ADVANCED FUEL CYCLE DEVELOPMENT AT CHALK RIVER LABORATORIES

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Abstract

Chalk River Laboratories (CRL) has a mandate from the Canadian government to develop nuclear technologies that support generation of clean, safe energy. This includes the development of advanced nuclear fuel technologies to ensure sustainable energy sources for Canadians.

The Fuel Development Branch leads CRL's development of advanced nuclear-reactor fuels. CRL capabilities include fuel fabrication development, irradiation testing, post-irradiation examination (PIE), materials characterization and code development (modeling). This paper provides an overview of these capabilities and describes recent development activities that support fuel-cycle flexibility in heavy-water reactors. This includes a review of irradiation testing and PIE for mixed-oxide, thoria, high-burnup UO_2 and low-void reactivity fuels and burnable neutron absorbers. Fabrication development, material characterizations and modeling associated with these tests are also described.

1. Introduction

Chalk River Laboratories (CRL) has facilities that enable fabrication, irradiation, post-irradiation examination (PIE) and materials characterization of various fuel types that can be deployed in pressurized heavy-water reactors (PHWRs). This includes $UO_{2,}$ thoria, MOX (mixed-oxide; plutonium-bearing) and burnable neutron absorber (BNA) materials. Collaboration with universities, industry and other laboratories is integral to CRL's nuclear science and technology mandate.

The Chalk River Advanced Fuel Technologies (CRAFT) laboratories include facilities for the development of advanced ceramic fuel pellets, in addition to the fabrication of fuel elements and bundles for testing in the National Research Universal (NRU) reactor. MOX fuels are fabricated in the Recycle Fuel Fabrication Laboratories (RFFL) that contains glove-boxes for plutonium (Pu) handling [1].

Irradiation tests are conducted in the U1 and U2 loop facilities of the NRU reactor at CRL. These loop facilities contain three vertical test sections that can accommodate a total of eighteen CANDU-geometry fuel bundles (six per test section). The axial thermal neutron flux in each test section is nominally cosine-shaped, and can be varied by up to 25%, depending on the reactivity of driver fuel adjacent to the loop sites. The combination of varied loop fuel enrichment and thermal neutron flux is used to simulate powers achieved in commercial power reactors. Coolant temperatures are typically in the range of 260-305 ^{o}C at pressures of 9.5-10.9 MPa. During 2009-2010, maintenance was conducted on the NRU reactor vessel, positioning it for operation into the next decade. The U2 loop (two test sections) was upgraded during 2005-2011 to support future operation.

Underwater visual inspections of fuel irradiated in the U1 and U2 loops take place in the NRU irradiated fuel bay. Detailed PIE occurs in the CRL hot cell facilities. Typical non-destructive examinations include:

- visual examination of bundles and disassembled elements
- dimensioning of bundles and elements (sheath diameter, element bow, element length and endplate distortion)
- axial gamma scanning (illustrates axial burnup gradients, end-of-life fuel power gradients, end-flux peaking, fission-product segregation and pellet dish filling)

Typical destructive examinations include:

- internal gas analyses (determining internal gas volume and composition, % fission-gas release (FGR) and internal void volume)
- sheath hydrogen (H) and deuterium (D) analyses
- sheath metallography (examining microstructure and oxidation)
- pellet ceramography (examining microstructure and determining grain growth)
- alpha/beta-autoradiography (typically used on MOX fuels)
- chemical burnup measurement
- root-cause failure determination

The Fuel Development Branch conducts investigations into fuel materials properties and behaviour using facilities at CRL and other laboratories and universities. Typical ceramic fuel pellet investigations include:

- mechanical properties
- oxidation behaviour
- phase diagrams
- thermal conductivity
- leaching behaviour

Mechanical properties of fuel sheath alloys are investigated, including the effects of microstructure, hydrogen/deuterium pickup and irradiation effects.

Historically, CRL fuel modelling efforts have focused on the behaviour of UO_2 fuel operating in CANDU-PHWRs (including normal operating conditions and design-basis accidents). Effort to model other fuel types is on-going to support the development of advanced fuel cycles.

2. UO_2 Fuel

In 2005, a review of irradiations and PIE supporting the development of advanced CANDU UO_2 fuel technology was conducted [2]. This comprised a description of advanced CANDU UO_2 fuel technology development during 1995-2005, including (i) development of the 43-element CANFLEX fuel bundle (developed for extended-burnup applications with reduced element powers), (ii) extended-burnup UO_2 technology (including investigations into the effect of pellet geometry on performance), and (iii) low-void-reactivity fuel technology (UO_2 fuel with dysprosium BNA added to the centre of the bundle to reduce coolant void reactivity).

During 2005-2011, advanced UO_2 fuel initiatives focused on technologies that can be applied to slightly-enriched uranium (SEU) fuel cycles and designs for deployment in enhanced CANDU-PHWR reactors. PIE was completed on BDL-437 bundle 'AJM' that achieved an outer element burnup of 520 MWh/kgU (22 MWd/kgU). AJM contained SEU pellets in a CANFLEX bundle geometry and sustained a power ramp from 42 kW/m to 65 kW/m at a burnup of 110 MWh/kgU (5 MWd/kgU). A test plan was developed for the DME-225 experiment, with the objective of demonstrating the effect of varied pellet geometry under power-ramp conditions (mitigation of stress-corrosion cracking (SCC)). Fabrication of a demountable fuel bundle (permitting element substitution during irradiation [2]) for the DME-225 experiment was completed in 2011; irradiation testing is expected to commence in 2012.

Considerable effort has been focused on understanding the effect of coolant ingress in defected UO_2 fuels. CRL has applied techniques to conduct measurements on pellets from fuels that experienced through-wall sheath defects during irradiation. Quantitative O/M (oxygen-to-metal atom ratio) measurements have been performed using a coulometric titration technique on fuel samples obtained by core drilling [3]. A shielded scanning electron microscope was used to determine the extent of oxidation at various locations within fuel pellets from defected fuels. CRL collaborates with the Royal Military College (RMC; Kingston, Ontario, Canada) to model the behaviour of UO_2 fuel; recently, a thermochemical model was developed and validated using coulometric titration data obtained at CRL [4]. In addition, RMC and CRL collaborate with industry partners to develop defected fuel diagnostic tools [5].

3. MOX Fuel

The development of various types of MOX fuels has been on-going at CRL for over 40 years [6, 7].

The BDL-419 experiment involved the fabrication, irradiation and PIE of fifteen $(U, Pu)O_2$ fuel bundles having the same geometry as those currently used in commercial CANDU-PHWRs [7]. Thirteen of the bundles have completed irradiation to outer element burnups up to 828 MWh/kgHE (35 MWd/kgHE). Bundle 'ADP' was irradiated in the NRU loops at a beginning-of-life (BOL) outer element power of 63 kW/m to a burnup of 557 MWh/kgHE (23 MWd/kgHE) when it experienced in-core failure (Figure 1). Preliminary examinations measured high internal gas volumes (up to 145 mL), indicating that the probable failure mode was SCC due to internal gas overpressurization (caused by high fission-gas release). UO_2 fuels with similar power histories have also experienced sheath SCC resulting from high internal fission-gas pressures [8]. Destructive PIE of selected elements from bundle ADP is expected to be completed in 2012. The objective is to contrast the behaviour of ADP with UO_2 fuel having similar power histories [8]. Two other BDL-419 bundles ('ABG' and 'ADR') presently have burnups of 800-900 MWh/kgHE (33-38 MWd/kgHE), and are continuing their irradiation in the NRU loops to > 1000 MWh/kgHE (42 MWd/kgHE).

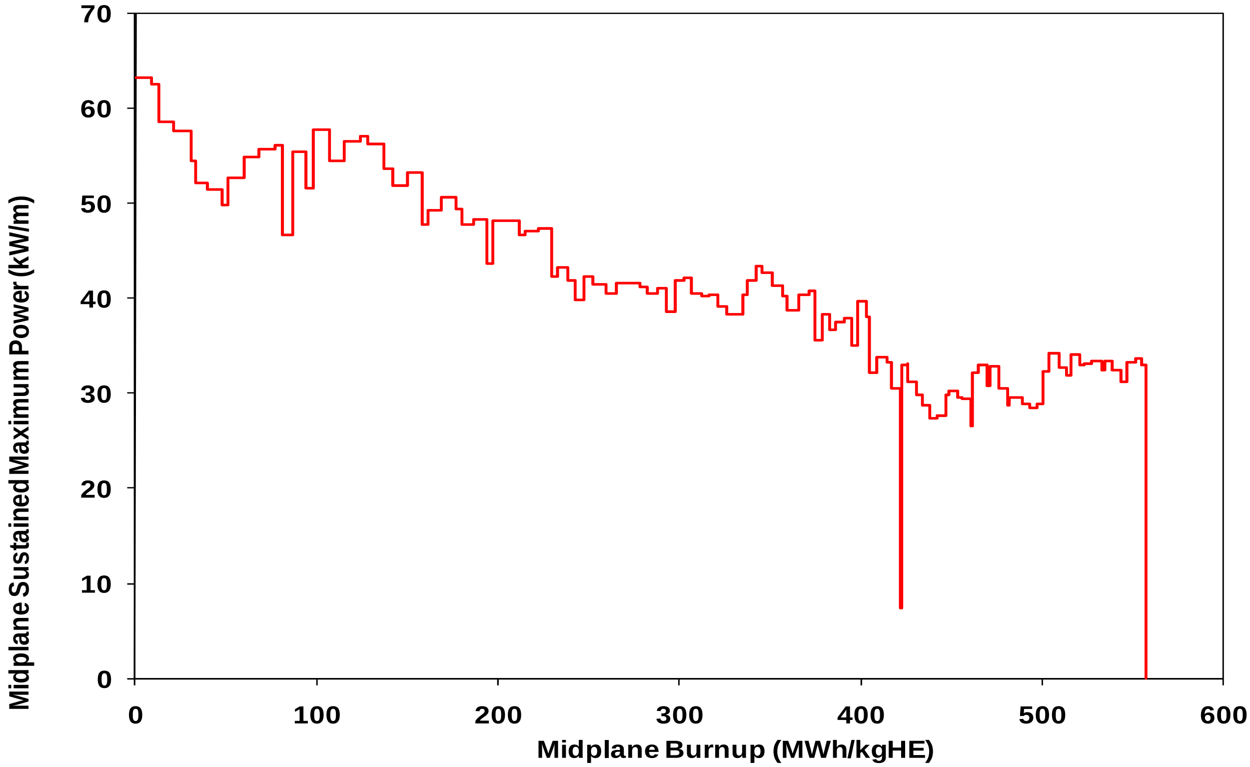

Figure 1 Power history for BDL-419 MOX bundle 'ADP' outer elements that experienced sheath failure due to high internal fission gas pressure.

The BDL-446 experiment was designed to investigate the effect of Pu homogeneity on fuel performance in (U, Pu)O_2 pellets produced at CRL using different fabrication techniques [9, 10]. The results of the irradiation and subsequent PIE were reported in 2010 [11]. FGR in the fuel containing pure Pu particles (similar to that in BDL-419) was higher than that exhibited by more homogeneous mixtures of Pu. Pellets characterized by microstructure with PuO_2 and UO_2 in solid solution exhibited similar FGR to "master mix" pellets containing Pu-rich particles (30 wt. % Pu). This investigation has demonstrated the effect of MOX fuel fabrication technology (i.e., pellet Pu homogeneity) on fuel performance. Further studies are envisioned to correlate fuel performance parameters with variations in master mix particle Pu concentration.

The behaviour of (U, Pu)O_2 MOX fuel has generally been observed to be similar to that of UO_2 fuel [7, 11]. As a result, existing UO_2 models have been used to approximate the behaviour of MOX fuel. MOX-specific models are envisioned, to account for differences in the two fuel types (e.g., effect of Pu homogeneity).

4. Thoria-Based Fuel

AECL has more than 50 years of experience with thoria-based fuel irradiations conducted in the Nuclear Power Demonstration (NPD), Douglas Point, WR1 (Whiteshell), National Research Experimental (NRX) and NRU reactors [12]. This includes irradiations of natural thoria (ThO_2) and thoria blended with fissile uranium or plutonium; i.e., (Th, U)O_2 and (Th, Pu)O_2. Many of the irradiations were performed with fuel pellets characterized by a granular, inhomogeneous microstructure – this fuel was found to exhibit performance comparable to that of UO_2 (e.g., FGR). During the 1990s, CRL successfully completed an initiative to achieve improved (homogeneous) microstructures in thoria-based fuels [13]. The performance of this fuel was demonstrated in the DME-221 experiment to be superior to that of similarly-operated UO_2 fuel.

Table 1 Operating History and Fission-Gas Release Data for DME-221 Elements

Fuel Type	Maximum Power (kW/m)	Burnup (MWh/kgHE)	% FGR
ThO_2	36-37	361-375	0.05-0.06
ThO_2	40-41	596-619	0.10
ThO_2 + 1.0% U-235	40-41	499-526	0.06-0.07
ThO_2 + 1.0% U-235	46	839	1.2
ThO_2 + 1.5% U-235	55	594	0.08-0.11
ThO_2 + 1.5% U-235	52-54	903-929	1.5-2.8

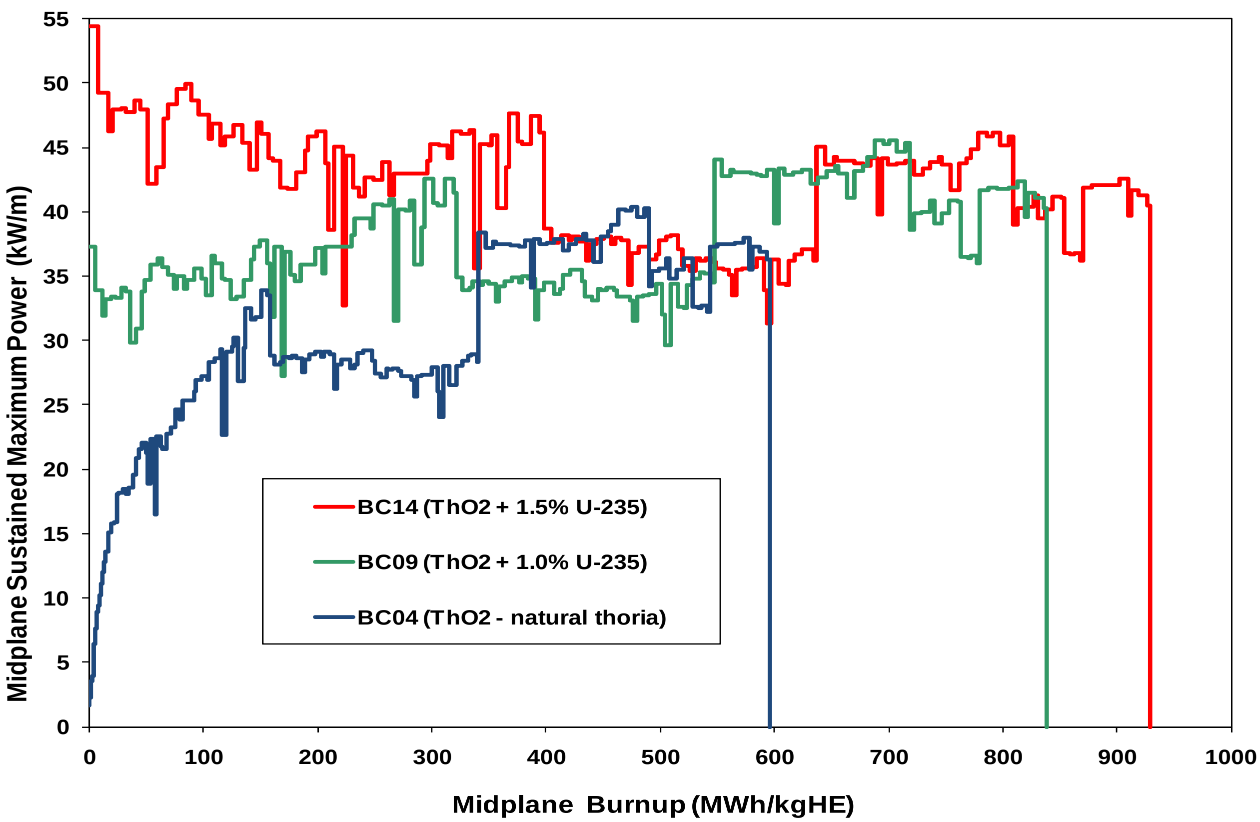

Figure 2 Power histories for thoria-based DME-221 elements 'BC04' (ThO_2), 'BC09' (ThO_2 + 1.0% U-235) and 'BC14' (ThO_2 + 1.5% U-235).

The DME-221 experiment includes the irradiation of eighteen CANDU-geometry fuel elements in a demountable configuration. 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.% U-235 (in total heavy elements), while the remaining six contain 1.5 wt.% U-235. The elements were irradiated to maximum powers of 36-55 kW/m (Table 1). Power histories for three DME-221 elements (representing each fuel type at high burnup) are shown in Figure 2. PIE has recently been completed on twelve DME-221 elements that achieved burnups of 361-929 MWh/kgHE (15-39 MWd/kgHE). FGR ranged from 0.05% (at 37 kW/m and 375 MWh/kgHE; Table 1) to 2.8% (at 54 kW/m and 929 MWh/kgHE; Table 1). These values are significantly lower than those experienced by similarly operated UO_2 fuels, especially above 500 MWh/kgHE (21 MWd/kgHE; Figure 3). This is attributed to the higher thermal conductivity of thoria (relative to that of UO_2). Six DME-221 elements are continuing their irradiation to burnups of 1000-1500 MWh/kgHE (42-63 MWd/kgHE). This is a major achievement in the development of thoria-based fuel technology, which has great potential for deployment in PHWRs [12, 14].

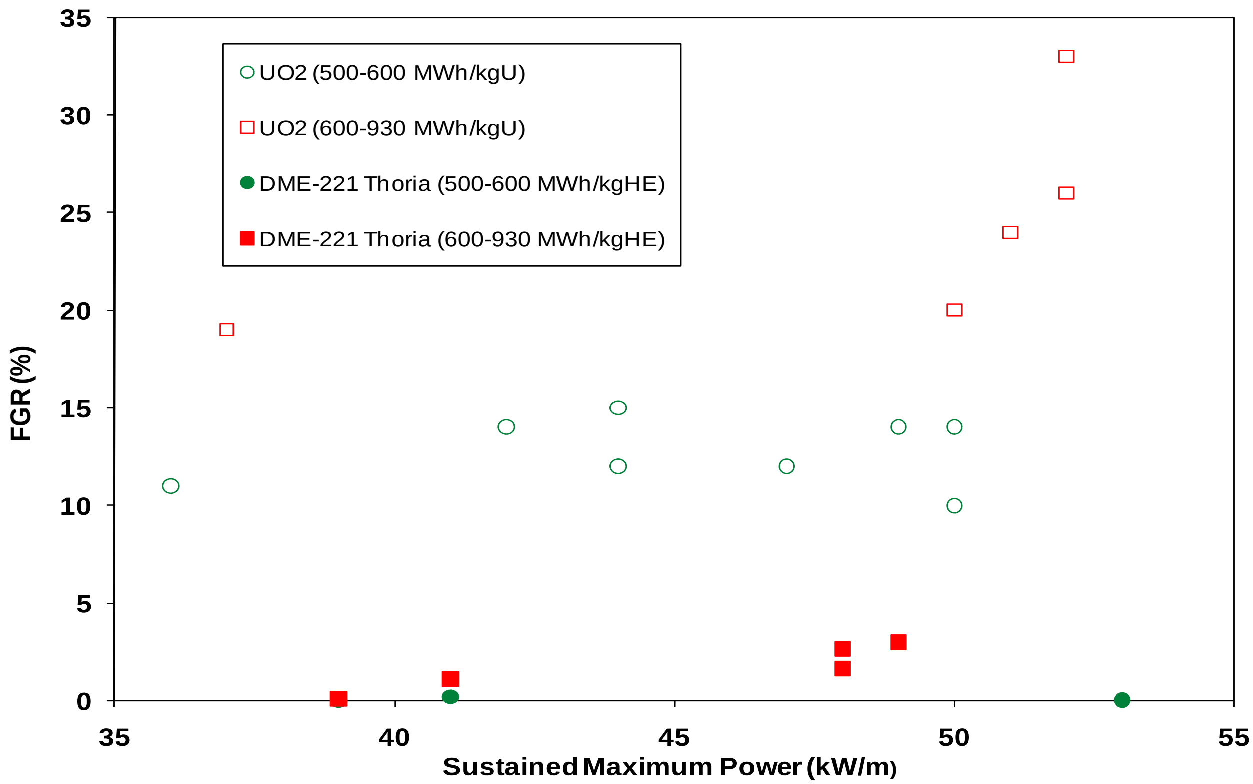

Figure 3 Fission-gas release (FGR) for UO_2 and DME-221 thoria fuels at burnups > 500 MWh/kgHE. Note low FGR for thoria (closed points) relative to that of UO_2 (open points).

The BDL-422 experiment involved irradiation testing of $(Th, Pu)O_2$ fuel bundles having the same geometry as those currently used in commercial CANDU-PHWRs. The fuel pellets contained 1.53 wt.% Pu in $(Th, Pu)O_2$. Three BDL-422 fuel bundles ('ADC', 'ADE' and 'ADF') were irradiated at maximum powers of 52-67 kW/m and burnups of 451-856 MWh/kgHE (19-36 MWd/kgHE; Table 2 and Figure 4); these exhibited performance characteristics superior to that of UO_2 fuel [15, 16]. This included benign FGR (1-5%) and grain growth, owing to the high thermal conductivity of thoria (relative to that of UO_2). Two BDL-422 fuel bundles ('ADA' and 'ADD') were irradiated at maximum powers of 54-73 kW/m and burnups of 1082-1181 MWh/kgHE (45-49 MWd/kgHE; Table 2 and Figure 4); these exhibited high FGR (20-33%), similar to that observed in UO_2 fuel irradiated at maximum powers of 50-60 kW/m at burnups of 500-900 MWh/kgU (21-38 MWd/kgHE) [8]. Columnar grain growth was not observed in the BDL-422 fuel, unlike UO_2 fuel irradiated at

> 60 kW/m and (U, Pu)O_2 fuel irradiated at > 55 kW/m [7]. The as-fabricated grain size of the BDL-422 fuel pellets was relatively small (3-4 μm) compared to typical grain sizes in UO_2 fuel (5-10 μm). This, in combination with the high operating power, appears to have contributed to high FGR at burnups > 1000 MWh/kgHE (42 MWd/kgHE). A future high-burnup (Th, Pu)O_2 irradiation is envisioned to investigate the effect of pellet microstructure (including initial grain size and Pu homogeneity) on performance. In addition, irradiation testing and materials characterization of thoria-based fuels with higher concentrations of initial U/Pu (> 10%) is envisioned to support other fuel cycles and reactor concepts (e.g., super-critical water reactor) [17]. Materials properties measurements are also envisioned to study the effect of U/Pu addition.

Table 2 Operating History and Fission-Gas Release Data for BDL-422 (Th, Pu)O_2 Outer Elements

Fuel Bundle	Maximum Power (kW/m)	Burnup (MWh/kgHE)	% FGR
ADC	67	451	5.3
ADE	64	597	1.2
ADF	52	856	2.8
ADA	54	1181	26.2-32.8
ADD	73	1082	19.6-26.9

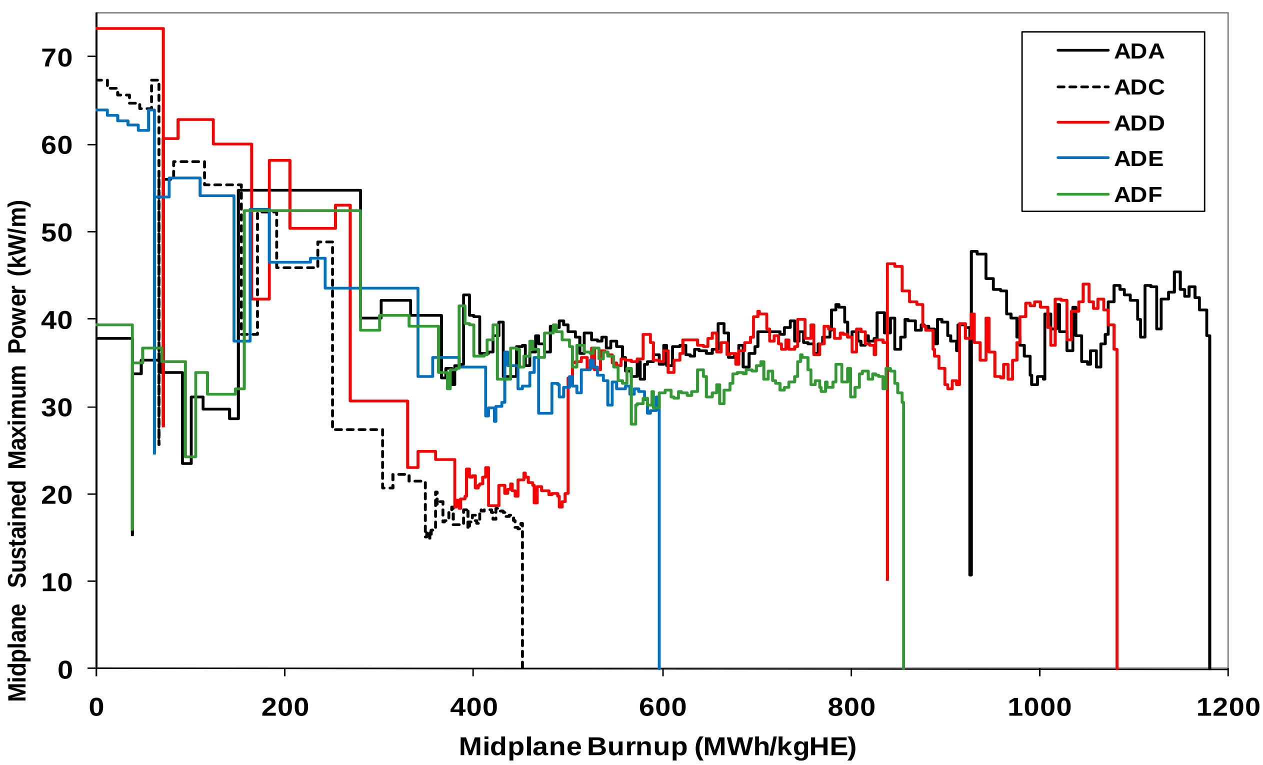

Figure 4 Outer element power histories for BDL-422 (Th, Pu)O_2 bundles ‘ADA’, ‘ADC’, ‘ADD’, ‘ADE’ and ‘ADF’.

Existing UO_2 models can generally be used to bound the behaviour of thoria-based fuel. Recently, work has commenced at CRL on thoria-specific fuel performance algorithms; notably, one that models FGR in thoria-based fuels.

CRL thoria irradiation experience includes several fuel elements that experienced through-wall failure. Work is in progress to summarize this experience and contrast it with defected UO_2 fuel behaviour. Unlike UO_2, ThO_2 pellets do not oxidize as a result of post-defect coolant ingress. Consequently, thoria pellet erosion and release to the coolant is lower. Fission-product release to the coolant is also lower in thoria-based fuel (compared to UO_2) due to its lower operating temperatures (higher thermal conductivity) and resistance to oxidation. Oxidation of UO_2 fuels results in lower thermal conductivity, which increases operating temperature and fission-product release.

5. Low-Void Reactivity Fuel and Burnable Neutron Absorbers

In 2005, irradiations and PIE supporting the development of low-void-reactivity fuel (LVRF) technology (UO_2 fuel with dysprosium (Dy) BNA added to the centre of the bundle to reduce coolant void reactivity) were conducted [2]. Twenty-one demountable fuel elements having UO_2 fuel pellets with varying Dy contents of 0-15% were tested at powers that gradually increased to a maximum of 24 kW/m and maximum burnups of 249 MWh/kgU (10 MWd/kgU). In addition, four CANDU-geometry bundles having SEU pellets in the two outer rings and UO_2 + 2% Dy in the inner elements of the bundle were tested. Performance of the Dy-bearing fuel elements was benign, including FGR < 0.1%.

Table 3 Sheath Strain Data for DME-224 Elements After 21 Days of Irradiation

Element Identity	Irradiation Type* (neutron energy characteristic)	Pellet Composition (Vol.% $Dy_2O_3/Gd_2O_3/Y_2O_3/ZrO_2$)	Average Sheath Strain (%)
BK01	Outer (high thermal neutron)	Dy+Gd+Y (12.0/12.0/15.3/60.7)	-0.02
BK03	Outer (high thermal neutron)	Dy+Gd (12.0/12.0/0.0/76.0)	+0.11
BK05	Outer (high thermal neutron)	Y-Stabilized (0.0/0.0/38.3/60.7)	-0.04
BK07	Outer (high thermal neutron)	Dy-Only (55.0/0.0/0.0/45.0)	-0.01
BK09	Inner (high fast neutron)	Dy+Gd+Y (12.0/12.0/15.3/60.7)	-0.16
BK11	Inner (high fast neutron)	Dy+Gd (12.0/12.0/0.0/76.0)	-0.12
BK13	Inner (high fast neutron)	Dy-Only (55.0/0.0/0.0/45.0)	-0.13

* location (by ring) in a 42-element demountable bundle resulting in different neutron energies

More recently, effort has been focused on the development of BNA technology associated with pellets made of non-fissile material. The DME-224 test includes fourteen demountable elements containing non-fissile ZrO_2 (zirconia) pellets with various compositions of BNA, including Dy_2O_3 (dysprosia) and Gd_2O_3 (gadolinia). Some of the pellets also contain Y_2O_3 (yttria) to stabilize the zirconia lattice structure under irradiation (Table 3). By placing these elements in both the outer and inner rings of a 42-element demountable bundle [2], the elements are exposed to different ratios of fast vs. thermal neutrons. The DME-224 irradiation test commenced in 2009 and was interrupted by a 14-month NRU shutdown and upgrades to the NRU loops during 2009-2011. Testing is expected to resume in 2012. To date, seven DME-224 elements have been irradiated for 47 days. Interim non-destructive examination of the seven elements took place after 21 days and included visual inspection and profilometry (diameter measurements). The elements were found to be intact and in good condition. Diameter changes (strain) were small and compressive except for one element that exhibited slightly tensile strains (Table 3). The duration of the DME-224 irradiation is planned to be 1000 days with interim examinations occurring after approximately 133, 277 and 565 days. Pending the outcome of non-destructive examinations at these intervals, some of the seven elements currently being irradiated may be replaced and subject to destructive examination. Considerable effort has also been applied at CRL to characterizing the material properties of ZrO_2 + BNA materials [18]. An understanding of the fundamental properties of these materials is prerequisite to modeling their irradiation behaviour.

6. Fuel Clad Technology Initiatives

The successful deployment of advanced fuel cycles is dependent on the fuel design, of which the clad (sheath) material an intrinsic part. CRL continues to undertake initiatives to understand the mechanical properties of zirconium alloys used in fuel cladding (e.g., Zircaloy-2, Zircaloy-4 and Zr-Nb), including the effect of hydrides [19-21]. These initiatives include collaboration with universities and industry partners. Work is also underway on advanced fuel cladding alloys to investigate material properties, including the effects of standard sheath joining methods. Future work is planned to investigate advanced sheath joining techniques (e.g., alternate braze material, resistance brazing, and high-frequency resistance welding). These advanced joining techniques seek to improve joint microstructure and properties for both advanced sheath materials and standard alloys, and improve fabrication worker safety by eliminating or reducing the use of hazardous fabrication materials (e.g., beryllium).

7. Summary

CRL has a vision to generate, access and share nuclear fuel science and technology, and work with others to apply and exploit it to produce clean, sustainable energy that is economical, safe and secure. The Fuel Development Branch continues to undertake initiatives in the development of advanced nuclear fuel technologies that can be deployed in PHWRs, as well as other reactor designs/concepts. Significant initiatives have been undertaken at CRL for UO_2, MOX, thoria-based, LVRF and BNA technologies, and are envisioned for the future (with respect to fabrication technology, irradiation testing, post-irradiation examination, materials characterization and modelling). These initiatives are enabled by CRL's facilities and collaboration with industry, universities and other laboratories.

8. Acknowledgements

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