12th International Conference on CANDU Fuel
Holiday Inn Kingston-Waterfront
Kingston, Ontario, Canada, 2013 September 15-18th

DME-221 THORIA FUEL: FABRICATION, IRRADIATION TESTING, AND POST-IRRADIATION EXAMINATION

S. Corbett, M.R. Floyd, H. Hamilton, S. Livingstone, N.F. Harrison
Atomic Energy of Canada Limited, Chalk River Laboratories, Chalk River, Ontario, Canada

ABSTRACT – Thoria (ThO_2) has several advantages over Urania (UO_2) as a nuclear fuel; its higher thermal conductivity results in lower operating temperatures, and its higher melting point and chemical inertness promise better stability under postulated accident conditions. Thorium is a "fertile" material, breeding mainly fissile ^{233}U as it is irradiated, making it more proliferation resistant. Thorium is also abundant as a natural resource, and is therefore an attractive future energy source. Past irradiation tests have shown that pellet microstructure greatly influences the irradiation performance of thoria fuels (e.g., fission-gas release is strongly influenced by the as-fabricated fuel microstructure). The "DME-221" test, conducted at AECL's Chalk River Laboratories, was designed to demonstrate the superior irradiation performance of thoria fuels characterized by homogeneous, high-density microstructures. Eighteen DME-221 fuel elements were irradiated in the National Research Universal (NRU) loops in a 36-element demountable bundle. Six of the elements had pellets comprised of natural thoria (ThO_2), and twelve comprised of thoria blended with 1.0 wt.% or 1.5 wt% ^{235}U in $(Th, U)O_2$. Various power histories were achieved as a result of the varied initial fissile concentration. To date, DME-221 thoria fuel has demonstrated excellent performance to burnups up to ~930 MWh/kgHE (39 GWd/tHE); fission-gas release is substantially lower than that expected from UO_2 fuels experiencing similar operating histories. This paper highlights the fabrication, irradiation testing and post-irradiation examination of twelve DME-221 fuel elements. The remaining six DME-221 elements are currently planned for irradiation in NRU to burnups up to 1500 MWh/kgHE (42-63 GWd/tHE).

Introduction

AECL has over 50 years experience with thoria fuel development, irradiation testing and performance assessments [1]. ThO_2-based fuel has many advantages when compared to UO_2 fuel, a result of thoria's superior physical and chemical properties such as higher melting point, higher thermal conductivity, and greater chemical inertness relative to UO_2. Previous studies have noted that ThO_2-based fuel will release fewer fission products than UO_2-based fuel operating under similar conditions [2].

The DME-221 experiment involves the fabrication, irradiation and post-irradiation examination (PIE) of eighteen demountable CANLUB-coated Zircaloy-4 elements fuelled with ThO_2 and $(Th, U)O_2$ pellets. The objectives are: to compare the performance of ThO_2 with UO_2 fuel; show that the fuel's microstructure is crucial to performance improvement [3]; and determine fission-gas release (FGR) from thoria fuels for three different power histories. The DME-221 elements were irradiated on the outer ring of a CANDU-type demountable element (DME) fuel bundle in the U1 and U2 test loops of the National Research Universal (NRU) reactor at Chalk River Laboratories (CRL). This paper presents the post-irradiation examination (PIE) results for twelve DME-221 elements, irradiated at maximum sustained mid-plane element powers ranging from 36 kW/m to 55 kW/m, to burnups ranging from 360 MWh/kgHE (15 GWd/tHE) to 930 MWh/kgHE (39 GWd/tHE). The remaining six elements are expected to continue their irradiation to proposed burnups of 1000 to 1500 MWh/kgHE (42 to 63 GWd/tHE).

1. Fuel Design and Fabrication

The DME-221 fuel was fabricated in the Chalk River Advanced Fuel Technologies (CRAFT) laboratories at CRL. The fabrication comprised six pellet types: natural ThO_2 featuring standard length pellets (L/D[1] ~1.3) and short pellets (L/D ~0.7); ThO_2 + 1.0 wt.% ^{235}U in heavy elements (HE) (standard L/D and short L/D pellets); and ThO_2 + 1.5 wt.% ^{235}U in HE (standard L/D and short L/D pellets). Shorter-length pellets are expected to reduce sheath ridging at the pellet interfaces and improve the resistance of the fuel to stress-corrosion cracking (SCC) at high burnup. Enriched UO_2 (93.1 wt.% ^{235}U in total U) was used to achieve the target concentrations of ^{235}U in the blended $(Th, U)O_2$ pellets. The elements also contained plena (made of Zr-4) installed at one end to accommodate fission-gas release. Plena were 17 mm long in elements containing reduced L/D pellets, and 24 mm long in elements containing standard L/D pellets.

Previous fabrication experience at AECL has demonstrated that ThO_2-based fuels with high (≥95% of theoretical) densities can be produced that contain micro-structural irregularities which affect fuel performance. In these cases, the granular structure of the press-feed was not destroyed during final pressing. The sintered pellets contained very dense regions, corresponding to the original press-feed granules, surrounded by porous regions. The irradiation performance of such fuels was below expectations with respect to fission gas release [3]. The fabrication route for DME-221 pellets was chosen to avoid producing this granular structure. The DME-221 fuel was fabricated to very high densities (98.7% of theoretical)[2] with a homogenous microstructure. An image of standard-L/D DME-221 pellets, as well as the pre-irradiated fuel microstructure, are shown in Figure 1; minimal porosity and no residual granules can be observed in the as-fabricated fuel.

[1] L/D is the ratio of pellet length to diameter.
[2] Theoretical density of pure thorium dioxide = 10.00 g/cm^3 at 298.15 K [2].

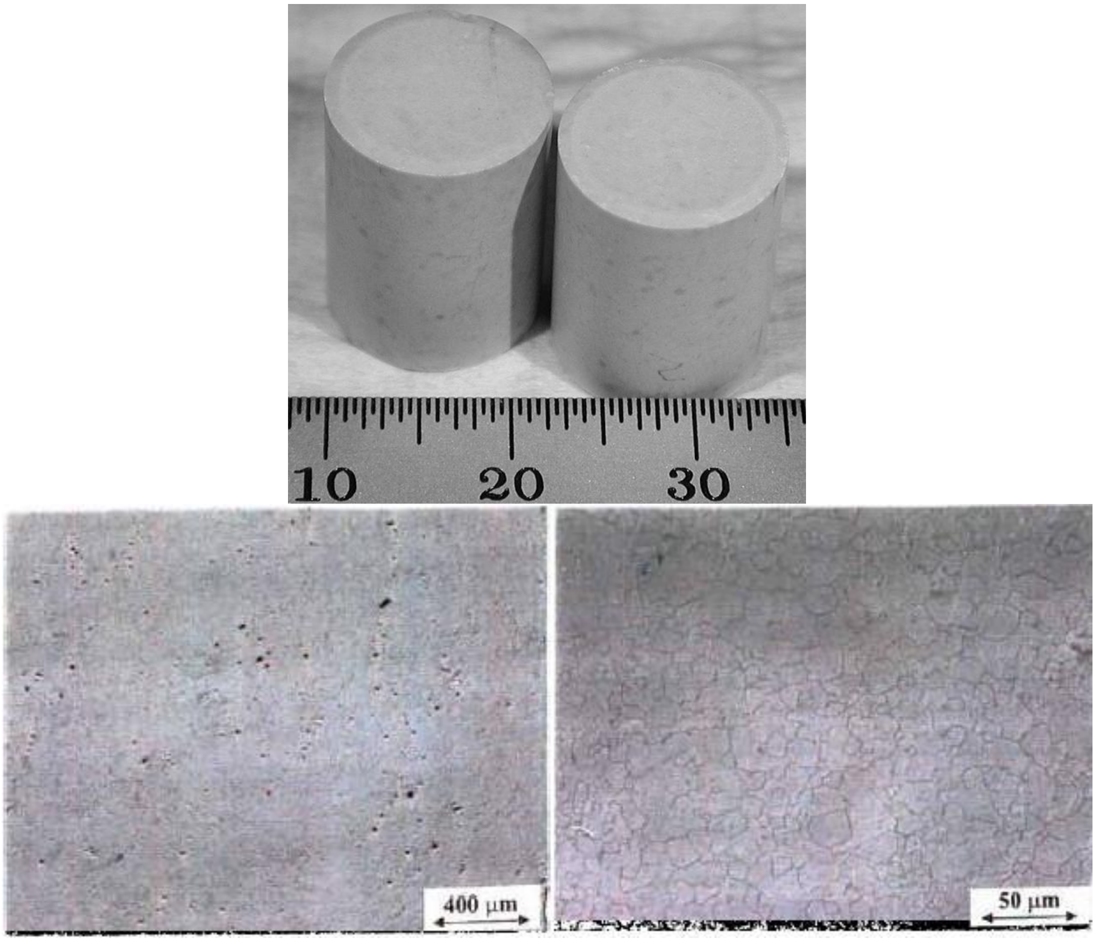


Figure 1 Typical DME-221 Fuel Pellets are shown in the top image (standard-length pellets). Typical pre-irradiation porosity and fuel microstructure are shown in the bottom two images. As-fabricated grain size averaged ~ 6-8 µm.

2. Irradiation History

Irradiation testing in the NRU loops is facilitated by the use of a fuel carriage that accommodates a string of six test bundles [4]. The centre element of CANDU-type bundles is replaced with a guide tube to accommodate the fuel carriage central tie rod which extends vertically through the central element position [4]. The flux shape along the string is roughly cosine, with the maximum flux slightly below centre line at bundle position #4 (Figure 2).

The DME-221 elements experienced three different operating histories, based on the three initial fuel compositions (summarized in Table 1). The natural thoria-fuelled elements (four examined in the hot cells) were subject to a steadily increasing power history, as fissile ^{233}U was produced in the fuel during irradiation. These elements achieved maximum sustained mid-plane powers ranging from 36 kW/m to 41 kW/m, to mid-plane burnups ranging from 361 MWh/kgHE (15 GWd/tHE) to 619 MWh/kgHE (26 GWd/tHE).

The ThO_2 + 1.0 wt.% ^{235}U fuelled elements (three examined) were subject to a constant power history with maximum sustained mid-plane powers ranging from 40 kW/m to 46 kW/m (the point at which FGR in UO_2 fuels starts to increase), to mid-plane burnups ranging from 499 MWh/kgHE (21 GWd/tHE) to 839 MWh/kgHE (35 GWd/tHE).

The ThO_2 + 1.5 wt.% ^{235}U fuelled elements (five examined) were subject to a declining power history with mid-plane beginning-of-life (BOL) powers between 52 kW/m and 55 kW/m (FGR in UO_2 fuel is relatively large in this range, but expected to be lower in thoria fuels), to burnups ranging from 594 MWh/kgHE (25 GWd/tHE) to 929 MWh/kgHE (39 GWd/tHE). BOL powers decreased to mid-plane powers of ~ 40 kW/m during the irradiation.

Table 1 Summary of DME-221 Element Operating Histories

DME-221 Fuel Composition	Elements Examined	Operating History	Normalized* Maximum Sustained Linear Power at Mid-Plane, kW/m	Normalized* Outer Element Burnup, MWh/kg HE (GWd/t HE)
Natural ThO_2	4	Increasing	36 – 41	361 – 619 (15 – 26)
ThO_2 + 1.0 wt.% ^{235}U	3	Constant	40 – 46	499 – 839 (21 – 35)
ThO_2 + 1.5 wt.% ^{235}U	5	Decreasing	52 – 55	594 – 929 (25 – 39)
* Normalized to Chemically-Measured Burnups				

The power history of DME-221 elements was calculated using AECL's BURFEL code [5]. Sections of fuel were removed from the mid-element location of DME-221 elements for burnup determination by ^{139}La (measured by HPLC) and U isotopic analysis (measured by TIMS). The derived burnups based on the La concentrations were determined using WIMS and WOBI codes [6]. WIMS was also used to calculate burnup based on the U isotopic ratios. The BURFEL-calculated burnups were then normalized to the chemically measured ones, and the normalized data were used to plot the power history for the DME-221 elements. Figure 3 to Figure 5 illustrate the element power histories for each of the three fuel compositions. Maximum sustained power is defined as the highest power experienced by the fuel element for any sustained 12-hour period during a given burnup step.

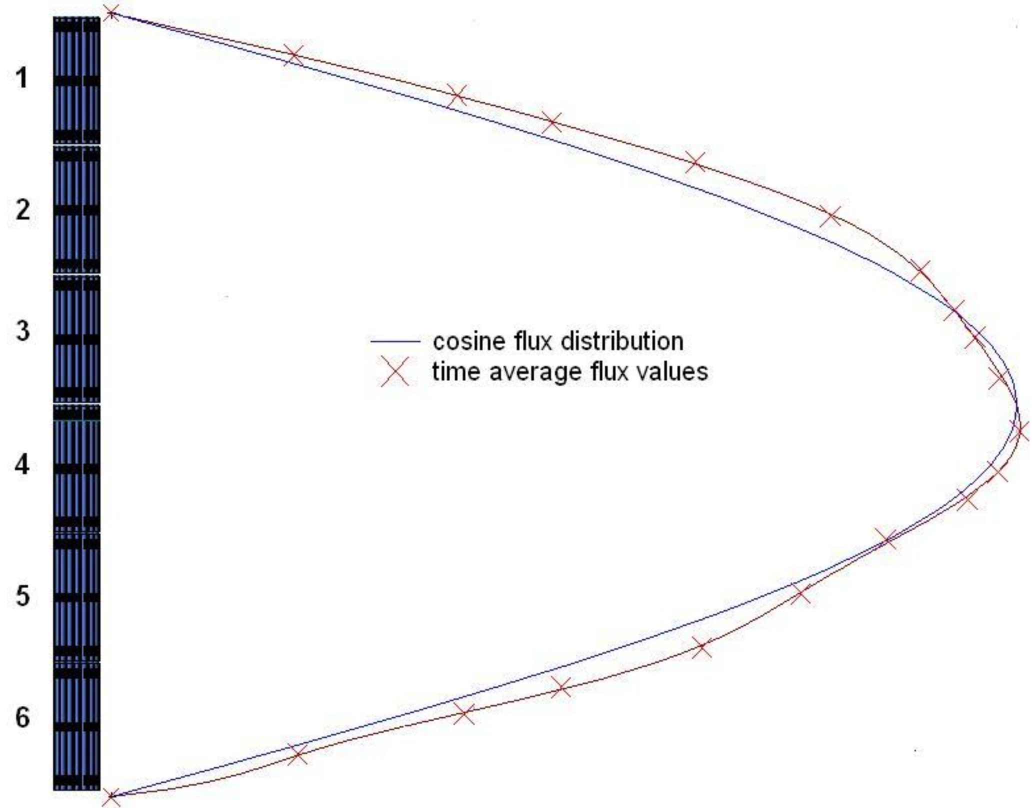


Figure 2 A Schematic of a Fuel String in an NRU Test Section Showing Bundle Positions and Typical BURFEL-Calculated Flux Profile. DME-221 elements were irradiated on a CANDU-type demountable fuel bundle in position #3 in the NRU loops.

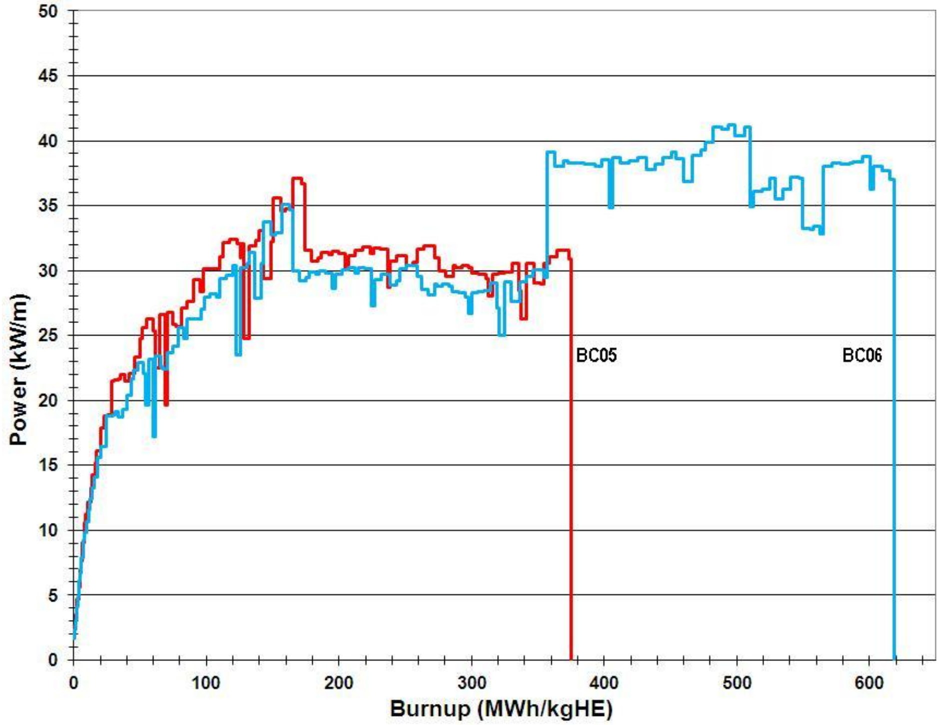


Figure 3 Increasing Power History Experienced by DME-221 Natural ThO_2 Fuelled Elements. Two of four elements (all with similar power histories) are shown.

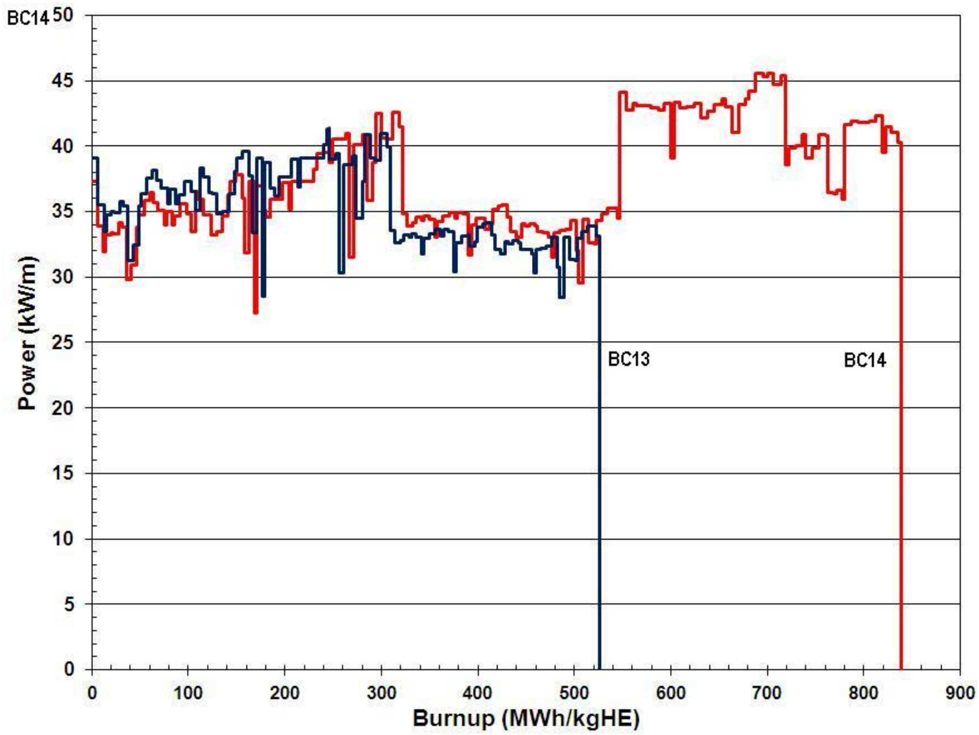


Figure 4 Constant Power History Experienced by DME-221 $(Th,U)O_2$ Elements with 1.0 wt.% ^{235}U in total HE. Two of three elements (all having similar power histories) are shown.

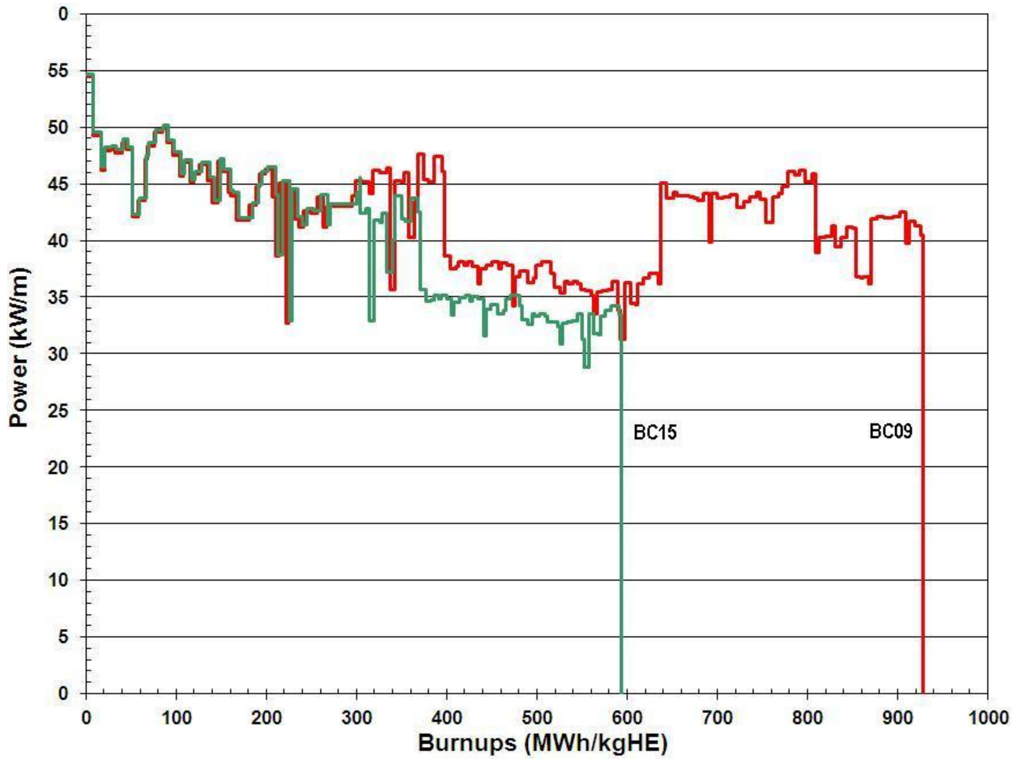


Figure 5 Declining Power History Experienced by DME-221 (Th,U)O_2 Elements with 1.5 wt.% ^{235}U in total HE. Two of five elements (all having similar power histories) are shown.

3. Post-Irradiation Examination

3.1 Element Profilometry

The outer elements were removed from the demountable bundle core, and profilometry was conducted in the CRL hot cells. The measured element diameters showed that the residual sheath strains were mainly compressive at the mid-pellet (MP) and pellet-interface (PI) locations for elements with mid-plane burnups < 600 MWh/kgHE. For elements achieving burnups of 600 - 930 MWh/kgHE, MP sheath strains were negligible (~ 0 %), but slightly tensile at PI locations.

The highest strains were observed in DME-221 elements BC17 and BC18, which experienced declining power histories from BOL powers of 53 kW/m and 52 kW/m, to burnups of 914 MWh/kgHE and 903 MWh/kgHE, respectively. These elements had slightly tensile average MP sheath strains of +0.1% (maximum of +0.3%), with average PI strains of +0.4% (maximum +0.5%). Table 2 summarizes the residual mid-plane sheath strain results for the twelve examined DME-221 elements. Figure 6 compares average mid-pellet sheath strains from DME-221 elements to that of UO_2 elements with similar operating histories and burnup ranges. Overall, the DME-221 elements that experienced a declining power history with a relatively high BOL power exhibited larger sheath strains, which tended to increase with burnup, as expected. DME-221 elements with reduced-L/D pellets did not appear to exhibit any significant reduction in sheath strain at the mid-pellet or pellet interface regions compared to elements containing standard L/D pellets.

Overall, the DME-221 thoria-fuelled elements exhibited lower MP sheath strains compared to standard UO_2 fuels with similar power histories (Figure 6). Purdy et al. [7] assessed 20 years of UO_2 fuel irradiation data in CANDU power plants. The frequency distributions of average outer element MP strain data for normally operated UO_2 bundles is +0.09% (standard deviation of 0.19%) [7]. Outer element MP sheath strains for UO_2 bundles within the normal operating envelope (discharged at burnups < 450 MWh/kgU) were mostly tensile (with 99% confidence, maximum sheath strains can be expected to reach 0.7%) [7]. Higher burnup natural UO_2 fuels (500-800 MWh/kgU) exhibit MP sheath strains up to 1.5% [8]. The DME-221 sheath strain results suggest that ThO_2-based fuels can be irradiated at higher power ratings and for longer burnups than UO_2-based fuels to produce the same dimensional changes.

Table 2 Residual Sheath Strain at Mid-Pellet (MP) and Pellet Interface (PI) Locations

DME-221 Fuel Composition	Burnup Range, MWh/kgHE (GWd/tHE)	Operating History	Sheath Strain	
			Average MP (%)	Average PI (%)
ThO_2	361 – 619 (15 – 26)	Increasing	-0.3 to 0.0	-0.1 to +0.2
ThO_2 + 1.0 wt% ^{235}U	499 – 839 (21 – 35)	Constant	-0.4 to 0.0	-0.1 to +0.3
ThO_2 + 1.5 wt% ^{235}U	594 – 929 (25 – 39)	Declining	-0.2 to +0.1	0.0 to +0.4

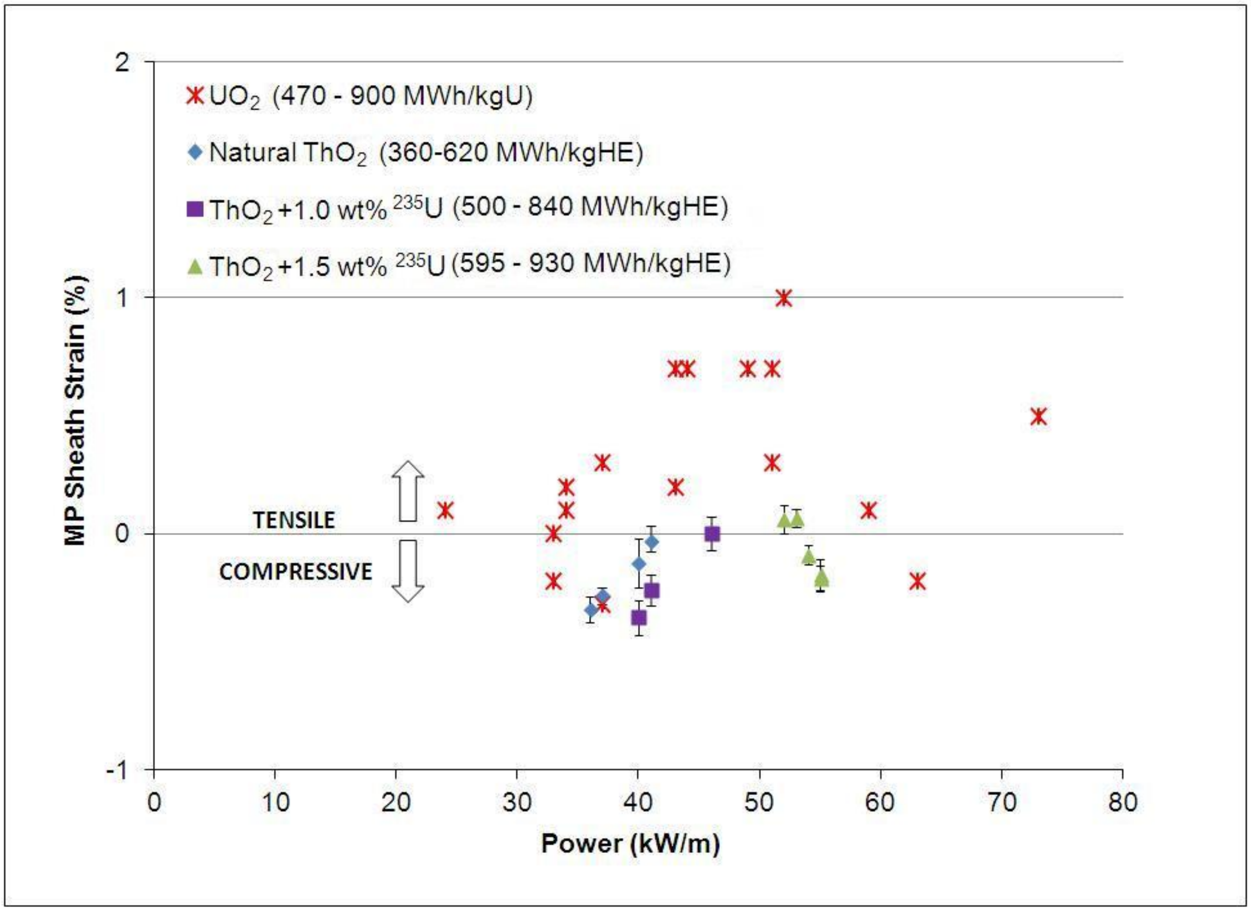


Figure 6 Comparison of Average Mid-Pellet Sheath Strains in DME-221 Elements Compared to UO_2 Outer Elements with Similar Burnup Ranges and Operating Histories. Error bars indicate the standard deviation from the average MP sheath strain value.

3.2 Fission Gas Analysis

Fission-gas release (FGR) is temperature-driven, originating mainly in the centre of fuel pellets where the highest temperatures are achieved, coinciding with grain growth, grain-boundary bubble formation, and swelling [8]. Some of the produced fission-gas (mainly xenon and krypton) ultimately migrates to the element internal void space. The FGR results for each of the three fuel compositions are given in Table 3.

The DME-221 elements generally exhibited very low FGR (< 3%), attributed to low operating fuel temperatures resulting from the higher thermal conductivity, and to a dense, homogeneous microstructure. DME-221 elements irradiated to burnups < 600 MWh/kgHE, regardless of their power history or fuel composition, achieved FGR below 0.11%. Elements with burnups between 840 MWh/kgHE and 930 MWh/kgHE showed a slight increase in FGR, ranging from ~1.2% to 2.8%. The highest FGR was observed in elements BC09 (2.8%) and BC17 (2.6%), both having a declining power history, and irradiated to burnups of ~930 MWh/kgHE and 914 MWh/kgHE, respectively. Figure 7 compares DME-221 FGR to that of UO_2 outer elements with similar operating histories; thoria FGR is observed to be significantly lower than UO_2 FGR.

Table 3 DME-221 Element Fission-Gas Release (FGR)

Element Identity	Fuel Composition	Burnup Range (MWh/kgHE)	Power History Type	%FGR (Xe + Kr)
BC03, BC04, BC05, BC06	ThO_2	~360 to 620	Increasing	0.05 to 0.1
BC07, BC13, BC14	ThO_2 + 1.0 wt% ^{235}U	~500 to 840	Constant	0.06 to 1.2
BC09, BC11, BC15, BC17, BC18	ThO_2 + 1.5 wt% ^{235}U	~595 to 930	Declining	0.08 to 2.8

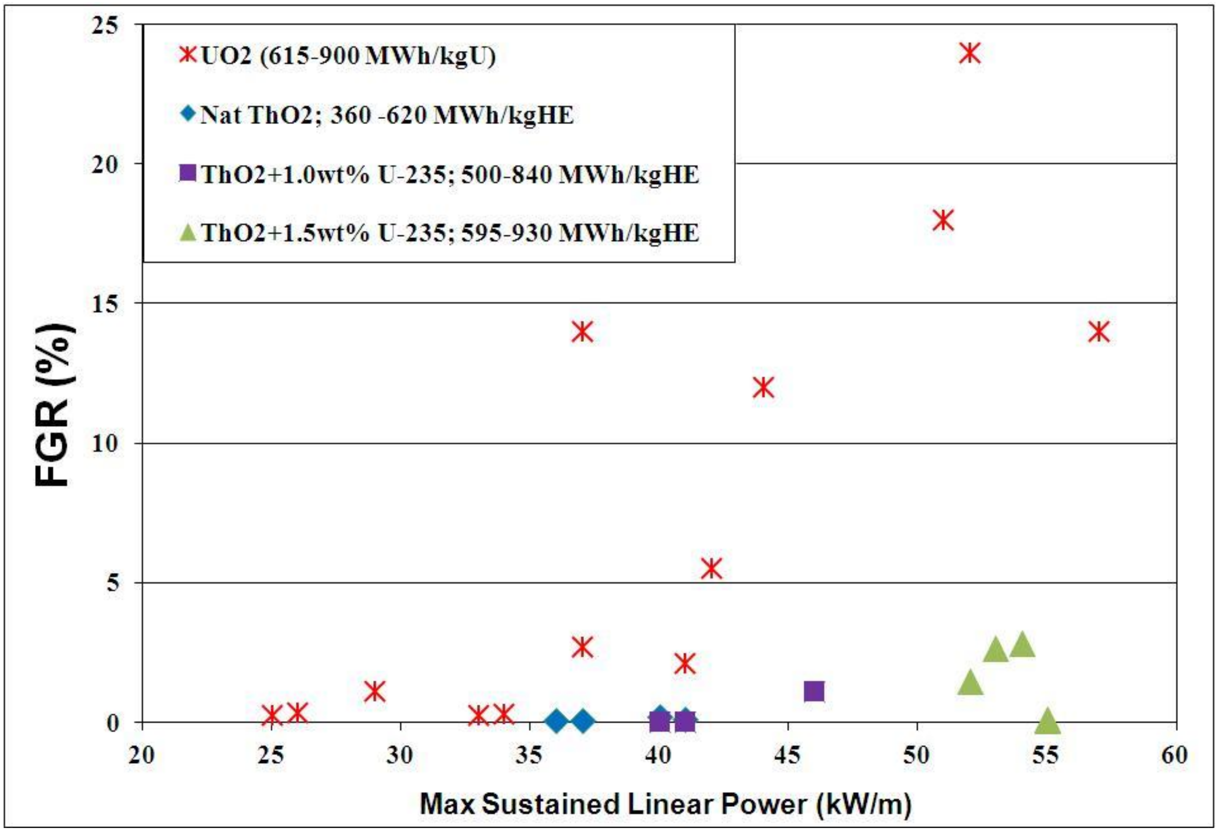
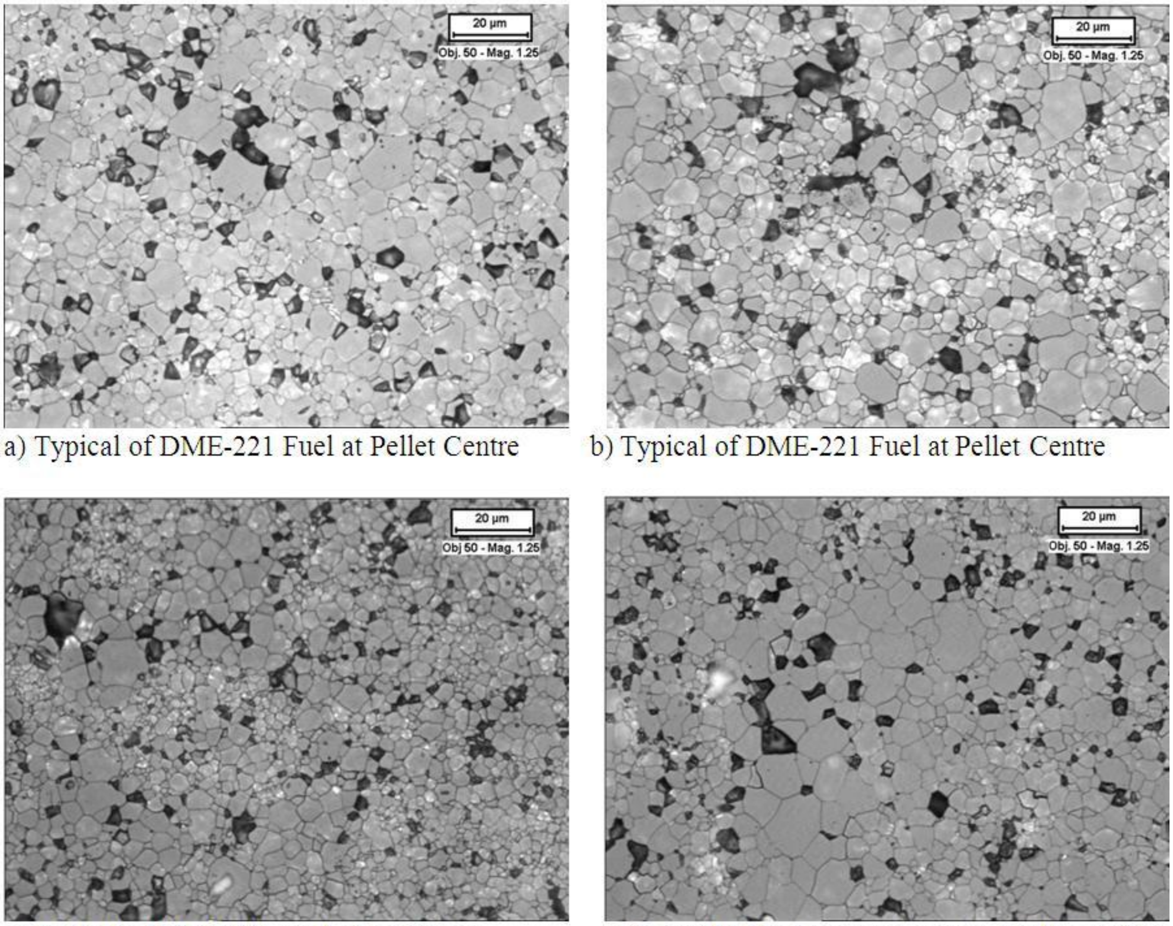


Figure 7 Comparison of Fission-Gas Release % (FGR) at Maximum Sustained Linear Powers in DME-221 and UO_2 Elements with Similar Operating Histories and Burnup Ranges.

3.3 Ceramographic Examination

DME-221 fuel was examined for grain size and microstructural features. The as-fabricated grain size of DME-221 fuel pellets averaged 6 to 8 μm. In general, little or no grain growth was observed at the pellet centre or other radial locations in irradiated DME-221 fuel up to 930 MWh/kgHE. Only a slight increase in grain size at the pellet centre was observed in the elements with declining power histories and relatively high BOL powers (52-55 kW/m). In comparison, UO_2 fuels irradiated at powers ~ 50 kW/m have exhibited grain-growth factors of 1.5 at 450 MWh/kgHE, to as high as 3.5 at 750 MWh/kgHE [8]. At 59 kW/m and 540 MWh/kgHE, grain-growth factors of 3.5 have been observed in UO_2 fuel [9]. More significant equiaxed grain growth at the pellet centre region would thus be expected in UO_2 irradiated at powers ~55 kW/m. 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.

Fuel temperature is the primary driving force for grain growth. The lower grain growth observed for DME-221 fuel compared to similarly operated UO_2 fuel is likely attributable to the same properties identified for the lower gas release; higher thermal conductivity, and a dense, homogeneous microstructure. The fact that thoria is a more refractory material than UO_2 may also be a contributing factor. Several ceramographic images depicting typical DME-221 fuel microstructure are shown in Figure 8.



a) Typical of DME-221 Fuel at Pellet Centre

b) Typical of DME-221 Fuel at Pellet Centre

c) Typical of DME-221 Fuel at Periphery

d) Typical of larger grains among smaller grains

Figure 8 Ceramographic Images Showing Typical DME-221 Irradiated Fuel Microstructure. Microstructure was observed to be similar for all three DME-221 fuel compositions. The sharp-angled pores are the result of grain pullout during sample preparation.

4. Summary

DME-221 thoria elements have exhibited excellent in-reactor performance, attributed to the fuel's dense, homogeneous, as-fabricated microstructure, and ThO_2-based fuels' inherent material properties.

The DME-221 elements exhibited lower sheath strains at the MP locations when compared to standard UO_2 fuels with similar power histories. Residual sheath strains were mainly compressive at the mid-pellet (MP) and pellet-interface (PI) locations for elements for mid-plane burnups < 600 MWh/kgHE (-0.4% to -0.2% at MP; and -0.1% to 0.0% at PI). Elements with burnups of 600-930 MWh/kgHE had negligible MP sheath strains, and slightly tensile strains at PI locations (+0.1% to +0.4%). The elements experiencing a declining power history (relatively high BOL power) had more significant sheath strains that increased with burnup. DME-221 elements with reduced-L/D pellets did not appear to exhibit a reduction in sheath strain at the mid-pellet or pellet interface regions when compared to standard L/D pellets.

Little to no grain growth at the pellet centre was observed in DME-221 fuels. Some grain growth was observed at the pellet centre region in elements with relatively high BOL powers of ~ 55 kW/m. Fission-gas release was significantly lower in DME-221 fuel than for similarly operated UO_2 fuels (ranging from <0.1% to ~3%), attributed to low centreline temperatures. FGR was negligible in DME-221 elements irradiated to burnups < 600 MWh/kgHE (~ 0.1%). DME-221 elements experiencing burnups ranging from 840 MWh/kgHE to 930 MWh/kgHE were observed to have slightly higher FGR (~1% to 3%).

5. Conclusions

Overall, the DME-221 elements exhibited excellent fuel performance for each of the three fuel compositions and operating histories when compared to that of similarly operated UO_2. No obvious difference in performance was observed in DME-221 elements having standard and reduced L/D pellets. The improvement in fuel performance parameters (i.e., benign FGR, low sheath strains, and low grain growth) observed in DME-221 fuel compared to similarly operated UO_2, are a result of the material property differences between ThO_2-based fuel and UO_2-based fuel. Given these observations, no operations-related failures should occur in thoria-based fuels having compositions and operating conditions similar to that of the DME-221 elements.

6. References

[1] S.J. Livingstone, and M.R. Floyd, "Thoria Irradiation and Post-Irradiation Examination Experience at AECL", Proceedings of the 12th International Conference on CANDU Fuel, CW-124950-CONF-006 (2013).

[2] J. Belle, and R.M. Berman, "Thorium Dioxide: Properties and Nuclear Applications", Naval Reactors Office, United States Department of Energy, Government Printing Office, Washington, D.C. (1984).

[3] A.D. Smith, J.A. Walsworth, R.E. Donders, and P.J. Fehrenbach, "In-Reactor Measurement of Operating Temperature in ThO_2-UO_2 Fuel: Effect of Pellet Microstructure", Proceedings of the 6th Annual Conference of the Canadian Nuclear Society (1985 June).

[4] N.F. Harrison, "AECL's Experimental Fuel and Materials Test Loops in NRU", presented at the IAEA Technical Meeting on In-Pile Testing and Instrumentation for Development of Generation-IV Fuels and Materials, Halden, Norway, 2012 August 21-24 (AECL report CW-124000-CONF-016).

[5] M.D. Atfield, "Calculation of Power Distributions for Experimental Bundles in the NRU Loops", Proceedings of the 22nd CNS Conference, AECL-CONF-638 (2004).

[6] D. Altiparmakov, "New Capabilities of the Lattice Code WIMS-AECL", Proceedings of the International Conference on Reactor Physics, PHYSOR 2008, Casino-Kursaal Conference Center, Interlaken, Switzerland (2008 September 14-19).

[7] P.L. Purdy, A.M. Manzer, R.H. Hu, R.A. Gibb and E. Kohn, "Assessments of Sheath Strain and Fission-Gas Release Data from 20 Years of Power Reactor Fuel Irradiations", Proceedings of the 5th International Conference on CANDU Fuel, Toronto, Ontario, Canada, Vol. 2, 134-14 (1997 September 21-25).

[8] M.R. Floyd, "Extended Burnup CANDU Fuel Performance," Proceedings of the 7th International Conference on CANDU Fuel, Kingston, Ontario, Canada, Vol. 2, 5A-1 (2001 September 23-27).

[9] M.R. Floyd, J. Novak, and P.T. Truant, "Fission-Gas Release in Fuel Performing to Extended Burnups in Ontario Hydro Nuclear Generating Stations", Proceedings of the IAEA Technical Committee Meeting on Fission-Gas Release and Fuel Rod Chemistry related to Extended Burnup, Pembroke, Canada, IAEA-TECDOC-697, pp. 53-59, (1992).