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Development of a Fuel Performance Model for Evaluating Conceptual Th-Based Canadian SCWR Fuel Designs

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ABSTRACT –The fuel assembly for the Canadian Super Critical Water Reactor (SCWR) is in the conceptual design phase. The proposed fuel pellets are made of ceramic Th-Pu mixed oxide ($(Th,Pu)O_2$). Neutronics and thermal hydraulics calculations are being undertaken by the nuclear industry to optimize the fuel assembly within a pressure tube. The SCWR working groupshave establishedtwo conceptual fuel element designs (outer diameter, fuel composition, cladding material, exit burnup etc.) within anassembly for performance assessment.

A detailed fuel element performance assessment underin-reactor conditions couldbe used to determine cladding material thickness/suitability and to optimize the fuel pellet geometry. This work reports the development of a fuel performance model to predict the behaviour ofthe Canadian SCWR fuel using the finite element method. Aninitial approach is to developa thorium-uranium mixed-oxide ($(Th,U)O_2$) model.Preliminary results from this model agree with fuel irradiationdata . Uranium dioxide (UO_2) fuel, under the same conditions, isalso being modeled and compared. Aplan to model $(Th, Pu)O_2$SCWR fuel is briefly presented here.

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 SuperCritical Water cooled Reactor (SCWR). Canada's participation in GIF is led by Atomic Energy of Canada Limited (AECL), 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% Pu content) pellets.For the purpose ofneutronics andthermal hydraulics calculations to optimize the fuel assembly design, two element designs are proposed (having different element outer diameters) with pellets being clad withina 310 stainless steel.The fuel will operate in supercritical water coolant, pressurized to 25 MPa and temperaturesranging from 315-625^{o}C.Linear element rating limits and target exit burnup goalshave also 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. The benefit of producing a fuel performance model at this timeis that it can provide a tool for the assessment of pellet and clad designs to meet fuel safety qualification limits [4].

Preliminary 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 (modeling UO_2 fuel pellets in SCWR coolant conditions), support the use of a finite-element solver (such as COMSOL Multiphysics)to predict fuel behavior [4]. Work is still required on the test model. The development of a model for Th-based ceramic fuel pellets has begun and the results of these efforts are presented in this paper.

1. Model Development

The SCWR fuel performance model is based on the Fuel And Sheath modeling Tool (FAST), a CANDU fuel performance model developed at RMCC [5]. Each successive step in the development of the SCWR model can be viewed as introducing new sets of equations that will enable FAST to model specific aspects of the SCWR fuel. Currently,FAST provides an approximate solution to the heat transport and deformation equations, using the finite element method on ahalf-axially symmetric UO_2pellet and sheath (Zircaloy-4) geometry. In the development of the test model[4], the SCWR coolant conditions and 310 stainless steel clad were examined. In order to model SCWR fuel, a model for the temperature dependent irradiation behaviour of a Th-based ceramic fuel needs to be developed as a first step.

Correlations are available for the temperature dependent behaviour of both thorium-uranium mixed oxide ($(Th,U)O_2$) and $(Th,Pu)O_2$ ceramics [6]. From an ongoing literature review, littledata has been found on the irradiation of $(Th,Pu)O_2$;specifically, there has been no data found on the diffusion of gaseous fission products throughsingle grain or poly crystalline $(Th,Pu)O_2$. The review is ongoing and may yet uncover data pertaining to this. However, data is available from the irradiation of $(Th,U)O_2$ fuel [7-10].With this information, a fission gas release model can be implemented for $(Th,U)O_2$.

The release of gaseous fission products from the fuel pellet into the free volume in a fuel element is of particular interest to fuel performance modeling. Gas release is thermally driven, dependent on fuel restructuring,and degrades heat transport from the fuel to the clad [11].Initial work on developing a model for Th-based ceramic pellets has focused on $(Th,U)O_2$, because it is expected that the fission gas release behavior (and other irradiation effects) of $(Th,Pu)O_2$will be more similar to $(Th,U)O_2$ than UO_2, and assumptions can be made if (Th,Pu) data does not exist.

The following subsections present the $(Th,U)O_2$ properties that are being used to develop this preliminary ceramic Th-based fuel model. Physical phenomena such as theoretical density,thermal conductivity, heat capacity, thermal expansion, Young's modulus, Poisson's ratio, equiaxed grain growth, fission gas production and granular fission gas diffusion are presented. All other aspects of the pellet behaviour are kept as those used by FAST for UO_2.

1.1 Theoretical Density

In 2006, the IAEA published a technical document that gave recommendations for correlations of material properties for many of the materials used in water cooled reactors [12]. A correlation for determining the theoretical density of $(Th,U)O_2$ is used within the model given in equation (1).

$$\rho^{Th}_{(\mathrm{Th,U})\mathrm{O2}} = 10.087 - 2.891 \times 10^{-4}T - 6.354 \times 10^{-7}y + 5.111 \times 10^{-6}y^2 \quad (1)$$

Where $\rho^{Th}_{(\mathrm{Th,U})\mathrm{O2}}$ is the density of the ceramic in g·cm^{-3} , T is the temperature in K and y is the weight percent (wt. %) of UO_2 in the ceramic. Equation (1) is valid for a temperature range of 298-1600K.The theroretical density is used to determinbe the initial porosity of the "as fabricated" ceramic pellet at 293 K. Density changes during irradiation are determined based on changes to the as-fabricated density.

1.2 Thermal Conductivity

FAST uses Lucuta's approach to modeling the irradiation effects on the thermal conductivity of UO_2[13]. The underlying assumption in this derivationofUO_2thermal conductivity, is that separate physical changes in the fuel during irradiationact individually on the thermal conductivity. The effect of physical changes on thermal conductivity can be accounted for as a multiplication factor on the thermal conductivity of 100% theoretical density, unirradiated UO_2. By assuming that these changes have the sameeffect on $(Th,U)O_2$, thermal conductivity can be calculated during irradiation by switching out the correlation for unirradiatedUO_2 for a similar $(Th,U)O_2$ correlation. Similar to Loewen etal. and Long etal., in the their development of Th modeling capabilities in FRAPCON [9],the unirradiated UO_2 correlation is replaced with one derived by Belle and Berman for $(Th,U)O_2$up to 30% UO_2 content below 2200 K given in equation (2) [14].

$$k_0 = \frac{1}{A+BT} \quad (2)$$

Wherek_0 is the thermal conductivity in W·m^{-1}·K^{-1}, A is given by equation (3) and B is given by equation (4)

$$A = \frac{1}{46.948 - 112.072M_U} \quad (3)$$

$$B = 1.597 \times 10^{-4} + 6.736 \times 10^{-4}M_U - 2.155 \times 10^{-3}M_U^2 \quad (4)$$

Where$M_U$is the mole percent of UO_2[14].

1.3 Heat Capacity

The heat capacity at constant pressure for $(Th,U)O_2$ as reported by Dash etal. is used in the development of the model. Their correlation is given in equation (5) [6].

$$C_p = (66.26 + 10.91y) + (0.00923 - 0.00065y)T - (7.70 \times 10^{-5} + 6.7 \times 10^{-5}y)T^2 \quad (5)$$

Where C_p is the heat capacity in J·mol^{-1}·K^{-1}, valid for temperatures 298-2000K.

1.4 Thermal Strain

The strain caused by thermal expansion, as described by Bakker et al., is given in equations (6) and (7)[15]and implemented within the model.

$$\frac{\Delta L}{L_0} = -0.179 - 0.087y + (5.097 + 4.705y) \times 10^{-4}T + (3.732 - 4.002y) \times 10^{-7}T^2 + (-7.594 + 11.98y \times 10^{-11}T^3 \quad (6)$$

Equation (6) is valid for temperatures of 273K- 923K. Above 923K, equation (7) describes the thermal strain in (Th,U)O_2.

$$\frac{\Delta L}{L_0} = -1.79 - 1.49y + (5.097 + 6.693y) \times 10^{-4}T + (3.732 - 4.002y) \times 10^{-7}T^2 + (-7.594 + 19.784y \times 10^{-11}T^3 \quad (7)$$

Equation (7) is valid up to 2000 K,where *y* (in both (6) and (7)) is the wt% UO_2 in the ceramic ranging from 0 to 1(i.e. 10 wt% UO_2 is *y*=0.1).

1.5 Young's Modulus

The Young's modulus used for the (Th,U)O_2 model is one reported by Belle and Bermanfor ThO_2[14] and given in equation (8).

$$E^{(Th,U)O_2} = E_{273}^{(Th,U)O_2} P\left(1.023 - 1.405 \times 10^{-4} T e^{\frac{-181}{T}}\right) \quad (8)$$

Where $E_{273}^{(Th,U)O_2}$ is the young's Modulus at 273 K with a value of 249.1 GPa and *P* is the porosity given by equation (9).

$$P = 1 - \frac{\rho_{(\mathrm{Th,U})\mathrm{O2}}}{\rho^{Th}_{(\mathrm{Th,U})\mathrm{O2}}} \quad (9)$$

With $\rho^{Th}_{(\mathrm{Th,U})\mathrm{O2}}$ given by (1) and $\rho_{(\mathrm{Th,U})\mathrm{O2}}$ is the current density of the (Th,U)O_2 pellet[14].The materials used to determine equation (8) included specimens that had as much as 5 wt% CaO.

1.6 Poisson's Ratio

Belle and Berman also report that the Poisson`s ratio for (Th,U)O_2 has a value of 0.28[14].

1.7 Equiaxed Grain Growth

The grain growth correlation used in the model is from a summary report from Light Water BreederReactor(LWBR)by Goldberg et al. They propose an equation ((10)) that describes equiaxed grain growth based on work by Nichols on the grain growth of UO_2[10].

$$D^3 - D_i^3 = K_f t e^{\frac{-Q}{RT}} \quad (10)$$

Where $D_i$is the initialgrain diameter within the ceramic fuel pellets in 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, and$Q$ is the vapour activation constant for $(Th,U)O_2$, with a value of $594\times10^3 J \cdot mol^{-1}$. The value of$Q$ was determined for 20-30% wt% UO_2.

1.8 Fission Gas Production

In the analysis of the LWBR irradiations, it was found that when ^{233}U undergoes fission, 1.6 timesthe amount of stable Kr (including ^{85}Kr) is produced (compared to ^{235}U). This leads to a fission gas production rate (Xe+Kr) that is~10% higher than that of UO_2 fuel [8, 13]. The fission gas production rate in the model has been adjusted to reflect the conservative estimate of ^{233}U to be the sole isotopic source of fission.

1.9 Single Atom Fission Gas Diffusion

In irradiations of experimental fuel at AECL- Chalk River Labs (CRL), it has been observed that the fission gas release from Th-based fuels characterized by homogeneous pellet microstructures, is significantly less than that observed in UO_2 fuel [16, 17].This finding indicates that, despite the increased production of fission gas from ^{233}U, less is escaping into the free volume of an element.During the irradiation test of a defected $(Th,U)O_2$ fuel element, the online monitoring of the ^{133}Xe released was found to be 1to 2 orders of magnitude less than that of UO_2 fuel [17] (comparing experiments with fabricated defects (drilled hole) on the sheath). The ingress of coolant into a defected element leads to fuel oxidation in UO_2, which affects the thermal conducitvity and leads to an increase ingas release. It is uncertain what reactions occur withdefected Th based fuel.Some reactions may alter the release rate of ^{133}Xe. However, in post-irradiation annealing tests on thoria-urania fuel (35% ThO_2, 65% UO_2) by Kim et al., it was found that the diffusion of ^{133}Xe in poly-crystaline $(Th,U)O_2$ was approximately an order of magnitude less than that of UO_2 [7].

Based on these measurements, an estimate of the fission gas diffusion coefficient has been implemented; its value is 1/10 that of FAST's current UO_2 coefficient [11].

2. Results and Discussion

2.1 Fission Gas Release

In order to determine if the model development for ceramic $(Th,U)O_2$ has been successful so far, the irradiation history and fuel composition data from [16] were used to see if the Percent

Fission Gas Release (%FGR) from the Post-Irradiation Examination (PIE) results of the DME-221 thoria fuel experiment could be replicated.

DME-221 is an experimental fuel irradiation test conducted at AECL-CRL. DME is short hand for "demountableelement"; this refers to how the experimental fuel is inserted into the U1 and U2 test loops at the National Research Universal (NRU) reactor. The experimental fuel is fabricated as elements that can be inserted into or removed (demounted) from the outer ring of a fuel bundle assembly. In the case of DME-221, the bundle has the same geometry as a 37-element CANDU fuel bundle, with the 18 elements in the outer ring available to house demountable elements, and the center element removed to allow the bundle to be irradiated on a fuel string in the NRU loops. Six variations of experimental fuel were fabricated for DME-221: 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) having two different pellet geometries, one set of pellets has a "standard" CANDU Length-to-Diameter (L/D) pellet ratio (~1.3) and the other set has a reduced L/D ratio (~0.7)[16].

It was decided to initially model the elements containing pellets with an L/D ratio of ~1.3. FAST has demonstrated good agreement with the modeling results of ELESTRES for the CANDU fuel design [5]. By limiting the initial analysis to the "standard" pellet design, changes made to pellet properties will be highlighted by results obtained from this new Th/U model.

Five of the twelve elements from DME-221 that have undergone PIE contain pellets with an L/D ratio of ~1.3. Table 1 displays the measured percentage of FGR results fromDME-221 fuel elements, compared to FAST predictionsforUO_2 and DME-221 fuel elements that experience the same power history. Results are presented based on the actual fuel power history(ThO_2, $(Th,U)O_2$ 1.0%, $(Th,U)O_2$ 1.5%) withthe dischargeburnup indicated in parentheses.

Table 1 Comparison of Percent Fission Gas Release Model Results to PIE Results

DME-221 Power History(Exit Burnup [MWh/kgHE])	**Modeled % FGR: UO_2 Fuel**	**Modeled % FGR: DME-221 Fuel**	**DME-221 Measured % FGR Range [16]**
ThO_2 (361)	1.2	0	0.05-0.11
ThO_2 (594)	11.8	0.2	0.05-0.11
$(Th,U)O_2$ 1.0% (499)	6.5	0	0.06-1.2
$(Th,U)O_2$ 1.5% (594)	11.0	0.2	0.08-2.8
$(Th,U)O_2$ 1.5% (929)	20.2	3.3	2.8

The modeled % FGR results forDME-221 fuel predict values that are close to the measurement range reported.This demonstrates that the model is producing realistic values for the % FGR. Note that a sensitivity analysis was not done to determine the uncertainty in the model, as the model is still under development. There is still a substantial amount of work that needs to be done on the model (see Section 3). In comparison to FAST predicted % FGR, the Th-based ceramic model is

also demonstrating significantly decreased fission gas release compared to UO_2 fuel, similar to what has been observed [16, 17].

Since fission gas release is dependent on fuel temperature, grain restructuring, and burnup, it can be viewed as an indicator of how well the model is predicting the overall irradiation behavior of the fuel. Although thecomparisons with available data are limited at this point in time, the model's ability to predict fission gas release is very encouraging.

2.2 Fuel Centerline Temperature

Th-based fuel is expected to have a lower temperature compared to UO_2 fuel irradiated in the same conditions [16]. The modeledDME-221 fuel provides an estimate for the fuel temperature throughout its irradiation, and provides a means to quantify how much of a temperature difference exists.

Figure 1 displays the declining power histories that the $(Th,U)O_2$ 1.5 wt.% ^{235}U fuel experiencedfor both "low" and "high" burnups[16]. Figure 2 plots the model-calculated centerline temperature of the $(Th,U)O_2$ with1.5% ^{235}Ufuel pellet and the FAST predicted results for a UO_2 fuel pellet enriched to the same ^{235}U content using the power history displayed in red from Figure 1.

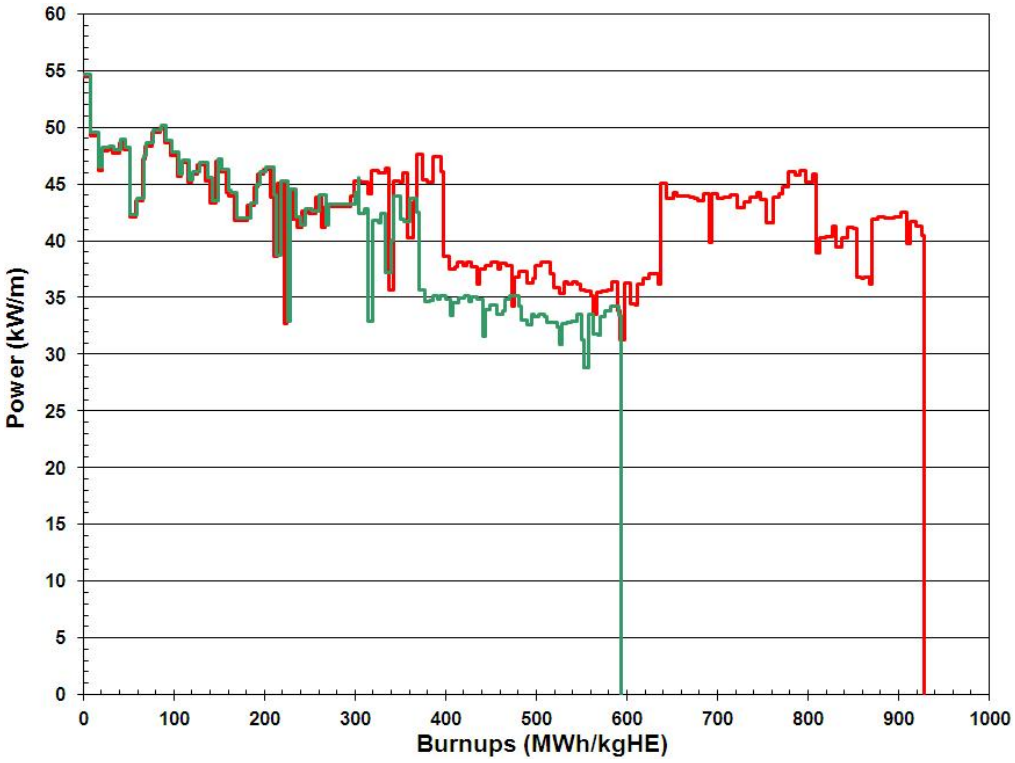

Figure 1Linear Power History of $(Th,U)O_2$ 1.5 wt.% ^{235}U Fueled Elements from [16]

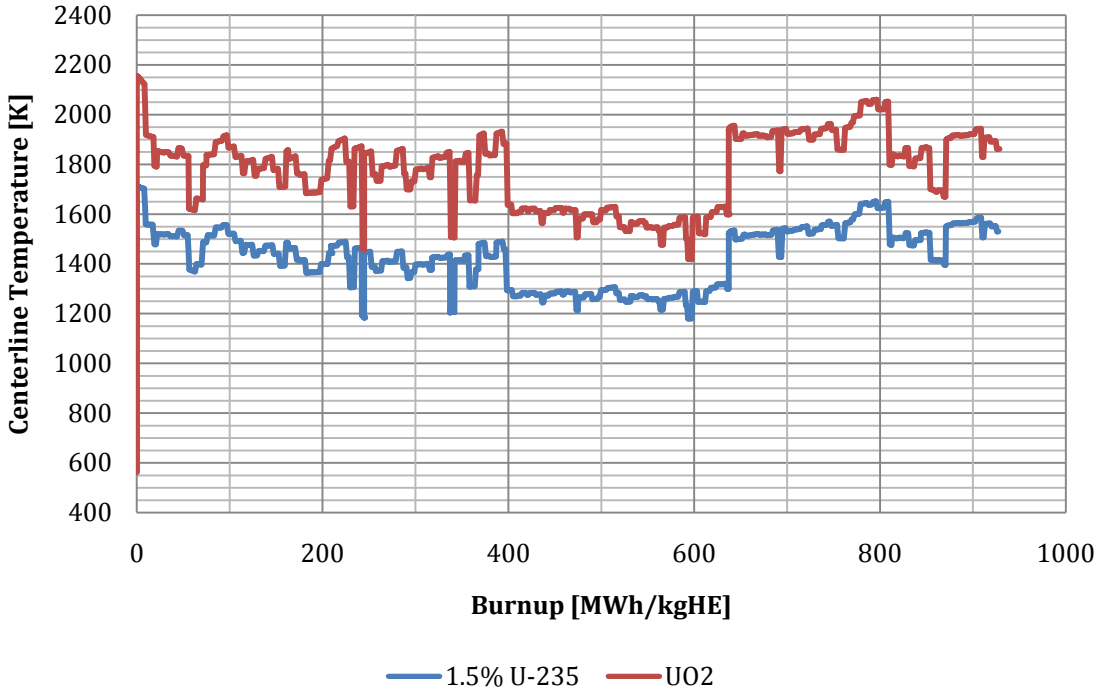

Figure 2 Comparison of DME-221 Element Fueled with (Th,U)O_2 1.5 wt.% ^{235}U and UO_2 Centerline Temperatures (high burnup power history)

Modeled centerline fuel pellet temperatures for each fuel type and corresponding power history, displayed the same response in temperature behaviour to changes in linear power. Table 2 presents the maximum modeled centerline temperature for the DME-221 fuel, UO_2 fuel (with the same power history) and the percent difference between the models.

Table 2 Comparison of Modeled Maximum Centerline Temperatures for DME-221 Fuel with UO_2 fuel

DME-221 Power History (Exit Burnup [MWh/kgHE])	Modeled Maximum Temperature: DME-221 Fuel (K)	Modeled Maximum Temperature: UO_2 Fuel (K)	Percent Difference
ThO_2 (361)	1149	1315	-13 %
ThO_2 (594)	1324	1700	-22 %
(Th,U)O_2 1.0% (499)	1253	1500	-16 %
(Th,U)O_2 1.5% (594)	1600	2008	-20 %
(Th,U)O_2 1.5% (929)	1711	2154	-21 %

Under the irradiation conditions of DME-221 fuel within the test loops at NRU, it is expected that the centerline temperatures of ThO_2 and (Th,U)O_2 fuel will be 13-22% cooler than that of UO_2 fuel. This means the overall centerline temperature is ~200-400 K lower during irradiation.

From the thermal conductivity correlations of $(Th,U)O_2$[14] and $(Th,Pu)O_2$ [18], it is expectedthat the thermal conductivity of $(Th,U)O_2$ is greater than $(Th,Pu)O_2$. Based on this, it is expected that the temperature of a $(Th,Pu)O_2$ pellet will be greater than a $(Th,U)O_2$ pellet, but less than UO_2.

Recalling equation (2) for the unirradiated thermal conductivity of $(Th,U)O_2$, the correlation is sensitive to the mole percent of U withinthe ceramic. With greater U content, there is a decreasein thermal conductivity. At this point in the model development, the mole percent remains constant throughout the irradiation. During irradiation,fertile ^{232}Th is converted into fissile ^{233}U,leading to variations in the U content within the fuel over the period of its irradiation. This will affect the thermal conductiviy, and currently is not accounted for. The significanceof any degradation in thermal conductivity is uncertain at this point, but will likely be dependent on the initial U content and neutron flux (power history).

3. Future Work

Work to be done on the development of an SCWR fuel model is substantial. A literature review is ongoing and may provide more physical properties of $(Th,Pu)O_2$. Modeling the remaining seven elements from DME-221 that have undergone PIE will provide further indication of the $(Th,U)O_2$ model'svalidity. With the framework for Th-based pellet behaviour established in this work, an initial $(Th,Pu)O_2$ model will be developed and compared to another experimental fuel irradiation conducted at AECL-CRL on $(Th,Pu)O_2$ fuel [19]. The activation energy used in the the grain growth model will need to be adjusted to reflect the wt% of UO_2 of the modeled fuel. There has been someindication while developing the $(Th,U)O_2$ model that greater attention to changes in U content during burnup is required.Some work is required to include a method to track the production of^{233}U from ^{232}Th. One of the current modeling assumptionsis that ^{233}U is the only fissile isotope that produces fission gas. While this assumption is valid for ThO_2 fuel, it does not account for the ^{235}U present in the other two fuel types. If the production of ^{233}U is calculated, the model will be able to determine if the source of fission gas is from^{233}U or from another fissile isotope.It would alsoallow the model to account for the difference in flux depression between UO_2 fuel and Th-based fuel that Long et al. demonstrate[8], and enable the thermal conductivity to be updated as the U content varies.

4. Conclusions

In aid of the development of a Canadian SCWR fuel perfomance model, an initial $(Th,U)O_2$ fuel pellet model has been developed. Selected irradiation histories from DME-221 fuel were modeled, and realistic values of the % FGR were predicted. A comparison is also made to the behaviour of FAST's UO_2 fuel model under the same irradiation conditions. Further work is required to produce a $(Th,Pu)O_2$ fuel model. Modeling the variation of Uin Th-based fuel during irradiation will be pursued.

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