CANDU FUEL:
Evolution towards Optimal Performance

PERFORMANCE OF HIGH BURNUP MOX FUEL

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ABSTRACT

Canadian Nuclear Laboratories (CNL; formerly AECL) has more than 40 years of experience in developing (U, Pu)O_2 mixed-oxide (MOX) fuel that supports pressurized heavy-water reactor fuel technology. The largest of CNL's MOX fuel experiments, BDL-419, comprises fifteen bundles irradiated in the NRU reactor. The pellets in these bundles contain 0.5 wt.% Pu in total heavy elements. To date, BDL-419 bundles have achieved burnups of up to ~ 900 MWh/kgHE (38 MWd/kgHE). Most recently, two BDL-419 bundles (ADM and ADP) have completed post-irradiation examination in the CNL hot cell facilities located at Chalk River. The outer elements of bundle ADM achieved beginning-of-life powers > 50 kW/m and burnups > 800 MWh/kgHE (33 MWd/kgHE). The outer elements of bundle ADP achieved beginning-of-life powers > 60 kW/m and burnups > 500 MWh/kgHE (21 MWd/kgHE); two of the outer elements experienced sheath stress-corrosion cracking failure as a result of high internal gas pressure (high fission-gas release).

This paper presents the performance assessment results of MOX BDL-419 bundles ADP and ADM and compares them with other CNL MOX fuel tests and UO_2 fuel having similar operating histories and designs.

Introduction

Canadian Nuclear Laboratories (CNL; formerly AECL) has more than 40 years of experience in developing (U, Pu)O_2 mixed-oxide (MOX) fuel that supports pressurized heavy-water reactor (PHWR) fuel technology [1-7]. This has included several CANDU-type irradiation tests having 19, 37 and 43-element geometries.

The largest of CNL's MOX fuel experiments, BDL-419, comprises fifteen 37-element geometry bundles irradiated in the NRU reactor [1-3, 7]. The pellets in these bundles contain 0.5 wt.% Pu in total heavy elements (Pu + natural uranium). To date, BDL-419 bundles have achieved burnups of up to ~ 900 MWh/kgHE (38 MWd/kgHE). Most recently, two BDL-419 bundles (designated "ADM" and "ADP") have completed post-irradiation examination in the CNL hot cell facilities located at Chalk River. These bundles incorporated three different coatings on the inner bore of alternating outer elements to mitigate stress corrosion cracking (SCC) of the Zircaloy-4 sheath. The outer elements of bundle ADM achieved beginning-of-life (BOL) powers of 54 kW/m and burnups of 865 MWh/kgHE (36 MWd/kgHE); none of the elements experienced failure. The outer elements of bundle ADP achieved BOL powers of 66 kW/m and burnups of 579 MWh/kgHE (24 MWd/kgHE); two of the outer elements experienced sheath SCC failure as a result of high internal gas pressure (high fission-gas release) [7].

This paper presents the performance assessment results of MOX BDL-419 bundles ADP and ADM and compares them with other CNL MOX fuel tests and UO_2 fuel having similar operating histories and designs.

1. Fuel Design and Fabrication

BDL-419 bundles ADM and ADP were of standard CANDU 37-element geometry, with the central element removed to facilitate irradiation in the U1 and U2 loops of the NRU reactor. The $(U, Pu)O_2$ pellets in bundles ADM and ADP were fabricated in CNL's Recycle Fuel Fabrication Laboratories (RFFL) and contained 0.5 wt% Pu (in total heavy elements) that was low-intensity blended with natural UO_2. The PuO_2 used for fabrication was passed through a mesh screen to ensure a maximum particle size of 63 µm. Alpha autoradiography of the as-fabricated pellets revealed a uniform distribution of distinct Pu particles within the UO_2 estimated to be 10-40 µm in diameter. The average density of the pellets was 10.54 g/cm^3, average grain size was 6-8 µm, and average pellet length was 12.1 mm with a length-to-diameter (L/D) ratio of 1. One end of the fuel elements was resistance welded, while the other was TIG-welded in the RFFL following pellet loading.

The inner bore of the inner and intermediate elements of bundles ADM and ADP were coated with standard graphite DAG-154 to provide mitigation for stress-corrosion cracking (SCC). The outer elements of the bundles were coated alternately with DAG-154, ES-242 and siloxane. ES-242 is similar to DAG-154, except that it is slurried in water prior to application (as opposed to alcohol slurry for DAG-154). Siloxane is commercially available vacuum grease. ES-242 and siloxane were investigated by AECL as alternatives to DAG-154 [8, 9]; both were shown to offer similar SCC mitigation properties to DAG-154.

2. Irradiation History

Bundles ADM and ADP were irradiated in the U1 and U2 loops of the NRU reactor during 1986-2001. Table 1 summarizes the power history data for the outer, intermediate and inner elements for both bundles. Outer element power histories are shown in Figure 1. Both bundles experienced declining power histories and did not experience significant power ramps. Maximum powers were achieved at the beginning of the irradiation. A wide range of powers and burnups were achieved by the outer, intermediate and inner elements (32-66 kW/m and 369-865 MWh/kgHE (15-36 MWd/kgHE), respectively).

The irradiation of bundle ADP was terminated because it experienced failure. Bundle ADM did not experience failure.

Table 1 Power History Data for BDL-419 (U, Pu)O_2 Bundles ADM and ADP

Fuel Bundle Identity	Element Type (Ring)	Burnup (MWh/kgHE) {MWd/kgHE}	Maximum Sustained Linear Power (kW/m)
ADM	Outer	865 {36}	54
	Intermediate	676 {28}	37
	Inner	598 {25}	32
ADP	Outer	579 {24}	66
	Intermediate	439 {18}	42
	Inner	369 {15}	32

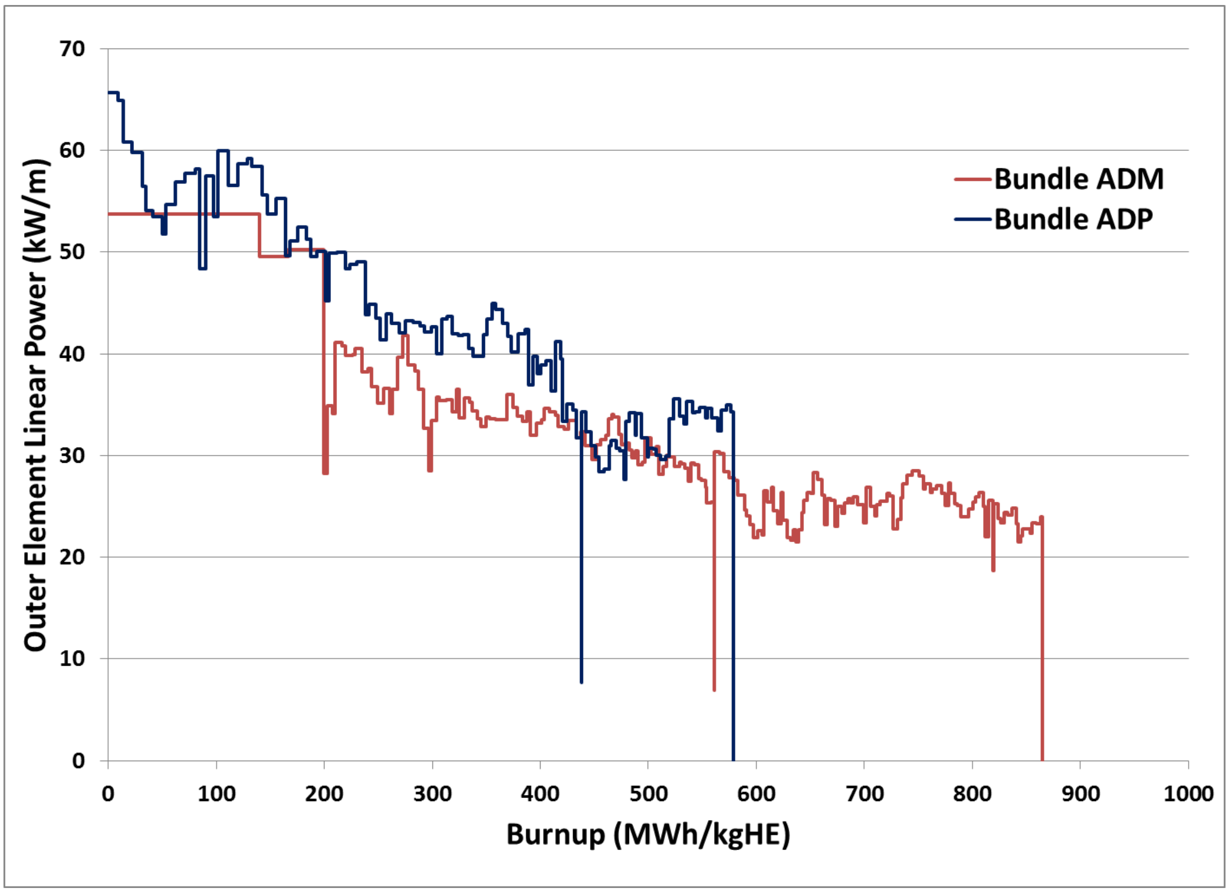

Figure 1 Outer Element Power History for Bundles ADM and ADP

3. Post-Irradiation Examination

Post-irradiation examination (PIE) of bundles ADM and ADP took place in CNL's hot cell facilities. The highlights are discussed below.

3.1 Bundle ADM

Visual examination of the elements of bundle ADM revealed them to be free of defects. Midpellet (MP) residual sheath strains were in the range of -0.1% to +0.1% for the outer elements and -0.6% to -0.2% for the intermediate and inner elements. Pellet interface (PI) residual sheath strains were in the range of +0.2% to +0.4% for the outer elements and -0.4% to 0.0% for the intermediate and inner elements. Outer element sheath strain did not exhibit any dependence on coating type. These strains are within the range of normally operated CANDU UO_2 fuel [10].

Internal gas volumes for the outer elements ranged from 31-51 mL (average 36 mL) and exhibited no dependence on coating type. Internal gas volumes in the intermediate and inner elements were < 7 mL. These volumes are well below the 100 mL gas over-pressurization failure threshold for conventional 37-element CANDU UO_2 elements [11]. Fission-gas release (FGR) in the outer elements ranged from 4% to 7% and exhibited no dependence on coating type. FGR in the intermediate and inner elements was < 1%.

Equiaxed grain growth was observed in ADM. Oxide on the inner bore of the sheath was observed in DAG-154 coated outer elements in thicknesses up to 12 μm. Siloxane and ES-242 coated outer elements did not exhibit a definitive oxide layer (<1 μm in thickness) resembling that observed in DAG-154 coated elements.

3.2 Bundle ADP

Visual examination of the elements of bundle ADP revealed through-wall SCC defects in the sheaths of outer elements 7 and 18, both coated with DAG-154 (Figure 2). MP residual sheath strains ranged from +0.1 % to +0.5 % for outer elements coated with ES-242 or siloxane, +1.4 % to +2.1 % for outer elements coated with DAG-154, and -0.3 % to -0.2 % for inner and intermediate elements (all coated with DAG-154). Pellet interface residual sheath strains ranged from +0.3 % to +0.8 % for outer elements coated with ES-242 or siloxane, +1.7 % to +2.4 % for outer elements coated with DAG-154, and -0.2 % to -0.1 % for inner and intermediate elements (all coated with DAG-154).

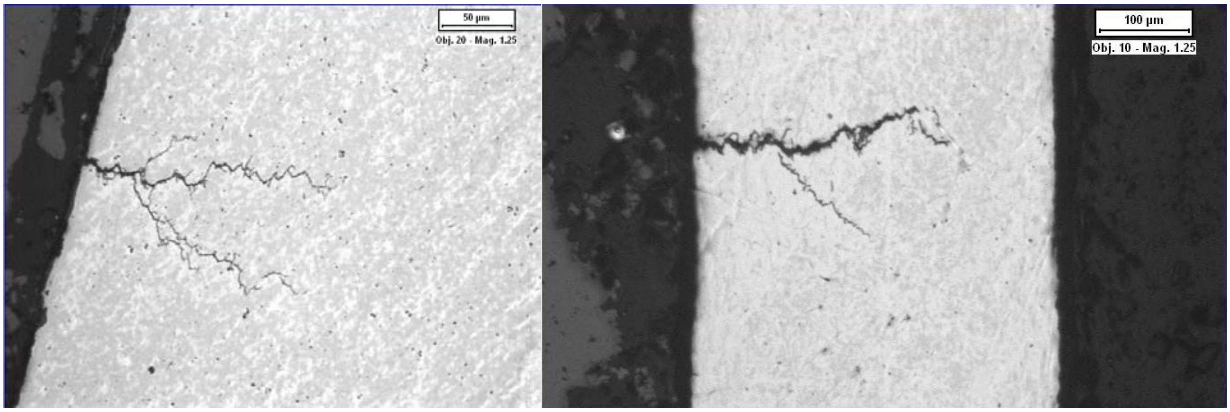

Figure 2 SCC Defects in DAG-154 Coated Elements ADP-7 (left) and ADP-18 (right).

Internal gas volumes of intact outer elements coated with ES-242 or siloxane ranged up to 98 mL, below the 100 mL defect threshold for 37-element CANDU UO_2 elements [11]. For intact outer elements coated with DAG-154, high internal gas volumes were measured (136-155 mL). Fission-gas release in outer elements coated with ES-242 ranged from 14-15%. In contrast, intact outer elements coated with DAG-154 had FGR ranging from 35-37 %. Fission-gas release was not calculated for ADP elements with a siloxane coating, but is expected to be similar to elements coated with ES-242 based on similar gas volumes.

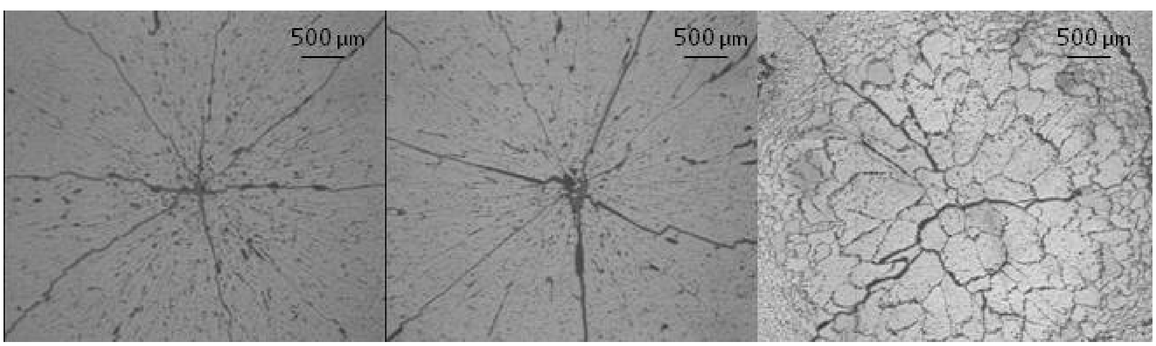

Figure 3 Typical Pellet Centre Grain Morphology Observed in ADP-4 (ES-242, Left; columnar grains), ADP-8 (Siloxane, Middle; columnar grains) and ADP-6 (DAG-154, Right; typical grain size > 100 µm).

Grain growth in pellets of DAG-154 coated elements differed from those coated with ES-242 or siloxane. DAG-154 coated elements exhibited an R/A ratio[1] of 0.84, while ES-242 and siloxane coated outer elements exhibited R/A values of 0.71 and 0.72, respectively (indicative of lower fuel temperatures than the DAG-154 coated elements). Bundle ADP outer elements coated with siloxane and ES-242 experienced columnar grain growth (Figure 3), similar to that observed in other DAG-154 coated BDL-419 fuel that operated at a power of 56 kW/m [3]. In contrast, ADP outer elements coated with DAG-154 revealed very large grains at the pellet centre (in excess of

[1] R/A = the ratio of the observed grain growth from the pellet centre (R) to the pellet periphery (A)

100 μm; Figure 3), similar to that observed in other DAG-154 coated BDL-419 fuel that operated at 59 kW/m [3].

Sheath oxide on the inner bore of ADP outer elements had thicknesses up to 9 μm, with no apparent relation to coating type.

4. Discussion

4.1 MOX Fuel Performance

The main objective of the BDL-419 experiment is to assess the performance of MOX 37-element geometry fuel bundles irradiated to extended burnup under CANDU power reactor conditions. Relevant to this discussion are other BDL-419 bundles for which PIE has been completed (ABB, ABC, ABD, and ABE) [2] [3], and other CANDU MOX fuel experiments (BDL-446 bundle AMC, BDL-447 bundle AMD, and NPD-40 bundle KB), all of which were coated with DAG-154. Irradiation history and fabrication data for these bundles are shown in Table 2. In general, the performance of MOX CANDU fuel bundles is considered to be similar to UO_2 CANDU fuel with similar power histories [2, 3, 11].

Table 2 Data for Selected MOX CANDU Bundles Irradiated in NRU and NPD

Experiment	Bundle	Maximum Sustained Linear Power (kW/m)	Burnup (MWh/kgHE) {MWd/kgHE}	Notes	Ref
BDL-419	ADM[1]	54	865 {36}	Varied sheath coatings	-
BDL-419	ADP[1]	66	579 {24}	Varied sheath coatings	-
BDL-419	ABB[1]	50-55	449-490 {19-20}	DAG-154 coating only	[3]
BDL-419	ABC[1]	60-62	538-547 {22-23}	DAG-154 coating only	[3]
BDL-419	ABD[1]	63-65	312-319 {13}	DAG-154 coating only	[3]
BDL-419	ABE[1]	54-56	346-360 {14-15}	DAG-154 coating only	[3]
BDL-446	AMC[2]	51-53	517 {22}	Varied Pu homogeneity	[6]
BDL-447	AMD[1]	46	250-294 {10-12}	Weapons-derived Pu	[5]
NPD-40	KB[3]	49	1181 {49}	Irradiated in NPD	[1]

1 – 37 element geometry with centre element removed for irradiation in NRU
2 – 43 element geometry with centre element removed for irradiation in NRU
3 – 19 element geometry

4.1.1 MP Residual Sheath Strains

Bundle ADM had MP residual sheath strains of -0.1% to +0.1% in outer elements, with no apparent differences between coating types. These benign strains were comparable to similarly powered bundles ABB and ABE (irradiated to lower burnups), with MP residual sheath strains of -0.1% to 0.0% and -0.3% to 0.0%, respectively [3]. These values are typical for similarly operated UO_2 fuel [11].

Higher-powered bundle ADP had MP residual sheath strains of +0.1% to +0.5% in ES-242 and siloxane coated outer elements. This is comparable to sheath strains observed in DAG-154 coated bundles ABC (BDL-419) and AMD (BDL-447), which achieved similar burnups but lower powers than ADP (there is no MOX bundle of comparable high power). Bundle ABC had MP residual sheath strains ranging from -0.1% to +0.1% [3] and DAG-154 coated bundle AMC (BDL-446) from -0.3% to -0.2% [6]. The relatively high MP sheath strains observed in DAG-154 coated bundle ADP outer elements (from +1.4% to +2.1%) are attributed to internal gas overpressure. A plot of MP residual sheath strain as a function of internal gas volume is shown in Figure 4. Note the high strain, high gas volume, DAG-154 coated intact ADP elements in the upper right corner.

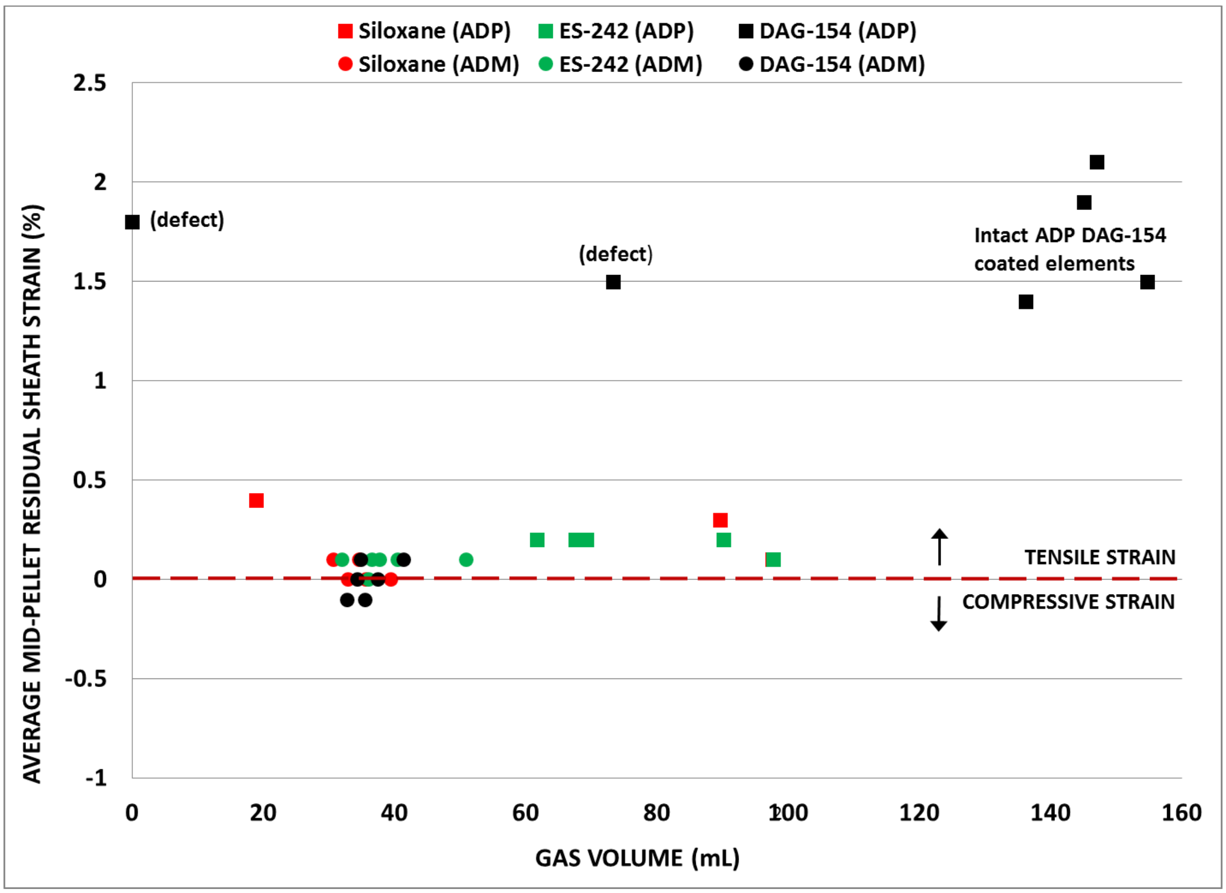

Figure 4 Average MP Residual Sheath Strain (%) as a Function of Gas Volume (mL) for ADM and ADP Outer Elements.

4.1.2 Fission-Gas Release

Fission-gas release (FGR) is a measurement of the quantity of fission-gas released from the fuel pellets to the element internal void space as a percentage of the calculated fission-gas produced during irradiation. Fission-gas release in bundle ADM outer elements ranged from 4-7 %, with no significant difference between coating types. These results are comparable to BDL-419 bundles ABB and ABE that experienced similar powers but lower burnups (FGR of 4-5 % and 7-9 %, respectively) [3]. NPD-40 bundle KB achieved a higher burnup than ADM (but also experienced lower power) and had FGR of <5 % [1]. This is consistent with previous observations that FGR has a stronger dependence on power than burnup [11].

Fission-gas release was 14-15 % in the ES-242 coated outer elements of bundle ADP. Fission-gas release was not measured for siloxane coated elements, but expected to be similar to those coated with ES-242, based on similar measured gas volumes. These values are in the range expected for similarly operated UO_2 fuels [11]. By comparison, DAG-154 coated outer elements exhibited high FGR of 35-37 %. DAG-154 coated bundle ABC experienced slightly lower power than ADP and had FGR of 12-13 % [3], similar to the ES-242 and siloxane coated elements on ADP. BDL-446 bundle AMC had elements with three different degrees of Pu homogeneity, noting that FGR decreased with increasing homogeneity [6]. Although dispersed uniformly throughout the UO_2 fuel matrix, the relatively large (10-40 μm) heterogeneously-distributed Pu-rich regions may have contributed to high FGR in high-powered bundle ADP. The results suggest that heterogeneously-distributed MOX fuel irradiated at BOL powers > 65 kW/m can lead to high FGR in DAG-154 coated elements if irradiated to extended burnup. This effect was not observed in ES-242 or siloxane coated elements under the same conditions, indicating that fuel performance was influenced by the coating type. This is consistent with past irradiation assessments of DAG-154, siloxane and ES-242 at CNL (see Section 4.2).

4.1.3 Grain Growth

Equiaxed grain growth was observed in ADM outer elements, while columnar grain growth was observed in the outer elements of ADP. Previous BDL-419 fuel irradiations (for bundles with similar power histories) indicate that the onset of columnar grain growth in MOX fuels occurs at > 55 kW/m [2, 3, 11]. In comparison, columnar grain growth is observed to occur in UO_2 fuel between 62-65 kW/m [11]. Similar to previous BDL-419 fuel assessments [2], columnar grain growth observed at lower powers than observed in UO_2 indicates either higher central temperatures in MOX fuel and/or different grain-growth kinetics [2]. Blanpain *et. al* measured higher operating temperatures in LWR MOX fuel containing 4-5 wt.% Pu, noting the difference in MOX and UO_2 centreline temperatures increases with increasing linear power, up to a difference of 100 °C at 40 kW/m [12].

4.1.4 Oxide on the Sheath Inner Bore

DAG-154 coated outer elements in ADM had oxide on the inner bore of the sheath up to 12 μm in thickness. ADP outer elements had thicknesses up to 9 μm. Bundles ABB and ABC had oxide thicknesses from 1 to 8 μm and bundles ABD and ABE had very little oxide [2]. While the sheath inner bore oxide thickness increases with burnup, no obvious effect on overall fuel performance has been observed. Similarly designed and operated UO_2 fuels typically exhibit oxide films <1 μm thick [12]. MOX fuels tend to have thicker oxide layers on the inner bore of the sheath because Pu-239 fission produces less oxide-forming fission products than the fission of U-235 [3][13], resulting in the liberation of more oxygen that can interact with the Zircaloy sheath.

4.2 Sheath Coating Performance

Bundles ADM and ADP were the first BDL-419 bundles examined with different types of sheath coatings on the outer elements. In high-powered bundle ADP, the performance of elements coated with ES-242 and siloxane was better than that of elements coated with DAG-154, evidenced by the significant difference in sheath strain, FGR, and grain growth. In contrast, no correlation between coating type and performance was observed in lower-powered bundle ADM (not withstanding its higher burnup than bundle ADP). The results indicate that coating type has an effect on the performance of MOX fuel that operates at high power. Coating type has also been observed to have an effect on fuel performance in high powered UO_2 fuels; a brief summary of CNL coating performance experience is presented here, with a particular focus on irradiations where DAG-154 coated elements were irradiated alongside ES-242 or siloxane coated elements. The majority of historical irradiations with alternate coatings were power ramps of relatively low burnup fuels.

The DME-159 experiment consisted of siloxane and DAG-154 coated elements being power ramped to 62 kW/m after achieving burnups up to 135 MWh/kgU (6 MWd/kgU). Post-irradiation examination revealed FGR of 18-20 % in DAG-154 compared to 2-6 % in siloxane coated elements. Grain growth ratios (R/A) ranged from 0.2 to 0.5 in siloxane compared to 0.5 in the DAG-154 element examined.

The DME-174 experiment had UO_2 fuel pellets of various surface roughness in sheaths coated with siloxane or DAG-154 mounted on two bundles that achieved powers of 50-60 kW/m and a burnup of 135 MWh/kgU (6 MWd/kgU). Fission-gas release was 18-25% in siloxane coated elements compared to 22-29 % in DAG-154 coated elements. Columnar grain growth was observed in all elements except for two coated with siloxane.

In the NRX-661 test, two elements coated with ES-242 and one with DAG-154 were irradiated at 42 kW/m before being power ramped to 87 kW/m at 69 MWh/kgU. The DAG-154 coated element experienced the greatest degree of fuel restructuring, indicating it experienced higher temperatures than the two ES-242 coated elements.

Although DAG-154 sheath coatings have shown adequate performance within the standard operating envelope for CANDU fuel, previous irradiations of siloxane and ES-242 coated elements have shown better performance than DAG-154 coated elements, particularly at high operating power. This is consistent with observations made for BDL-419 bundles ADM and ADP.

5. Summary and Conclusions

MOX fuel performance is comparable to that of UO_2 under CANDU power reactor conditions. Both MOX and UO_2 exhibit similar internal gas overpressure failure thresholds for fuel that has operated at high power to extended burnup. High-powered bundle ADP outer elements exhibited less grain growth and FGR when coated with ES-242 and siloxane rather than DAG-154. This is consistent with past experience with high-powered UO_2 fuel where DAG-154 coated elements were irradiated with ES-242 or siloxane coated elements.

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