PREDICTING THE BEHAVIOUR OF DEFECTED CANDU FUEL OXIDATION IN AIR WITH THE USA DOE SEMI-EMPIRICAL MODEL FOR THE OXIDATION OF DEFECTED LWR FUEL

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ABSTRACT – The USA Department of Energy (DOE) semi-empirical model for the oxidation of defective LWR fuel in air was extended to CANDU fuel. Model results were consistent with available experimental test data for irradiated CANDU fuel in the temperature range 200-400 ^{0}C.

For the only available data at lower temperatures (150^{0}C) from the AECL Controlled Experiment (CEX-1, 1980-1997), the model predicted full conversion to U_3O_7/U_4O_9 in 14.8 years in agreement with the experimental value of 15.5 years. The model also indicated that when terminated in 1997, the CEX-1 experiment was midway to cladding breaching due to oxidation to U_3O_8.

Introduction

A semi-empirical model was developed by the USA Department of Energy (DOE) for predicting breachment of the fuel cladding due to the oxidation of defective PWR and BWR fuel stored in air [1]. A similar approach was developed by [2] and [3]. The model is based on Canadian and USA databases and opens the possibility of its application to CANDU fuel, particularly, at the lower temperatures of storage of CANDU fuel for which no predictive formulations for whole fuel elements exist.

The DOE model has been reviewed by the USA Nuclear Regulatory Commission (NRC) and addressed in an interim guideline to deal with the "potential rod splitting due to exposure to an oxidizing atmosphere" of defected fuel during storage and transportation [4]. Through "Interim Staff Guidance-22", the NRC reviews the use of the DOE model in safety assessments as an alternative option to assess the potential for defective fuel oxidation and its consequences. Further, the model was applied to predict the behaviour of LWR fuel for long term storage at the once proposed Yucca Mountain repository [5]. Application of the DOE model to CANDU fuel to test its potential to predict the behaviour of defective CANDU fuel for dry storage and in the used fuel containers for a Deep Geological Repository is of particular importance.

1. The DOE Model for Predicting Defective Fuel Oxidation

The DOE model is semi-empirical and is based on the experimental evidence that the oxidation of LWR fuel takes place in two stages: (1) oxidation of UO_2 to U_4O_9 and (2) oxidation of U_4O_9 to U_3O_8. The CANDU fuel oxidizes to an intermediate U_3O_7/U_4O_{9+x} and then to U_3O_8, although

there is some doubt about the formation of U_3O_7. However, the activation energies for conversion of the

UO_2 to U_3O_7 or U_4O_9 are nearly identical. Hence, the DOE model approach should be applicable to CANDU fuel. In fact, based on experimental evidence, the two oxidation mechanisms included in the DOE model are nearly identical to the Canadian experimental evidence. For instance, the activation energies for the observed stages of the oxidation of UO_2 to U_3O_7/U_4O_{9+x} are nearly identical – 106 kJ/mol against −107 kJ/mol [1]. Similarly, the activation energy for conversion of U_3O_7/U_4O_{9+x} to U_3O_8 is well represented by a value of 150 kJ/mol [5].

On U_3O_8 formation a volumetric expansion of the pellet takes place leading to the eventual fracturing of the cladding. The mode of unzipping the cladding is usually longitudinal along the length of the fuel element but can also be spiralling or simply circumferential around the clad at the location of the defect. The end result is that the pellets contents are spilled out of the cladding as fine grains. Volatile fission products such as krypton, xenon and iodine are released, as well as, the semi-volatiles like cesium and cadmium become mobile.

The two oxidation stages are modelled with Arrhenius equations where the time to completion of the oxidation stage is logarithmically linear as a function of 1/T. The Arrhenius equation parameters are derived from the available database by a best fit line. The model also accounts for the fuel burnup in the second stage oxidation to U_3O_8. The fuel element physical characteristics are also taken into account including the pre-irradiation fuel/clad gap or "cold" gap and the fuel pellet radius. The strain required to split the cladding is also included. For CANDU fuel, a cladding strain of 2% was chosen to result in the breach of the fuel cladding [6].

The time to oxidize the fuel to the first stage is approximated by the following relation:

$$t_{2.4} = k_{2.4}\, e^{\frac{Q_{2.4}}{RT}} \qquad \text{..... (1)}$$

where:

$t_{2.4}$ is time in hours, where the subscript 2.4 is the O/M hyper-stoichiometry ratio for the transformation of UO_2 to U_4O_{9+x} rather than the 2.25 ratio of U_4O_9.

$k_{2.4}$= 1.40 x 10^{-8} hr

$Q_{2.4}$= 105 kJ/mol, and R= 8.314 J/mol/K

Oxidation of the fuel to the second stage of U_3O_8 oxidation and fuel cladding splitting is additive to the time taken to oxidize the fuel to U_4O_9. The relation for the second stage is:

$$t_{7.5} = \lambda_{7.5}\, k_{7.5} e^{\frac{(Q_{7.5}+\propto B)}{RT}} \qquad \text{..... (2)}$$

where:

$t_{7.5}$ is the incubation time (hours or years) for stage 2: $UO_{2.4} \rightarrow U_3O_8$ which is also hyper-stoichiometric at an O/M ratio of 2.75, the subscript is a short form of the decimal part of this ratio for ease of distinguishing it from the first stage oxidation.

$\lambda_{7.5}$ is a correction factor that takes into account the fuel dimensional characteristics (also shown below).

$k_{7.5}$ empirically derived coefficient (hr) with a value of 4.84 x 10^{-14}

$Q_{7.5}$ is the activation energy determined to be 150,000 (J/mol)

α is a constant used for the effect of the fuel burnup on the oxidation of $UO_{2.4}$ to U_3O_8 and set to 1,000 (J/mol)/ (GWd/MTU).

B is the fuel burnup in GWd/MTU

R is the Universal Gas constant as 8.314 J/mol/K

As indicated above, $\lambda_{7.5}$ is a correction factor that takes into account key parameters of the fuel dimensions and the cladding as well as the strain required to split the cladding [5]. For used CANDU fuel, the strain is 2% as per [6]. This correction factor is defined as follows [5]:

$$\lambda_{7.5} = 1 - \frac{r_{11}}{r_{01}} \quad \text{..... (3)}$$

and

$$\frac{r_{11}}{r_{01}} = \left[\frac{(1+x)^3(1+s)^3 - z_1 z_2}{z_1 - z_1 z_2}\right]^{\frac{1}{3}} \quad \text{..... (4)}$$

where

x is the ratio of the cold fuel pellet gap to the fuel pellet radius

s is the strain necessary to split the cladding. For CANDU fuel a value of 2% is recommended.

z_1 is the ratio of the UO_2 volume to $UO_{2.4}$ volume (being equal to 0.9929)

z_2 is the ratio of U_3O_8 volume to $UO_{2.4}$ volume (being equal to 1.3807)

Once the expansion of the fuel pellets reaches the cladding, fracturing of the cladding at the defect such as a pinhole or hairline crack follows. The time that takes the fuel pellets to oxidize and breach the cladding is the incubation time or the time to breach the cladding. Upon fracture, the initiating crack continues to grow along the whole length of the fuel element. The time for the cladding to slit open along its length is known as the unzipping time and is calculated for CANDU fuel by an empirical correlation based on Canadian tests on CANDU Bruce fuel at temperatures higher than 220°C [7a]:

$$V = 1.31 \times 10^4 e^{\frac{Q}{RT}} \quad \text{..... (5)}$$

where V is the velocity of the crack breaching the cladding in cm/min. Q is the activation energy of the clad crack given by a value of – 81.5 kilojoules per mol. R is the Universal Gas constant as 8.314 J/mol/K and T is the fuel temperature in degrees Kelvin.

This correlation is similar in form to the DOE model equation and based on an Arrhenius type. Therefore, it should allow reliable predictions at the lower temperatures since the mechanism of oxidation to U_3O_8 leading to cladding breachment is identical at the lower temperatures.

2. Application of the DOE Model to CANDU Fuel

Data generated from application of the DOE model to the Pickering and Bruce type fuel bundles is presented. The bundles physical characteristics are given in Table 1 and were applied to equations (2) and (4) of the previous section as follows:

Table 1: Typical Fuel Bundle Parameters to Use in DOE Correlation

Physical Parameters	Pickering 28-Element Fuel	Bruce-37 Element Fuel
Cladding OD (mm)	15.218	13.07
Cladding ID (mm)	14.38	12.23(*)
Cladding Wall Thickness (mm)	0.419	0.419
Fuel Pellet OD (mm)	14.25	12.141
Fuel/clad gap (mm)	0.065	0.0455

OD= Outside diameter; ID= Inside diameter; (*) value derived from Clad OD

1. The formula describing the time for stage 1 oxidation of the UO_2 to U_3O_7/U_4O_{9+x} for CANDU fuel is applicable to any CANDU fuel type because the DOE correlation is independent of the fuel physical characteristics. In essence, the fuel pellet densifies and decreases in volume, by assuming the oxidation occurs uniformly across the pellet, the oxidation becomes independent of the fuel physical dimensions. It is given below as:

$$t_{2.4} = 1.40 \times 10^{-8}\ e^{\frac{12\,629}{T}} \text{ in hours} \qquad .....(6)$$

where $t_{2.4}$ is the time required to convert UO_2 to U_4O_9 but also assumed to describe conversion to U_3O_7/U_4O_{9+x}.

2. The second stage (stage 2) of the UO_2 oxidation to U_3O_8 takes into account the physical characteristics of the fuel elements as given in Table 1. The gap width is based on the measure of the gap dimension before the fuel is irradiated or "cold" gap dimension. As a result of the different fuel element dimensions between the Bruce and Pickering bundles, an equation for the oxidation of the fuel in the second stage exists for each fuel bundle type. Douglas Point fuel is well represented by Pickering fuel since the fuel/clad gap and the fuel pellet radius are identical.

   Further, since conversion is also influenced by burnup, different equations are also obtained for each fuel bundle type depending on its burnup. Three different burnups have been considered: CANDU fuel of average burnup equal to 8.3 GWd/MTU, higher burnup of 10 GWd/MTU and no burnup (0 GWd/MTU). The intention was to compare the significance of burnup on the required incubation time. The equation has the form given in equation (2). After introducing the values for the various parameters such as: $\lambda_{7.5}$, $k_{7.5}$, Q and R, the equation is simplified to the form:

$$t_{7.5} = k\ e^{\frac{B}{T}} \text{ in hours.....} \quad (7)$$

where T is temperature of oxidation in degrees Kelvin and k is given by the product of $\lambda_{7.5}$ and $k_{7.5}$. Values for $\lambda_{7.5}$ and $k_{7.5}$ are shown in Table 2. Based on these values, the values for k are

calculated for equation (7). The B values are obtained from the bundle burnup as per equation (2) in section 2.0. Both the k and B values are shown in Table 3 for Bruce and Pickering fuel.

Table 2: Values of $\lambda_{7.5}$ and $k_{7.5}$ used to calculate the coefficient k in Equation (7)

Bruce Fuel		**Pickering Fuel**	
$\lambda_{7.5}$	$k_{7.5}$	$\lambda_{7.5}$	$k_{7.5}$
0.0892	4.84E-14	0.0948	4.84E-14

Table 3: Parameters for Equation (7) for Bruce and Pickering Fuel of various burnups

Burnup (GWd/MTU)	**Bruce Fuel**		**Pickering Fuel**	
	Equation (7) Parameters		Equation (7) Parameters	
	k	**B**	**k**	**B**
0	4.32E-15	18042	4.59E-15	18042
8.3	4.32E-15	19040	4.59E-15	19040
10	4.32E-15	19245	4.59E-15	19245

3. Data Results and Verification of the DOE Model for CANDU Fuel

Estimated values for oxidation incubation times using the DOE model for CANDU fuel are compared against CANDU fuel experimental data from full fuel elements in Table 4. The data are from [6], [7a], [7b] and [8], and cover oxidation tests above 200°C. Fuel bundles from Bruce, Pickering and Douglas Point are included at various burnups.

Table 4: Comparison of Incubation Times from Tests and DOE Model Predictions

Fuel Type/[Ref.]	**Burnup GWd/MTU/ (MWh/kgU)**	**Temperature (°C)**	**Experimental Results (t_{inc}) (hrs)**	**Model Time (hrs)**		
				$t_{2.4}$	$t_{7.5}$	t_{inc}(**)
Bruce /[6]	9.2/(220)	400	<24	2.0	0.01	2.0
Pickering/[8]	Unirradiated	300	4 and 7	52	0.2	52
Pickering /[7a]	Unirradiated	250	>942	430	4.4	434
Douglas Point/[6]	6.7/(160)	250	173.5	430	21	451
Pickering/[7a],[7b],[6]	7.9/(190)	250	118 - 208 (*)	430	27	457
Bruce/[7b]	7.1/(170)	230	~601	1122	89	1211
Pickering/[6]	7.9/(190)	230	500	1122	114	1236
Bruce/([6]	9.2/(220)	200	>>700	5517	1647	7164

(*) Indicates variability in the test time to breachment for nominally similar fuel elements

(**) The incubation time is the sum of the oxidation times at stage one ($t_{2.4}$) plus stage two ($t_{7.5}$)

The Douglas Point fuel elements are nearly identical to the Pickering bundle with identical cold clad/fuel gap and fuel pellet radius. As a result, the estimates from the DOE Model for both fuel bundle types are the same.

The data from the tests on fuel elements in Table 4 indicate that oxidation of the fuel above 200°C is rapid. This leads to incubation times which are of the order of a few hours to half a year at most. Except for the CEX-1 experiments at 150°C, there is no other work that tested full fuel elements at temperatures below 200°C.

The data also show that the estimates provided by the DOE Model for CANDU fuel can be favourably compared with experimental results. The estimated values are for the most part overestimating the experimentally obtained incubation times for breaching the cladding by 50-100% in the temperature range 230-300°C. This variation is well within the variation in the DOE model for this temperature range. Factors contributing to the difference between the model and the experimental data include: the heterogeneity of the fuel post-irradiation due to the fuel power history, accuracy of the measured fuel cladding breach time, lack of measurements for the fuel clad/ gap prior to irradiation for the fuel tested, and precise determination of the strain of the fuel cladding leading to fuel cladding breach.

Also, the DOE model considers that oxidation occurs uniformly across the fuel pellet for simplicity. However, this is not the case due to the variation of the irradiation flux and radial temperature profile of the pellets while in-reactor. In fact, for CANDU fuel as well as LWR fuel, the inner core of the pellets oxidizes more rapidly than the intermediate zone of the pellet. Additionally, fuel pellets are also cracked due to the temperature gradient while in-reactor, thus producing another level of heterogeneity. Swelling in the inner core of the fuel pellet ahead of the periphery might lead to an earlier breach than predicted. This might be, particularly true, at temperatures higher than 200°C when the oxidation to U_3O_8 is quite rapid. In another words, these factors due to heterogeneity are likely to play a greater role influencing the time to breach the clad at the higher temperatures when oxidation is fast than at the much smaller oxidation rates below 200°C.

To verify values below 250°C, the Taylor and McEachern correlation [9] was used. This correlation is based on the oxidation of UO_2 discs of unirradiated sintered fuel to U_3O_8 by observing the extent of conversion to U_3O_8 on the surface of the discs by X-ray diffraction thus avoiding interference from the reaction of UO_2 to U_3O_7/ U_4O_{9+x}. One drawback of using this correlation is that the results are for open UO_2 discs and cannot include the additional barrier to mass transport resulting from the fuel pellets being enclosed in the fuel cladding. Therefore, it is expected that this correlation will overestimate the time of incubation. However, it should allow for an approximate estimate of the incubation times at temperatures below 200°C [9].

The calculations are based on the criterion that defective CANDU fuel oxidation splits the cladding when there is a 15% conversion to U_3O_8 from UO_2 which corresponds to a diametral strain of 2% (see Hastings et al., 1984). The correlation is defined by the following equations:

$$\alpha(t) = 1 - \exp(-\pi\kappa t^3 / 3) \quad \text{..... (8)}$$

$$\ln \kappa = \frac{(-52808 \pm 3442)}{T} + 86.165 \quad \text{..... (9)}$$

where α is the extent of oxidation of UO_2 to U_3O_8, κ is a rate constant that includes both nucleation and growth rate constants in (hr^{-3}), T is temperature in degrees Kelvin and *t* is time in hours.

Since the experimental data is for unirradiated fuel, the data was compared against predictions of the DOE model for fuel with zero burnup for both CANDU fuel and LWR fuel (Table 5). Mindful that the DOE model is based on irradiated fuel which behaves differently than unirradiated fuel, the comparison indicates that the predicted values are reasonable and provide a good estimate of the incubation times. In fact, the correction for the fuel burnup used in the DOE model works well and it appears to be supported by the experimental evidence.

Further, for temperatures below 150°C, the DOE model values for zero burnup fuel are significantly smaller than those predicted by the Taylor and McEachern correlation suggesting that the incubation times predicted by the DOE model are conservative. At 150°C, the difference is a factor of 1.5 but at 100°C is a factor of 9. Considering that the unirradiated tests represented by the Taylor and McEachern correlation do not take into account the impact of the fuel cladding restricting the diffusion of air to the pellets enclosed in the fuel element, the DOE model predictions are truly conservative and consistent with the finding that irradiated CANDU fuel oxidizes faster than unirradiated fuel [6]. A double check with the LWR data [5] included in Table 5 shows that the CANDU fuel incubation times are about 30-50% greater than those for LWR fuel. This is consistent with the differences between the calculations for LWR and CANDU fuel. In the case of LWR, the incubation times are based on 0% strain and the fuel gap equals zero. The CANDU calculations are based on a cladding strain of 2%.

Table 5: Comparison of Incubation Times for Zero Burnup CANDU Fuel and LWR Fuel Using the DOE Model against the Correlation of Taylor and McEachern [9]

	Incubation Times (Years)		
Temperature (°C)	**Bruce Fuel Zero burnup**	**Unirradiated (*)**	**LWR Fuel Zero burnup**
25	1.0×10^{8}	9.28×10^{8}	-
50	1.0×10^{6}	9.6×10^{6}	-
75	2.54×10^{4}	1.9×10^{5}	1.04×10^{4}
100	1.31×10^{3}	6.45×10^{3}	8.44×10^{2}
150	16	24.4	14.9
200	0.64	0.30	0.631
250	4.9×10^{-2}	8.54×10^{-3}	4.91×10^{-2}

(*) Values calculated with the Taylor and McEachern Correlation [9]

The incubation times to breach the cladding of Bruce and Pickering fuel bundles obtained from the DOE model are shown in Table 6 for Bruce and Pickering fuel, respectively as a function of temperature. Also, times to fully unzip the fuel elements along the length of the fuel element

based on the Novak correlation [7a] are included. The calculations assumed the crack initiates at the mid-point of the fuel element. Since the lengths of CANDU fuel elements are identical, cladding unzipping times are applicable to other CANDU fuel types. The results show that the cladding unzipping times are large below 150°C and provide significant additional time before the oxidized pellets are spilled from the fuel element.

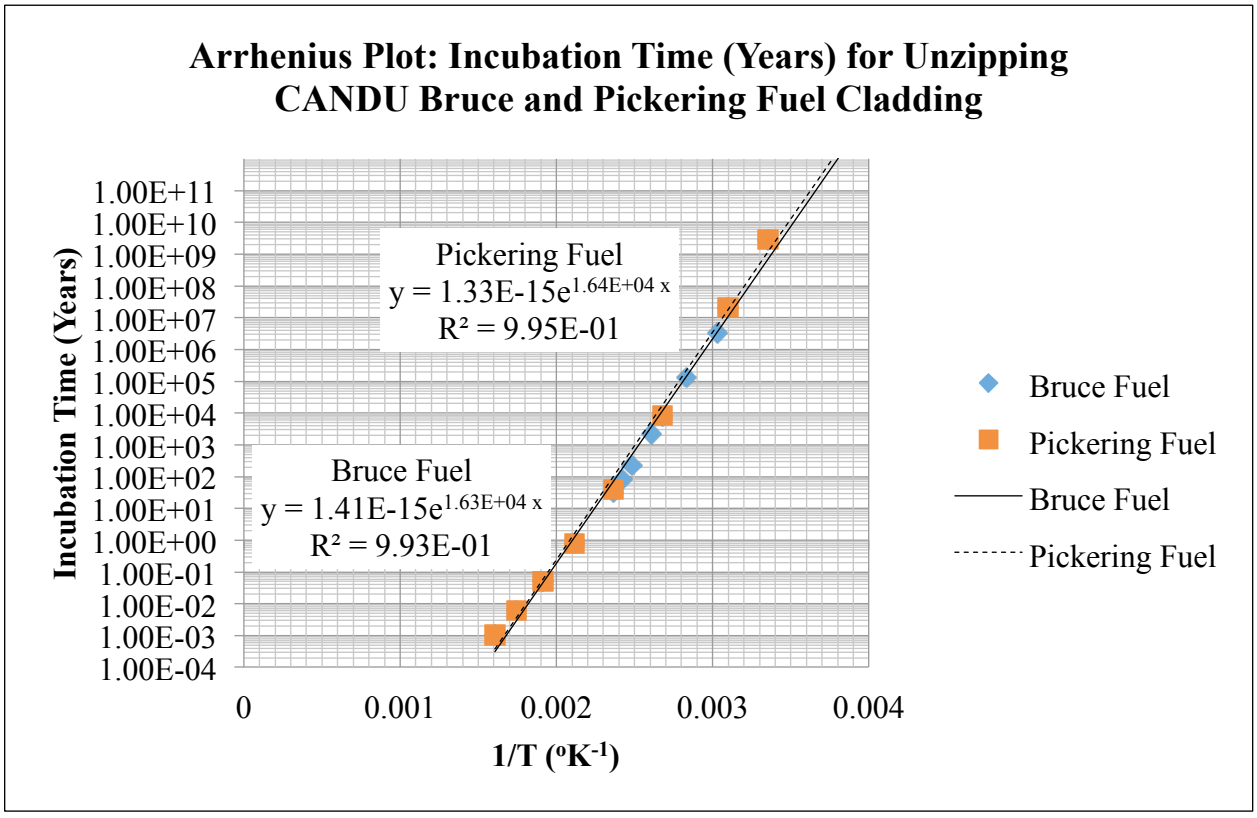
Table 6: Incubation Time to Cladding Breachment and Cladding Unzipping Times for Defected Irradiated CANDU Bruce Fuel and Pickering Fuel of Average Burnup (8.3 GWd/MTU) as a Function of Temperature in Ambient Air

	Bruce Fuel			Pickering Fuel	Bruce Fuel
Temperature (°C)	**Stage 1 to U_4O_9 $t_{2.4}$ (Years)**	**Stage 2 to U_3O_8 $t_{7.5}$ (Years)**	**Cladding Breachment Time (Years)**	**Cladding Breachment Time (Years)**	**Cladding Unzipping Time (Years)**
25	4.1×10^{6}	2.8×10^{9}	2.8×10^{9}	2.9×10^{9}	6.7×10^{5}
50	1.5×10^{5}	2.0×10^{7}	2.0×10^{7}	2.1×10^{7}	5.4×10^{4}
100	809	7284	8.1×10^{3}	8.5×10^{3}	8.9×10^{2}
120	144	542	686	719	238
130	65.1	163	228	238	128
140	30.5	51.9	82	85.5	71
150	14.8	17.4	32	33	40
160	7.4	6.1	13.5	14.0	24
200	0.62	0.15	0.77	0.78	3.5
250	0.049	3.2×10^{-3}	0.052	5.2×10^{-2}	0.050
300	6.0×10^{-3}	1.3×10^{-4}	6.1×10^{-3}	6.14×10^{-3}	9.4×10^{-2}

A significant finding in Table 6 is the prediction that the conversion of UO_2 to U_4O_9 is complete at an incubation time of 14.8 years at 150°C. This value is consistent with the CEX-1 observation that at 15.5 years the conversion of the UO_2 to U_4O_9 was complete or nearly complete for the last fuel examined. It is also for the first time that the experimentally found value at the lower temperature of 150°C has been scientifically estimated within an error of about 4.5%.

The results also predict that breachment of the fuel cladding of defected fuel at the constant temperature of 150°C, as used in the Controlled Experiment at AECL, would be expected to occur at about 32 years of storage. This suggests that the AECL experiment which was terminated after about 15 years, was ended about mid-way to fuel cladding breachment.

The data in Table 6 were plotted in an Arrhenius type plot as shown in Figure 1 and yielded a linear plot with a best fit line that accurately represents the results of the DOE Model in the temperature range of 25-350°C. Figure 1 also illustrates that the difference between defective



Bruce and Pickering fuel incubation times is insignificant.

Figure 1: Incubation Time (years) as a Function Temperature (degrees Kelvin) for Unzipping the Clad of Defected CANDU Bruce and Pickering Fuel

Table 7: Correlations for Incubation Times (Years) as a Function of Temperature (°K) for Defected Used CANDU Bruce and Pickering Fuel of Average Burnup (8.3 GWd/MTU)

Temperature (°C)	Bruce Fuel	Pickering Fuel
25-350	$t_{inc} = 1.41 \times 10^{-15}\, e^{\frac{16300}{T}}$	$t_{inc} = 1.33 \times 10^{-15}\, e^{\frac{16400}{T}}$
25-150	$t_{inc} = 4.016 \times 10^{-18}\, e^{\frac{18330}{T}}$	$t_{inc} = 4.514 \times 10^{-18}\, e^{\frac{18360}{T}}$
150-300	$t_{inc} = 8.84 \times 10^{-13}\, e^{\frac{13000}{T}}$	$t_{inc} = 8.30 \times 10^{-13}\, e^{\frac{13000}{T}}$

where T is in degrees Kelvin and t_{inc} is incubation time in years.

Table 6 also shows that the second stage oxidation to U_3O_8 from U_4O_9 is faster than the first stage from UO_2 to U_4O_9 above 150°C to the extent that the first stage to U_4O_9 becomes the dominant component of the incubation time. Conversely, below 150°C, the oxidation to U_3O_8 from U_4O_9 dominates the time it takes to breach the fuel cladding. As a result, correlations with greater accuracy than the correlations shown in Figure 1 are achieved when dividing the data into two ranges, below and above 150°C. These correlations are documented in Table 7 for Bruce and Pickering fuel with variances above 0.99.

Finally, the data in Table 6 suggest that obtaining further experimental values from defective fuel elements below 220°C could be done in a relatively short time of a few years in the temperature range 170-220°C.

Table 8: Effect of Burnup on Incubation Times (Years) for CANDU Bruce Fuel

Temperature (°C)	Zero burnup	Burnup 8.3 GWd/MTU	Burnup 10 GWd/MTU
	Incubation Time (years)		
25	1.0×10^8	2.8×10^9	5.5×10^9
50	1.0×10^6	2.0×10^7	3.7×10^7
100	1.31×10^3	8.1×10^3	1.3×10^4
150	16	32	43
200	0.64	0.77	0.86
250	-	0.050	-

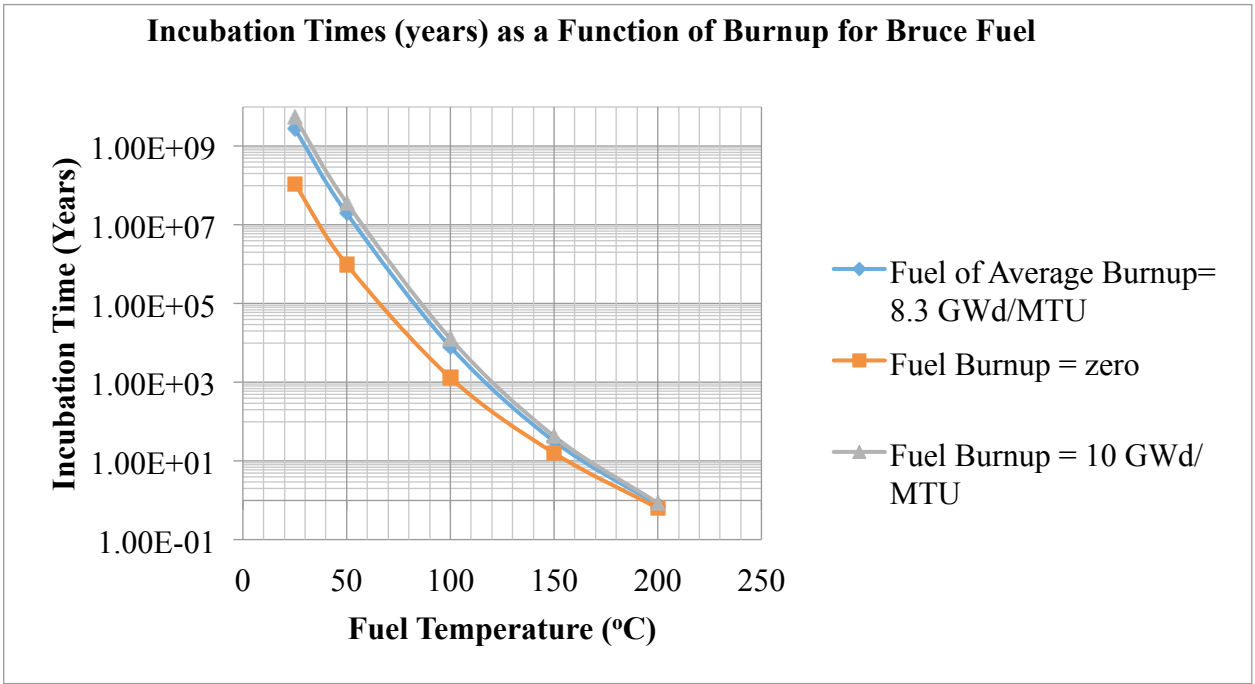


Figure 2: Effect of Burnup on time to Breach Defective CANDU Fuel

The influence of burnup on CANDU fuel is documented in Table 8. Incubation times for Bruce fuel burnups of zero and 10 GWd/MTU were calculated from the DOE Model and plotted as data points in Figure 2. The results indicate significantly shorter incubation times for fuel of zero burnup. This is consistent with evidence that fission products such as Gadolinium have been observed to influence and decrease the rate of oxidation to U_3O_8 [10]. Note that while the fuel is zero burnup, it is, however, irradiated. For burnup values greater than the CANDU fuel average burnup such as 10 GWd/MTU, the increase in the incubation time is minor. All three fuels incubation times asymptotically reached the same common value for the incubation time at 200°C. This is because the oxidation time in the first stage of the UO_2 to U_4O_9 or U_3O_7 becomes the dominant mechanism as the temperature increases to 200°C and is independent of burnup in the DOE model. The impact of burnup becomes increasingly significant as the burnup increases into the domain of LWR fuel with averages burnups of 40 GWd/MTU.

4. CONCLUSIONS AND RECOMMENDATIONS

The USA Department of Energy (DOE) semi-empirical model that predicts the oxidation of defective LWR fuel in air to fuel cladding breaching was extended to defective CANDU fuel. Model predictions from the model were consistent with available experimental test data for irradiated CANDU fuel in the temperature range 200-400°C. For the only available data at lower temperatures (150°C) from the AECL Controlled Experiment (CEX-1, 1980-1997), the model predicted full conversion to U_3O_7/U_4O_9 in 14.8 years in agreement with the experimental value of 15.5 years. The model also indicated that when terminated in 1996, the CEX-1 experiment was about midway to cladding breaching due to oxidation of the pellets to U_3O_8.

Therefore, the extension of the USA DOE semi-empirical model to CANDU fuel for oxidation of defected fuel elements appears to be supported by the data available from the AECL experiments in the range 150°C to 400°C. The results presented confirm the rationale as to why the USA DOE model can be extended to CANDU fuel and offer proof that it predicts well actual experimental values, particularly, below 200°C where the data is difficult to obtain because of the very long times needed to oxidize the pellets to U_3O_8 which is of the order of decades at 150°C and much longer below this temperature. Temperatures below 150°C are the temperatures relevant to CANDU fuel storage systems and the used fuel containers.

The results from the DOE Model were used to develop correlations that predict the incubation time for fuel cladding breaching for commercial defected CANDU fuel elements of average burnup (8.3 GWd/MTU) in the temperature range 25-350°C. These correlations simplify the calculations from the DOE Model and are useful in assessing the behaviour of defective fuel in dry storage systems, transportation and long term storage in disposal containers in a Deep Geological Repository.

In view of the results, it is observed that incubation times are predicted to be relatively short for the temperature range 170-220°C where there is no experimental data available. Obtaining data in this temperature range will be important both for refining the model and extending the database in the area relevant to CANDU fuel.

5. References

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