fuel pellets in SCWR coolant conditions), support the use of a finite-element solver (such as COMSOL Multiphysics)to predict fuel behavior [4]. The development of a model for Th-based ceramic fuel pellets is ongoing. Aninitial $(Th,U)O_2$ model has been developed. The model's results were compared to PIE data from DME-221,an irradiation experiment conducted at CNL's Chalk River Laboratories (CRL) that examined three different fuel compositions:ThO_2, $(Th,U)O_2$ with 1.0 wt.% ^{235}U and $(Th,U)O_2$with 1.5 wt.% ^{235}U. It was found that the model demonstrated the expected reduction in fuel temperature compared to UO_2 fuel, and predicted fission gas release results comparable to the PIE data [5,6].

This model has been used to attempt to replicate the irradiation behaviour of another irradiation experiment conducted at CRL. BDL- 422 was a fuel irradiation experiment that was undertaken to demonstrate the ability of $(Th,Pu)O_2$ bundles to operate to high burnups (>1000 MWh/kgHE) (Heavy

Element [HE]) [7,8]. Six bundles were irradiated in test loops of the NRU reactor. Each bundle was fuelled exclusively with (Th,Pu)O_2 with 1.53 wt% Pu. In order to avoid potential over pressurization caused by fission gas release, the outer ring elements were fabricated with plena at both ends of each element. The BDL-422 fuel pellets had an average initial grain size of 3-4 µm and an average initial pellet density of 9.469 g cm^{-3}. The six BDL-422 bundles were labelled ADA – ADF. A selection of outer ring elements (OE) and an additional element from the intermediate bundle ring (IE) were chosen for PIE from ADA, ADC, ADD, ADE and ADF[7, 8].The modelling assumptions and results will be discussed within the context of model predictions of: pellet centerline temperature, centerline grain size, and fission gas release, with the grain size and fission gas release results compared to PIE data.

2. Centreline Temperature

The model calculates the fuel teperature by approximating the solution to the heat conduction equation (equation (1)) using finite element methods.

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + Q_{prod} \quad (1)$$

Here,T is the temperature (K), t is time (s), ρ is the density of the material (kg m^{-3}), C_p is the specific heat capacity of the material (J K^{-1}), k is the thermal conductivity of the material (W m^{-1} K^{-1}), and Q_{prod} accounts for the heat produced within the fuel pellet (J $m^{-3}s^{-1}$).

A sensitivity analysis that examined which material properties had the greatest effect on the temperature results of a UO_2 fuel performance model, found that changes in the thermal conductivity (k) of the UO_2 was the dominant factor [9]. This would indicate that any modelling assumption concerning the thermal conductivity of the fuel pellet will be significant to the model's overall performance. The assumptions made to model the thermal conductivity of (Th,Pu)O_2 will be outlined for the purposes of discussingthe model's results for the fuel centerline temperature of the BDL-422 experimental fuel.

Lucuta presented a method to describe the effects of fuel burnup on the thermal conductivity of UO_2 [10].The underlying assumption in this derivation of UO_2 thermal conductivity is that separate physical changes in the fuel during irradiation act individually on the thermal conductivity. The effect of physical changes on thermal conductivity can then be accounted for as a multiplication "correction" factor on the thermal conductivity of 100% theoretical density, unirradiated UO_2.The primary modelling assumption for the thermal conductivity of (Th,Pu)O_2 is that the correction factors developed for UO_2 are applicable to (Th,Pu)O_2. The application of Lucuta's thermal conductivity factors to (Th,U)O_2simulated the temperature conditions required to model the fission gas release of that fuel composition [5,6,11].

The (Th,Pu)O_2 fuel performance model uses the Cozzo correlation for unirradiated (Th,Pu)O_2 for a ceramic at 95% theoretical density as given in equation (2) [12].

$$k_{Cozzo} = \frac{1}{A + BT}$$
$$A = 0.006071 + 0.572 wtPu - 0.5937 wtPu^2 \quad (2)$$

$B = 0.00024$

Here, $wtPu$ is the wt% of PuO_2 in the fuel matrix (0-100% PuO_2), with the correlation being derived from a series of measurements in the temperature range of 500 K to 1600 K. The model currently uses this correlation unaltered; a correction factor still needs to be applied to the Cozzo correlation in order for it to reflect the thermal conductivity of 100% dense $(Th,Pu)O_2$.

Figure 1 presents the unirradiated thermal conductivity for BDL-422 fuel as predicted by the Cozzo correlation in comparison to the unirradiated UO_2 thermal conductivity predicted by the ELESTRES fuel codefor the same temperature range[9, 13].

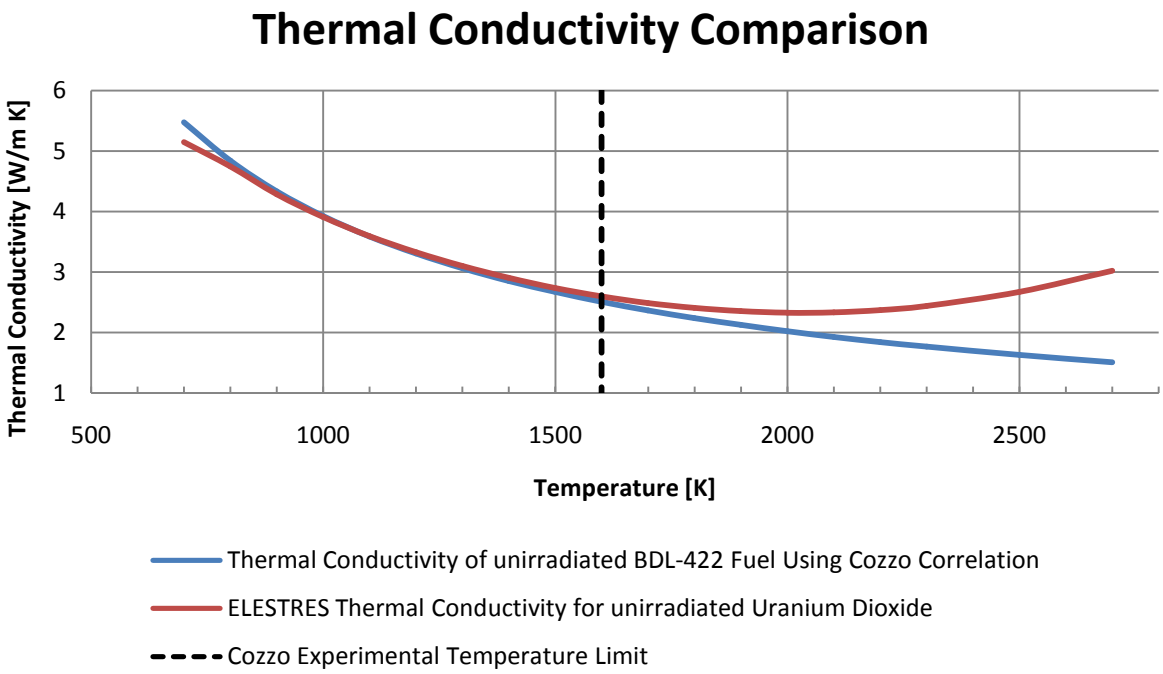

Figure 1Thermal conductivity of BDL-422 fuel compared to UO_2.

As the temperature of the BDL-422 fuel increases to about 1100 K, there is little difference in the thermal conductivity of the $(Th,Pu)O_2$ in comparison to UO_2. Once the $(Th,Pu)O_2$ fuel begins to exceed approximately 1500 K, the correlation begins to diverge from theUO_2 prediction. Beyond the experimental temperature measurements made by Cozzo, it continues to diverge to the point where the difference is a factor of ~2 at 2700 K.

Table 1 presents the modeled maximum centerline temperature results for the BDL-422 fuel and UO_2 fuel experiencing the same power histories as those experienced by the BDL-422 fuel.

Table 1 Modeled maximum centerline temperature within the fuel.

BDL-422 Power	Modeled Max. Temp.	Modeled Max. Temp.	Temperature

History (Max. Linear Power [kW/m], Exit Burnup [MWh/kgHE])	UO_2 Fuel (K)	BDL-422 Fuel (K)	Difference (K)
Outer Elements			
ADA (54, 1181)	1953	2020	-67
ADD (73, 1082)	2468	2845	-377
ADC (67, 451)	2394	2717	-323
ADE (64, 597)	2156	2355	-199
ADF (52, 856)	1887	1896	-9
Intermediate Elements			
ADA (27, 718)	1364	1415	-51
ADD (45, 665)	1578	1636	-58
ADC (22, 288)	1047	1086	-39
ADE (32, 308)	1223	1276	-53
ADF (39, 531)	1358	1413	-55

All of the predicted temperatures for the BDL-422 fuel with power histories that experienced < 60 kW m^{-1}arehotter (within 70 K) of the UO_2 under the same irradiation conditions. For the high power cases (> 60 kW m^{-1}), the temperature difference is ≥200 K. These results reflect the divergence of the thermal conductivity correlations at temperatures greater thanCozzo's experimental temperature limits.

A similar continuous decrease in thermal conductivity is reported by Long et al. in the correlation used in their (Th,U)O_2fuel model [11]. In the development of their model to account for temperatures exceeding 2200 K (the limit on the thermal conductivity correlation), an additional term with T^3 dependence to the correlation is usedto fit the conductivity of ThO_2 at its melting point. The inclusion of this term inthe correlation leads to an increase in the thermal conductivity beyond 2200 K. Since there is limited high temperature data on the thermal conductivity of (Th,Pu)O_2, the same exercise applied to Cozzo's correlation may provide a more realistic estimate. This will be examined in order to determine if it leads to significant changes in the modeled fuel temperature.

3. 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 by Goldberg et al. for the grain growth of ThO_2-based ceramic fuel given in equation (3), based on the assumption that grains can be approximated as spheres [14].

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

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[15], T is the temperature in K, and Q is the vapouractivation constant for (Th,U)O_2, with a value of 594×10^3 J·mol^{-1}[14]. Equation (3) is the steady state solution to Nichol's formulation for grain growth rate given by equation (4).

$$\frac{dg_d}{dt} = \frac{k_g}{g_d^2} \quad (4)$$

The second grain growth model examined is given by Khoruzhii et al. for the grain growth of UO_2 during irradiation presented in equation (5) [16].

$$\frac{dg_d}{dt} = k_g \left(\frac{1}{g_d} - \frac{1}{g_{max}} - \frac{1}{g_{ir}}\right) \quad (5)$$

Here, k_g is the grain growth rate in $m^2\ s^{-1}$, with its value found using equation (6), g_{max} is the maximum stable grain size (m) as a function of temperature given in equation (7), and g_{ir} is a function of temperature and fission rate that accounts for the irradiation effects on the grain size given in equation (8).

$$k_g = 1.46 \times 10^{-8} \exp\left(\frac{-32100}{T}\right) \quad (6)$$

$$g_{max} = 2.23 \times 10^{-3} \exp\left(\frac{-7620}{T}\right) \quad (7)$$

$$g_{ir} = \frac{6.71 \times 10^{18} \exp\left(\frac{-5620}{T}\right)}{F_{rate}\ T} \quad (8)$$

In equation (8), F_{rate} is the rateof fission in the fuel.

Figure 2 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 $(Th,Pu)O_2$. It is unknown if the growth rate in equation (4) is entirely accurate; however, the Khoruzhii model is more representative than the Goldberg model. From the results of modelling ADC, the inclusion of the g_{max} and g_{ir} are apparently beneficial by limiting grain growth and lead this model to be physically representative.

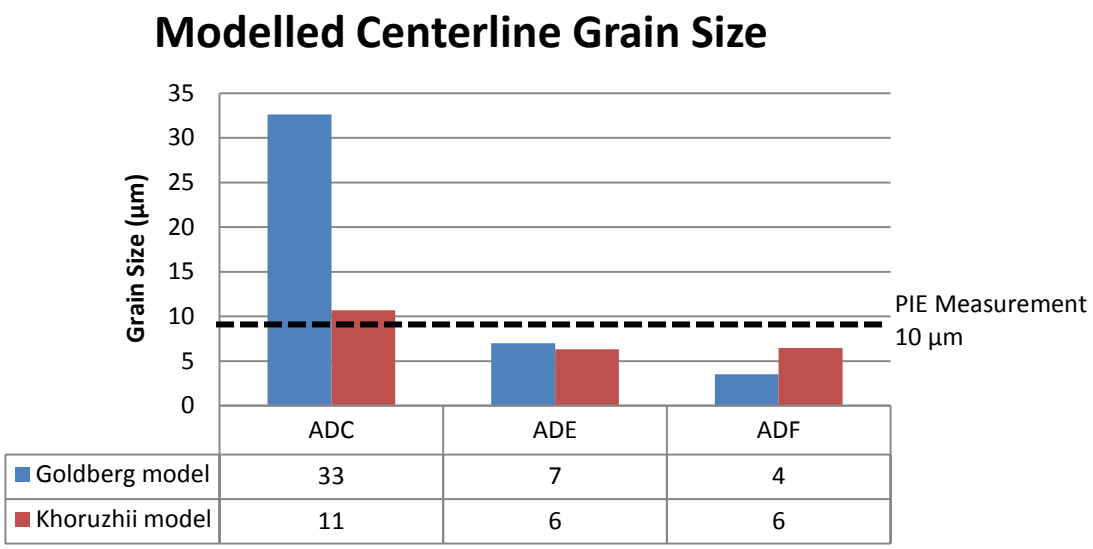

Figure 2Comparison of modelled centerline grain sizes, using two different grain growth models.

4. Fission Gas Release

Fission Gas Release (FGR) is modeled as a two step process. The first step is to solvefor the concentration of fission gas within fuel grains using the Booth diffusion model.This solution leads to the release rate of fission gas atthe grain boundaries. Fission gas then accumulates at the grain boundaries. The second step within the model is the determination of the boundary saturation conditions; once the boundaries become saturated with fission gas, any fission gas that reaches the grain boundary past this point is released to the free volume of the element. The Booth diffusion equation is given in equation (9).

$$\frac{\partial C}{\partial t} = D\nabla^2 C + P_{fg} \tag{9}$$

In equation (9),C is the concentration of fission gas atoms (atoms m^{-3}), ∇^2 is the Laplacian in spherical coordinates, and P_{fg} is the volumetric production rate of fission gas (atoms $m^{-3}s^{-1}$), and D is the net fission gas diffusion rate ($m^2\,s^{-1}$). P_{fg} is determined through the percent yield of stable fission gas per fission multiplied by the rate of fission (F_{rate}). In the development of the UO_2 fuel performance model at RMCC, the determination of D is based on the work by Turnbull et al. and given by equation (10) [17-19].

$$D = \frac{D_0 b'}{b' + g_a} \tag{10}$$

Here,b' is the intragranular resolution rate in s^{-1}, g_a is the trapping rate in s^{-1}, and D_0 is the single fission gas atom diffusion coefficient for a fully dense UO_2 crystal as given. The value of D_0 for UO_2 is found by weighted sums of diffusion coefficients that describe the contribution of three separate mechanisms given in equation (11).The details of the b' andg_a are omitted for the sake of brevity.

$$D_0 = D_{thrm} + 4D_{irr} + 4D_{athrm} \quad (11)$$

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(12) – (14) respectively; all are in units of $m^2\ s^{-1}$

$$D_{thrm} = 7.6 \times 10^{-9} exp\left(\frac{-2.93\times10^5}{RT}\right) \quad (12)$$

$$D_{irr} = 10^{13}\Omega^{2/3}C_v^0 \frac{\Omega^{2/3}\mu^2+Z_n C_v^0}{2Z_n}\sqrt{1+\frac{2\times10^6 F_{rate}\,\Omega Z_n}{v_v\left(\Omega^{2/3}\mu^2+Z_n C_v^0\right)}} \quad (13)$$

$$D_{athrm} = 2 \times 10^{-40} F_{rate} \quad (14)$$

Ω is the atomic volume in m^3 given by equation (15).

$$\Omega = \frac{M_U}{\rho_{UO_2STP}\, N_{Av}} \quad (15)$$

$C_v^0$is the vacancy concentration of unirradiated UO_2 given by equation (13)

$$C_v^0 = exp\left(\frac{-27780}{T}\right) \quad (16)$$

Here,$\mu \approx 10^{15}$ m^{-2} is the sink strength for crystallographic point defects (dislocations and vacancies),$v_v = 10^{13}C_v^0$ is the vacancy jump frequency,$Z_n \approx 100$ is the average number of sites available for recombination around defects which will inevitably recombine, M_U is the molar mass of natural uranium, ρ_{UO_2STP} = 10980 kg m^{-3} is the density of UO_2 at STP and N_{Av} is Avogadro's number.

The modelling assumption made to develop the fission gas release modelfor (Th,Pu)O_2 is that the grain boundary saturation and release to the free volume of an element are unchanged (fission gas mixture is still predominately comprised of Xe and Kr), but the ability of a fission gas atom to diffuse through a grain of fuel is different from UO_2. 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 thoria.

So far, 16 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 (17)-(19).

$$D_{0,ThPu,1} = 0.1D_{thrm} + 0.1D_{irr} + 0.1D_{athrm} \quad (17)$$

$$D_{0,ThPu,15} = D_{thrm} + 0.1D_{irr} + 0.1D_{athrm} \quad (18)$$

$$D_{0,ThPu,16} = 0.1D_{thrm} + 0.5D_{irr} + D_{athrm} \quad (19)$$

All fission gas diffusion through a single grain of (Th,Pu)O_2 permutations examined are of the form demonstrated in equations (7)-(19), $D_{0,ThPu,N}$, where N (at the end of each subscript) is an index number. Figures 3 - 7 display the modelled percent fission gas release results using each of the different diffusion coefficients for outer ring fuel elements from the five bundles that underwent PIE, 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 ADA and ADD achieved high burnup > 1000 MWh kgHE^{-1}; multiple elements were measured for fission gas release, hence the ranges in %FGR in Figures3 and 4) [7, 20].

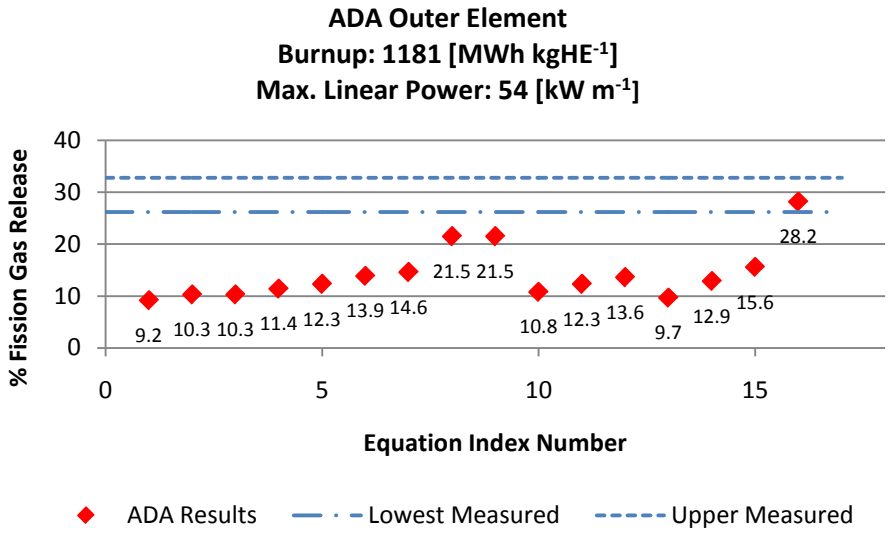

Figure 3 % FGR modeled with various single grain diffusion coefficients.

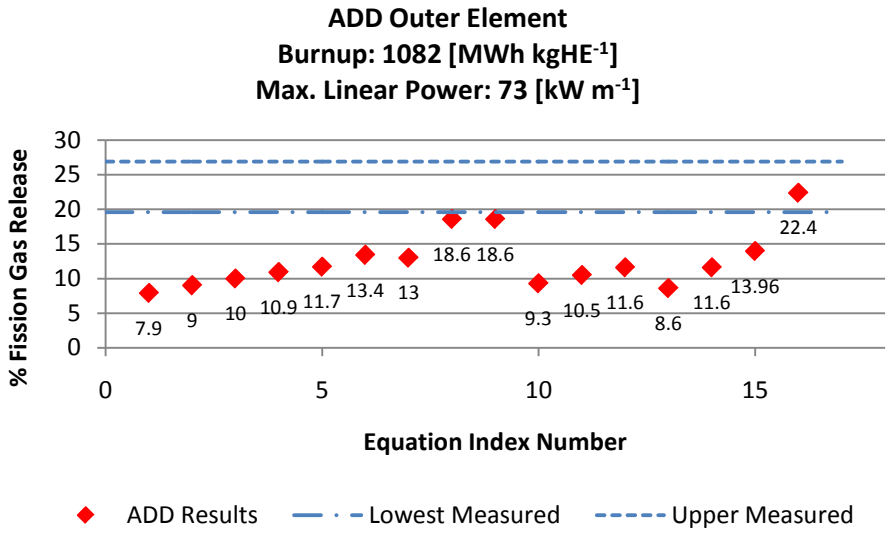

Figure 4% FGR modeled with various single grain diffusion coefficients.

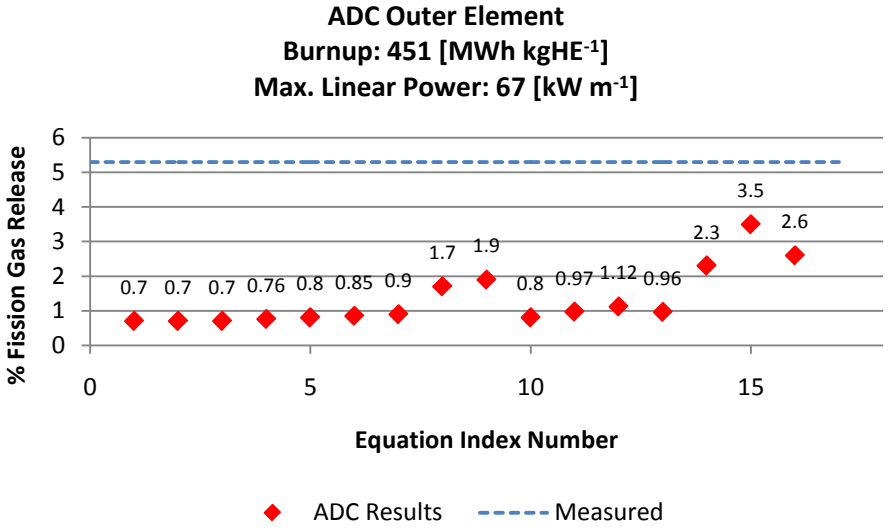

Figure 5 % FGR modeled with various single grain diffusion coefficients.

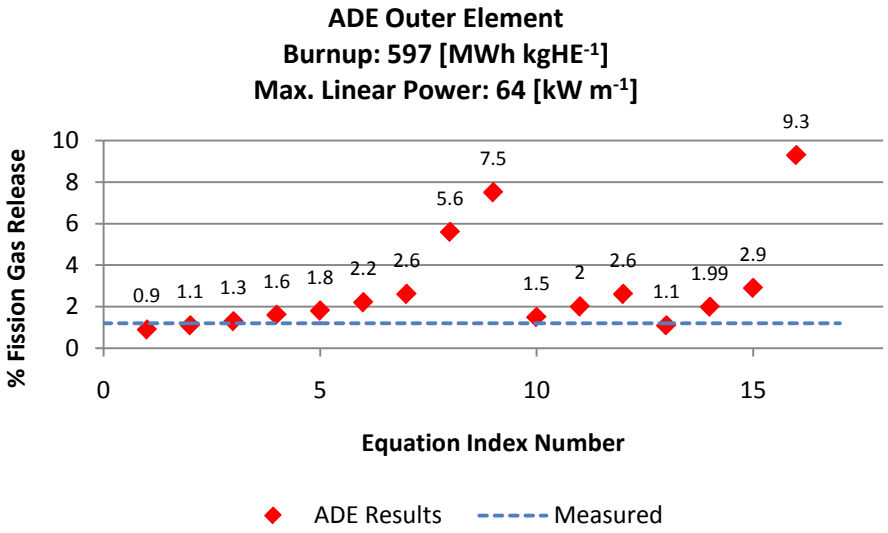

Figure 6 % FGR modeled with various single grain diffusion coefficients.

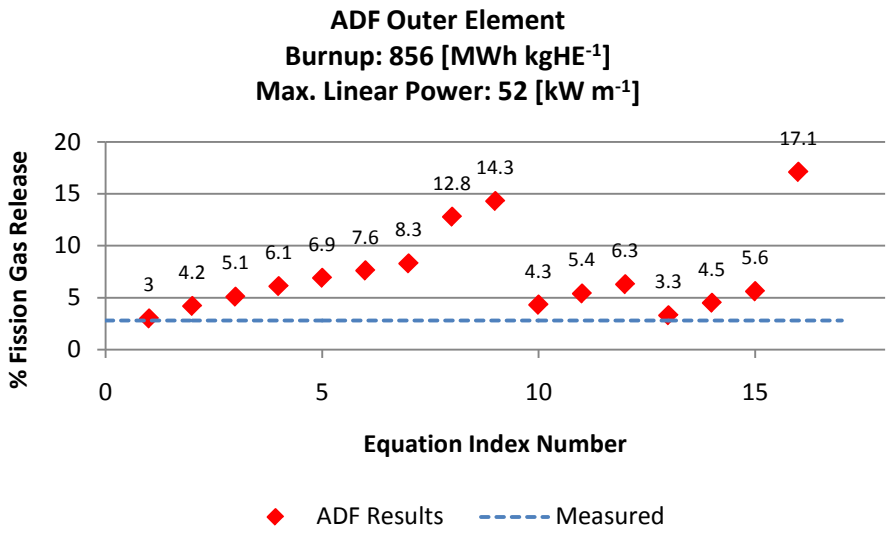

Figure 7 % FGR modeled with various single grain diffusion coefficients.

$D_{0,ThPu,1}$(equation (17)) was the initial attempt, as most of the fission gas diffusion data on (Th,U)O_2 suggested that Th-based fuel is well approximated by 0.1*D [5]. As seen in Figures 6 and 7,$D_{0,ThPu,1}$produces decent results for ADE and ADF, but drastically under predicts ADA, ADC, and ADD. $D_{0,ThPu,1}$was the diffusion coefficient used in the comparison of modelled centerline temperatures; this analysis will be repeated when a more representative diffusion coefficient is determined. $D_{0,ThPu,16}$(equation (19)) produced the best agreement with ADA and ADD, under predicts ADC, but greatly over predicts ADE and ADF. $D_{0,ThPu,15}$(equation (18)) generated the closest agreement to ADC than any of the other 16 combinations, showed significant improvement in replicating ADA and ADD, while not drastically over predicting the release of ADE and 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 16 different cases represent. From this data, it is envisionedthat an optimized series of weightings can be found and applied to each component of D_0 in order to replicate the behaviour of the BDL-422 gas release.

5. Summary and Conclusions

A preliminary (Th,Pu)O_2 model has been developed, and is still in progress.An attempt to validate the model to the PIE results of BDL-422 demonstrates that Khoruzhii's model for UO_2 grain growth provides more realistic grain size predications, and that considerable work is still required in order to find a fission gas diffusion coefficient that will replicate the fission gas release behavior of BDL-422. The examination of the centerline fuel temperature and the behaviour of the Cozzo correlation for (Th,Pu)O_2 thermal conductivity has highlighted the need to attempt to extend the current capabilities beyond 1600 K.

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