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THORIA IRRADIATION AND POST-IRRADIATION EXAMINATION EXPERIENCE AT AECL

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ABSTRACT –Thorium is envisioned to play a significant role in future global nuclear fuel cycles. Thoria (ThO_2) based fuels are under consideration for deployment in the Canadian-designed pressurized heavy water reactor and super-critical water reactor. AECL's experience in developing thoria fuels is a key enabler to the Canadian nuclear industry availing itself of thorium as a sustainable source of nuclear energy. This paper summarizes 50 years of AECL experience with thoria irradiation tests and performance assessments from ~1962 to the present. The paper reviews irradiation tests conducted in two experimental reactors, and the Nuclear Power Demonstration pressurized heavy water reactor.

Introduction

Various thoria-based fuels have been studied by AECL including natural thoria (ThO_2), $(Th,U)O_2$, and $(Th,Pu)O_2$, with ^{235}U (in HEU) or Pu content ranging from 1-4 wt.% [1]. Fuel burnups up to 1130 MWh/kgHE (47 GWd/tHE) have been achieved, with linear powers up to 73 kW/m. Tests that studied defected fuel, power ramp behaviour, high burnups, different pellet fabrication methodologies, and pellet microstructure investigations are summarized. Key findings and lessons learned from irradiation tests and post-irradiation examinations are described.

Experience is contrasted with that of CANDU UO_2 fuel. Specific fuel performance attributes are discussed, including fission gas release, pellet microstructure behaviour, sheath strain/corrosion/hydriding, and defected fuel performance. Irradiation tests that are in progress (targeting burnups up to 1500 MWh/kgHE (63 GWd/tHE)) are described, as well as envisioned initiatives that will provide additional thoria fuel science and technology to Canada and the world.

This paper focuses on AECL experience with ThO_2-based fuel irradiations in the NRU (National Research Universal), NRX (National Research eXperimental) and NPD (Nuclear Power Demonstration) reactors. Physics experiments in the ZED-2 (Zero Energy Deuterium) critical facility using a variety of thoria based fuels (including ^{233}U experiments) are not discussed [2]. AECL also has experience with irradiating thoria fuels in the WR-1 organic-cooled reactor (high temperature, low pressure coolant [3]), LWR-type thoria fuel rods in NRX [4], and CANDU geometry fuels in the NRU moderator (low temperature and low pressure coolant) [5]. Irradiations of Th-based alternate fuel fabrication technologies (i.e., sol-gel, spherepac, extruded fuel, and annular fuel) have also been conducted by AECL; only solid ceramic pellet ThO_2-based fuels fabricated from powders are described in this paper.

1. Fission Gas Release (FGR)

Fission gas release (FGR) is a key parameter of fuel performance, particularly at high power (i.e., high temperature) and extended burnup [6]. Inert and volatile fission gases are produced in the fuel matrix as a result of fission, and a fraction of this gas diffuses to the element free volume. High FGR may result in element internal gas pressures exceeding that of the coolant, leading to sufficient stress on the fuel cladding to cause fuel failure by stress corrosion cracking (SCC).

Based on its superior thermal conductivity (relative to UO_2), thoria-based fuels are expected to operate at lower temperatures compared to UO_2 operating at the same power. Thoria is also a more refractory material than UO_2; since FGR is primarily a thermally activated diffusion process, it is expected that thoria-based fuels should experience reduced FGR compared to UO_2. AECL experience with thoria-based fuels has shown their FGR to be reduced, or bounded by those observed from similarly operated UO_2, depending on the microstructure of the as-fabricated fuel. Smith et al. [7] performed an instrumented $(Th,U)O_2$ irradiation experiment that showed thoria-based fuel with as-fabricated granules (resulting from pre-pressing and granulating stages) operating at higher temperatures than fuel with a homogeneous, dense microstructure; this behaviour was observed in several experiments as shown in Figure 1 and Figure 2. Figure 1 and Figure 2 are plots of identical FGR data from UO_2 [6] and various thoria-based fuels plotted against their element power (Figure 1) and burnup (Figure 2) (primary dependence on power, with a secondary dependence on high (> 900 MWh/kgHE, 38 GWd/tHE) burnup).

From Figure 1 and Figure 2, it is apparent that granular thoria-based fuels have similar FGR characteristics to conventional UO_2 based fuels, while irradiations (DME-221 [8]) with homogeneous pellet microstructures and standard (5- 10 μm) as-fabricated grain size have shown superior performance. In the case of non-granular BDL-422 fuel (1.5 wt% Pu in $(Th,Pu)O_2$ [9]), the FGR was low (< 5 %) below 1000 MWh/kgHE (42 GWd/tHE), and high (> 23 %) above 1000 MWh/kgHE (42 GWd/tHE) [10]. For the high burnup elements in BDL-422, the power and burnup combination (54 – 73 kW/m to 1181 – 1082 MWh/kgHE, respectively) is above that of AECL experience with UO_2, and the small initial grain size (3 – 4 μm versus 5 – 10 μm in production CANDU fuel) may also have contributed to the high FGR [11]. For fuels operating at < 30 kW/m, it appears that FGR remains low regardless of fuel composition or microstructure (e.g., granular NPD-51 thoria fuel was irradiated in the Nuclear Power Demonstration reactor at < 30 kW/m up to 1130 MWh/kgHE (47 GWd/tHE) with only 0.5 % FGR).

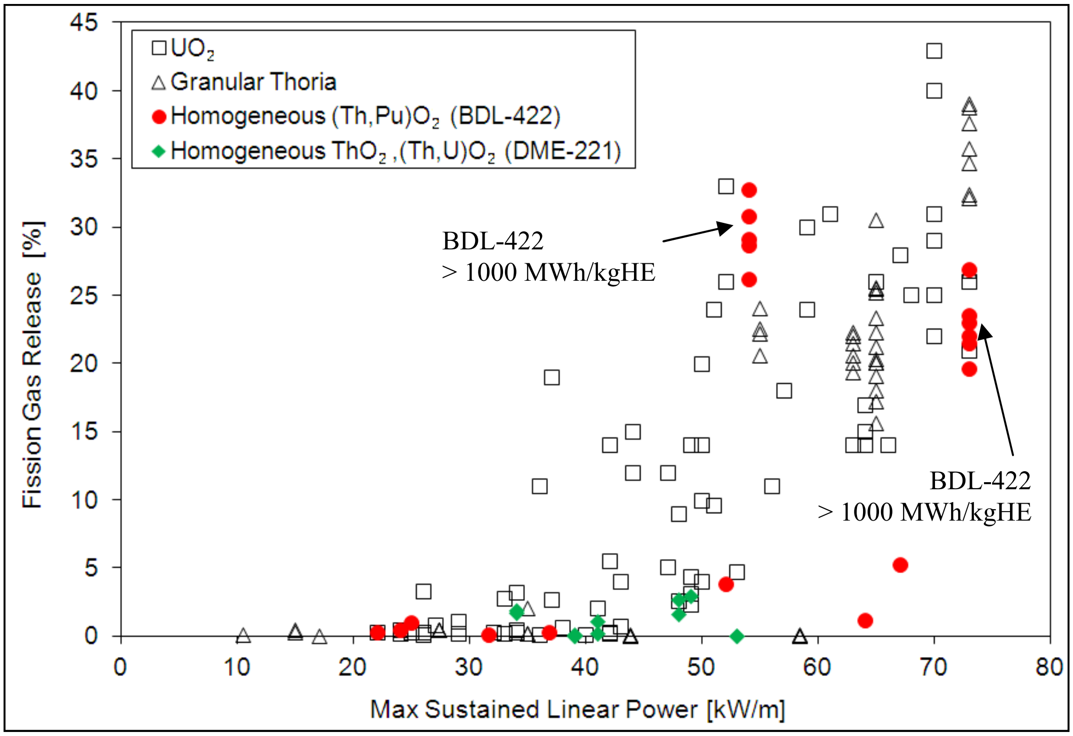

Figure 1: FGR [%] as a Function of Maximum Sustained Linear Power [kW/m]. The BDL-422 $(Th,Pu)O_2$ with FGR > 15% have Burnups > 1000 MWh/kgHE (42 GWd/tHE).

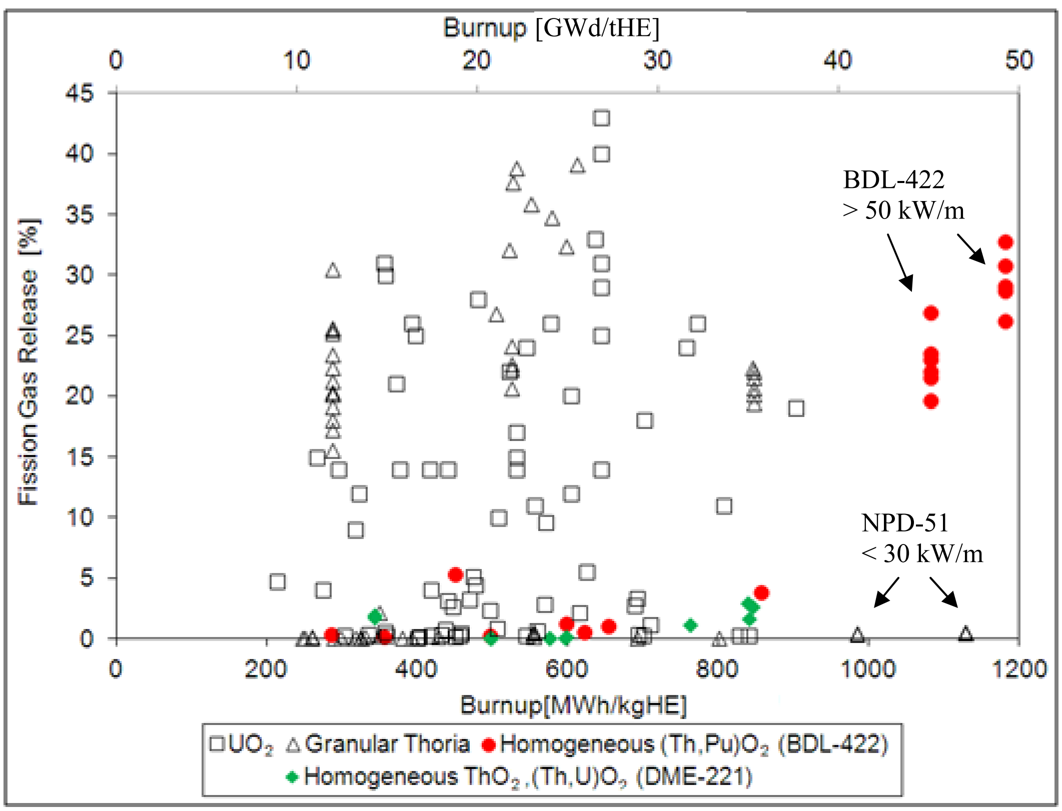

Figure 2: FGR [%] as a Function of Burnup [MWh/kgHE (bottom) and GWd/tHE (top)].

2. Pellet Microstructure Behaviour

Thoria fuels with homogeneous microstructure generally exhibit less grain growth than UO_2 fuel. For example, BDL-422 bundles ADC, ADE and ADF that contained $(Th,Pu)O_2$ pellets were irradiated at powers of 52-67 kW/m to burnups of 451 – 856 MWh/kgHE (19 – 36 GWd/tHE); a grain growth factor of ~ 3 was observed [9]. In comparison, at lower power (47 – 53 kW/m) and a burnup of 450 – 750 MWh/kgU (19 – 31 GWd/tHE), UO_2 is expected to have a grain growth factor of the order 1.5 to 3.5 [6]; this demonstrated that higher power and higher burnup Th-based fuel had grain growth comparable to lower power and lower burnup UO_2 fuel. Grain growth factor is the ratio of the observed pellet-centre grain size to that at the pellet periphery, assumed to represent the as-fabricated pellet-centre grain size.

No thoria fuel irradiated by AECL has exhibited columnar grain growth, despite several having powers exceeding 65 kW/m; UO_2 typically exhibits columnar grain growth at this power [6]. BDL-417 and BDL-422 operated at ~ 73 kW/m at the beginning of life (BOL) and only exhibited equiaxed grain growth [6] [10]. This is consistent with reports that suggest significantly higher temperatures are required for columnar grain growth to occur in thoria-based fuels (relative to UO_2) [12] [13]; columnar grain growth is affected by various material properties, for example the fuel vapour pressure (lower in Th than UO_2), and the temperature to melting point ratio (thoria has a higher melting point and improved thermal conductivity). Apparently, conditions for columnar grain growth in thoria are not achieved at powers up to 73 kW/m

3. Sheath Strain, Corrosion, Hydriding Behaviour and CANLUB Retention

Sheath residual strains (mid-pellet and pellet-pellet interface) of thoria fuels with homogeneous pellet microstructures are bounded by ±0.6 %, despite high burnups and powers, varying pellet geometries, and different fuel compositions. The residual sheath strains of the thoria fuels are comparable to UO_2 sheath residual strains [6]. Sheath hydriding/deuteriding and oxidation (internal and external) behaviour show no significant difference from UO_2 fuels. CANLUB retention in thoria-based fuel is consistent with that observed in UO_2 fuels.

4. Performance of Defected Thoria-Based Fuels

A fuel defect refers to a breach in the fuel sheath that allows heat transport system (HTS) coolant to enter the element, and fission products (and likely fuel) to escape the element. This section describes defect root causes, post-defect degradation, and fission product release from defected Zircaloy sheathed thoria-based fuel.

4.1 Defect Root Causes

To date, AECL experience has shown no defect root causes that are unique to thoria-based fuel. Defect root causes in thoria-based fuel experienced at AECL include: SCC (stress-corrosion cracking; from internal gas overpressure or power ramping), primary hydriding, longitudinal sheath ridging (from low density fuel pellets), and incomplete endcap-to-sheath closure welds. All of these defect mechanisms occur in standard CANDU UO_2 fuel [14], and are related to manufacturing or operational issues.

Thoria-based fuels irradiated by AECL have exhibited similar power ramp SCC defect thresholds to those developed empirically for UO_2 fuels. The differences in ThO_2 and UO_2 material properties (e.g., thermal conductivity and melting point) do not appear to have a major net effect on power ramp SCC defect thresholds, and may be offsetting in nature. Further investigation is required to determine the impact of differing ThO_2 and UO_2 material properties on power ramp performance.

4.2 Post-Defect Degradation

Both thoria and UO_2 fuels appear to exhibit similar secondary sheath degradation (sheath hydriding resulting from coolant ingress via primary defects). AECL experience with thoria-based and UO_2 fuels follow UO_2 secondary hydriding thresholds developed by Locke [15]. No unique post-defect degradation mechanism has been observed in thoria-based fuel; in some aspects, defected thoria fuel appears to experience reduced degradation relative to UO_2, as described below.

When UO_2 fuels operate in a defected state, the pellets experience oxidation (O/M ratio > 2.00). As a result, grain boundary oxidation increases fuel loss (to the HTS; i.e., erosion) and thermal conductivity is degraded leading to higher fuel temperatures. Higher fuel temperatures cause increased grain growth and enhanced fission gas release to the HTS [16]. Since ThO_2 is chemically stable, fuel oxidation does not occur and, therefore, fuel loss remains low and thermal performance is not expected to change. Further investigations are required to determine if grain growth (and fuel operating temperature) in thoria-based fuels are affected by fuel defects, and how increasing the Pu or U content in ThO_2 might impact defect performance.

Thoria fuels irradiated by AECL have consistently exhibited reduced pellet erosion and fission product release (Section 4.3) as compared to UO_2 fuel. For example, an axial split in DME-166 fuel (SCC failure resulting from a power-ramp) resulted in minimal erosion of the $(Th,U)O_2$ pellet (1.66 wt% UO_2 with 93 wt% ^{235}U in total U) under the large axial sheath split. Furthermore, during the parallel FDO-680 $(Th,U)O_2$ and FDO-681 UO_2 experiments (discussed in Section 4.3), negligible $(Th,U)O_2$ loss was observed, while significant UO_2 fuel loss occurred (Figure 3). Figure 3 shows typical erosion from the UO_2 pellet, but the $(Th,U)O_2$ pellet under the defect site shows no evidence of erosion; it is not clear if the unidentified phase observed in the $(Th,U)O_2$ micrograph is real or an artifact of the metallographic preparation process.

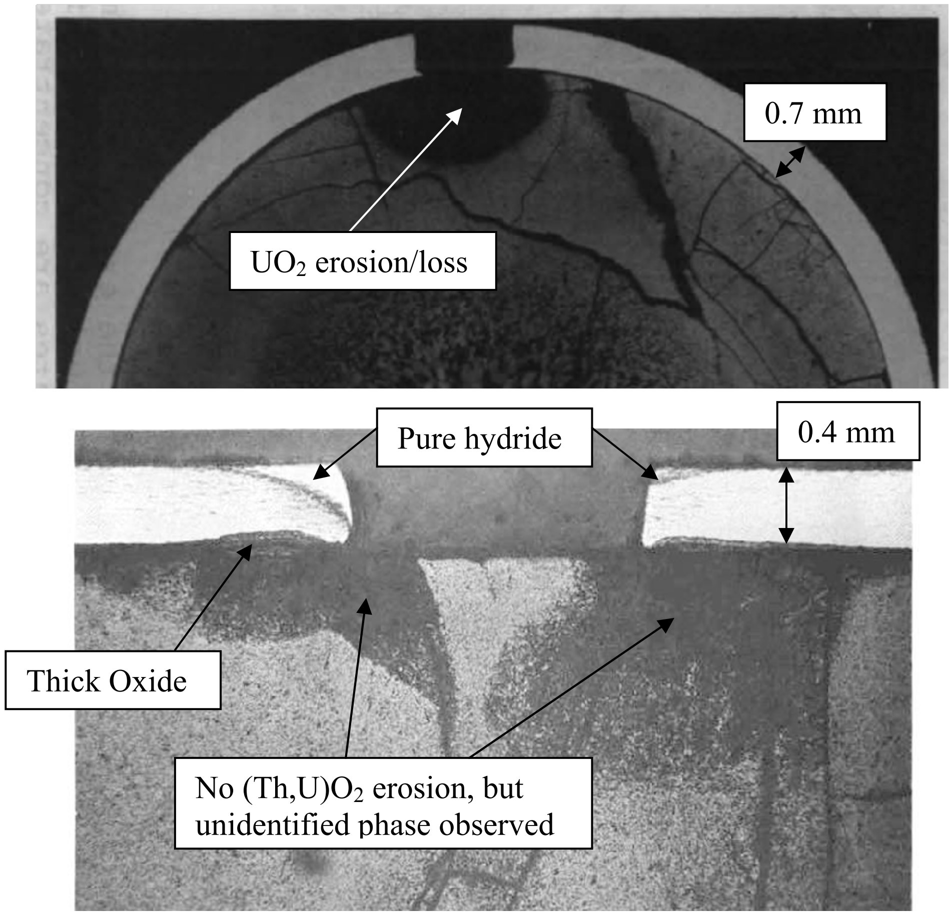

Figure 3: Fuel Erosion from UO_2 (top) and $(Th,U)O_2$ (bottom) under Similar Conditions

4.3 Fission-Product Release

Fission product release from thoria-based fuel has generally been less than that from UO_2 fuel operating under similar conditions. Early (pre-1970) AECL experience with defected thoria fuels did not include the collection of detailed fission product data; notwithstanding, anecdotal evidence existed at that time that radioiodine and fission gas release were lower in thoria than similarly operated UO_2 fuels that experienced failure.

In the mid-1970's, AECL conducted a parallel defect experiment where a defected UO_2 element (FDO-681 element RPL with 4.5 wt% ^{235}U in total U [17]) and defected $(Th,U)O_2$ element (FDO-680 element HKL with 4 wt% UO_2 in ThO_2 (93 wt% ^{235}U in total U)) were irradiated separately in the X-2 loop of the NRX reactor. While a complete analysis of this experiment is outside the scope of this paper, a simple analysis using ^{133}Xe release is described below.

Due to the inert nature of ^{133}Xe and its 5.2 day half-life, its release from defected fuel can be approximated as independent of defect size and uranium contamination. This approximation is shown in Equation (1) as a simple model for *R/B* (release over birth rate) for steady-state release [17], [18] from a defect. Equation (2) is the measured value, modified to include the element length (*L*) as FDO-680/1 were short elements.

$$\frac{R^{eq}}{B} = \frac{3\nu}{(\nu+\lambda)}\sqrt{\frac{X^2 D'(P)}{\lambda}}H + c \xrightarrow{Single\ defect\ (X=1),^{133}Xe\ (\nu\gg\lambda, c\ll defect\ release, H\approx 1)} \approx 3\sqrt{\frac{D'(P)}{\lambda}} \quad (1)$$

$$\frac{R^{eq}}{B} = \frac{M_c}{B\lambda}\left(\left(\lambda + \frac{\beta_p IX_{eff}}{M_c} + \frac{\epsilon}{M_c}\right)C_{eq}\right) \xrightarrow{^{133}Xe\ (IX_{eff}=0),\ \ no\ leakage} \approx \frac{M_c C_{eq}}{\left(B = F_f y_c L P\right)} \quad (2)$$

Where:

R^{eq}/B	=	Equilibrium release rate to birth ratio
ν [s^{-1}]	=	Escape rate coefficient
λ [s^{-1}]	=	Decay constant
X	=	Number of defects
$D'(P)$ [s^{-1}]	=	Emprical diffusion coefficient
H	=	Pre-cursor correction factor
M_c [kg]	=	Coolant mass
$\beta_p IX_{eff}$ and ϵ [kg.s^{-1}]	=	Purification and leakage terms
C_{eq} [Bq.kg^{-1}]	=	Activity concentration
F_f [fission.s^{-1}.kW^{-1}]	=	Fission rate per kilowatt
y_c [%]	=	Cumulative yield
L [m]	=	Defected element length
c	=	Uranium contamination contribution
P [kW.m^{-1}]	=	Defected element power

Figure 4 shows the results of Equation (2) applied to CANDU reactor data [19] and NRX data from a suite of UO_2 defect experiments [17] compared with FDO-680 and FDO-681 data. The FDO-681 UO_2 and FDO-680 $(Th,U)O_2$ data are from short element irradiations (~ 170 mm stack length versus ~ 480 mm in the other experiments and reactor data); other experiment and reactor parameters are similar. This preliminary analysis indicates significantly reduced (10 – 100 times) release of ^{133}Xe from defected $(Th,U)O_2$ versus UO_2 in the 50 – 60 kW/m power range. Further investigations are required to confirm this finding, and to further explore fission-product release behaviour from defected thoria-based fuels.

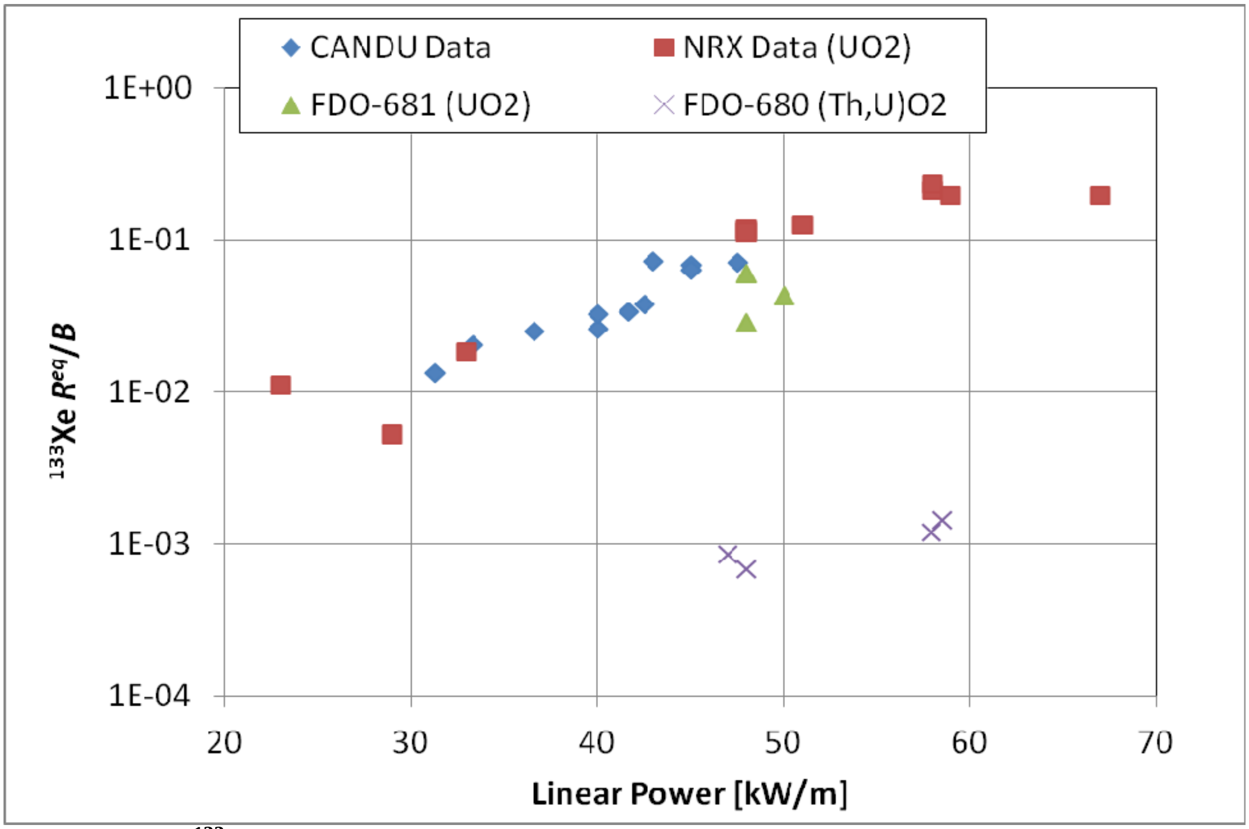

Figure 4: ^{133}Xe Release Fraction (R^{eq}/B) as a Function of Power. 'CANDU Data' is Commercial Reactor Data, 'NRX Data' is UO_2 Defects in the NRX X-2 Loop, and 'FDO-680/FDO-681' were Short Elements Irradiated in the NRX X-2 Loop.

5. Current and Future Irradiations

The DME-221 experiment includes the irradiation of eighteen fuel elements in a demountable configuration [8], [10]. Six of the elements contain natural thoria, while the other twelve contain $(Th,U)O_2$. Six of the $(Th,U)O_2$ elements contain 1.0 wt.% ^{235}U (in total heavy elements), while the remaining six contain 1.5 wt.% ^{235}U. The elements were irradiated to maximum powers of 36-55 kW/m. To date, PIE has been completed on twelve DME-221 elements that achieved burnups of 361-929 MWh/kgHE (15-39 GWd/tHE). Six DME-221 elements are planned to continue their irradiation to burnups of 1000-1500 MWh/kgHE (42-63 GWd/tHE).

The "Thoria Roadmap Project" is a major development in AECL's recent initiatives to ensure sustainable nuclear energy for future generations. Launched in 2012, this project seeks to identify knowledge gaps in science and technology (S&T) areas associated with thoria-based fuels and fuel cycles. This is being accomplished by measuring fundamental requirements against current understanding in various S&T areas. The outcome will be a "roadmap" to address gaps in current understanding, which will better position Canadian industry to implement thoria-based fuels in commercial nuclear power reactors. Irradiation Testing is one of the S&T areas being investigated. Preliminary investigations indicate that additional irradiation testing is required to understand the effect of pellet grain size variations and Pu homogeneity in $(Th, Pu)O_2$ fuels irradiated to high burnup. These irradiation tests are presently in the early stages of planning.

6. Conclusions

AECL has 50 years of experience with thoria irradiation tests and performance assessments (ThO_2, $(Th,U)O_2$, and $(Th,Pu)O_2$) with burnups up to 1130 MWh/kgHE (47 GWd/tHE) and linear heat ratings up to 73 kW/m. Experience includes defected fuel experiments, power ramp tests, instrumented fuel, various pellet fabrication methodologies, and pellet microstructure variations.

In general, AECL thoria-based fuel performance is bounded by that of UO_2 fuel. Thoria-based fuels having high-density pellets with homogeneous microstructures exhibit superior fission-gas release behaviour to UO_2. Defected thoria fuels exhibit lower fission-product release and less fuel degradation; one experiment showed 10-100 times reduced release of ^{133}Xe from defected $(Th,U)O_2$ versus UO_2 in the 50-60 kW/m power range.

Work is on-going to identify and address thoria fuel S&T gaps. This includes extending the burnup of DME-221 ThO_2 and $(Th,U)O_2$ fuels to burnups of 1500 MWh/kgHE (63 GWd/tHE), and to investigate the effect of pellet grain size and Pu homogeneity on $(Th,Pu)O_2$ fuel performance at high burnup. Other initiatives are expected to be undertaken as additional S&T gaps are identified by AECL's Thoria Roadmap Project.

7. References

[1] I.J. Hastings, K. Bradley, J. Hopwood, P. Boczar, S. Kuran, S. Livingstone, "We Can Use Thorium", Nuclear Engineering International, 2009 August 28.

[2] P.G. Boczar, B. Hyland, E. Hagberg, "Qualification of Reactor Physics Toolset for a Thorium-Fuelled CANDU Reactor", Proceedings of the 18th International Conference on Nuclear Engineering, Xi'an, China, 2010 May 17-21.

[3] Canadian Nuclear Society (CNS), "WR-1: Whiteshell Rector No.1", CNS webpage, http://media.cns-snc.ca/history/wr-1/wr-1_1.html, 2013 March.

[4] G.L. Olson, R.K. McCardell, D.B. Ilum, "Fuel Summary Report: Shippingport Light Water Breeder Reactor", Idaho National Engineering and Environmental Laboratory Bechtel BWXT, LLC, INEEL/EXT-98-00799 Rev 2, 2002 September.

[5] A.S. Bain, J. Christie, I.J. Hastings, "Performance of ThO_2-UO_2 Fuel Irradiated in the NRU Reactor at CRNL", Transactions of the American Nuclear Society, Volume 27, Page 307, 1977.

[6] M.R. Floyd, "Extended-Burnup CANDU Fuel Performance", Proceedings of the 7th International CNS CANDU Fuel Conference, Kingston, Canada, 2001 September 23-27.

[7] A.D. Smith, J.A. Walsworth, R.E. Donders, P.J. Fehrenbach, "In-Reactor Measurement of Operating Temperature in $(Th,U)O_2$ Fuel: Effect of Pellet Microstructure", Proceedings of the 6th Annual Conference of the Canadian Nuclear Society, Ottawa, 1985 June 3-4.

[8] S. Corbett, H.B. Hamilton, M.R. Floyd, N.F. Harrison, S.J. Livingstone, "DME-221 Thoria Fuel: Fabrication, Irradiation Testing and Post-Irradiation Examination", Proceedings of the 12th International Conference on CANDU Fuel, Kingston, Canada, 2013 September 15-18.

[9] M. Karam, F.C. Dimayuga, J. Montin, "Post-Irradiation Examination of CANDU Fuel Bundles Fuelled with $(Th,Pu)O_2$", Proceedings of the 11th International Conference on CANDU Fuel, Niagara Falls, Canada, 2010 October 17-20.

[10] M.R. Floyd, "Advanced Fuel Cycle Development at Chalk River Laboratories", Proceedings of the International Conference on Future of Heavy Water Reactors (HWR-Future), Ottawa, Canada, 2011 October 2-5.

[11] I.J. Hastings, "Effect of Initial Grain Size on Fission Gas Release from Irradiated UO_2 Fuel", Journal of the American Ceramic Society, Vol. 66, No. 9, 1983 September.

[12] A.S. Bain, "Crack Healing and Void Movement During Irradiation of ThO_2 – 2 wt% UO_2", AECL Report, AECL-3008, 1967 May.

[13] A.E. Walter, "Fast Spectrum Reactors", Springer Publishing, ISBN 1441995714, 2012.

[14] A.M. Manzer, "CANDU Fuel Performance", Proceedings of the Water Channel Reactors Fuels and Fuel Channels: Design, Performance, Reasearch and Development, Technical Committee Meeting, IAEA-TECDOC-997, Vienna, 1996 December.

[15] D H Locke, "The Behaviour of Defective Reactor Fuel," Nuclear Engineering and Design, vol. 21, pp. 318-330, 1972.

[16] A. El-Jaby, B.J. Lewis, W.T. Thompson, F. Iglesias, and M. Ip, "A General Model for Predicting Coolant Activity Behaviour for Fuel-Failure Monitoring Analysis," Journal of Nuclear Materials, no. 399, pp. 87-100, 2010 April.

[17] B.J. Lewis, "Fundamental Aspects of Defective Nuclear Fuel Behaviour and Fission Product Release", Journal of Nuclear Materials, Vol. 160, Pg 201-217, 1988 December.

[18] S.J. Livingstone, "Development of an On-Line Fuel Failure Monitoring System for CANDU Reactors", PhD Thesis, Royal Military College of Canada, 2012 March.

[19] S. Livingstone, "Progress in Developing an On-Line Fuel-Failure Monitoring Tool for CANDU Reactors", Proceedings of the 11th International Conference on CANDU Fuel, Niagara Falls, Canada, 2010 October 17-20.