STATUS OF IRRADIATION TESTING AND PIE OF MOX (Pu-CONTAINING) FUEL

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ABSTRACT

This paper describes AECL's mixed oxide (MOX) fuel-irradiation and post-irradiation examination (PIE) program. Post-irradiation examination results of two major irradiation experiments involving several (U, Pu)O_2 fuel bundles are highlighted. One experiment involved bundles irradiated to burnups ranging from 400 to 1200 MWh/kgHE in the Nuclear Power Demonstration (NPD) reactor. The other experiment consisted of several (U, Pu)O_2 bundles irradiated to burnups of up to 500 MWh/kgHE in the National Research Universal (NRU) reactor. Results of these experiments demonstrate the excellent performance of CANDU® MOX fuel. This paper also outlines the status of current MOX fuel irradiation tests, including the irradiation of various (U, Pu)O_2 and (Th, Pu)O_2 bundles. The strategic importance of MOX fuel to CANDU fuel-cycle flexibility is discussed.

INTRODUCTION

As an integral component of AECL's mixed oxide (MOX) fuel program, irradiation testing and post-irradiation examination (PIE) of various types of MOX (Pu-containing) fuel have been on-going for more than thirty years. Initially, the program consisted of multi-element tests aimed at studying the basic ceramic properties and behaviour of MOX fuel. More recently, the program has involved multi-bundle demonstration testing of Canadian-fabricated MOX fuel.

This paper discusses the results of two multi-bundle irradiations, namely the NPD-40 and the BDL-419 experiments. These experiments consisted of irradiating several (U, Pu)O_2 bundles in the Nuclear Power Demonstration (NPD) and the National Research Universal (NRU) reactors. The PIE results of both tests are highlighted. In addition, this paper outlines the status of current MOX fuel irradiation tests, including the irradiation of various (U, Pu)O_2 and (Th, Pu)O_2 bundles in NRU.

THE NPD-40 EXPERIMENT

Objective

The NPD-40 experiment was a demonstration-scale irradiation test in NPD involving six bundles containing (U, Pu)O_2 fuel [1]. The objective of the experiment was to show that MOX fuel could operate successfully to extended burnup, under typical CANDU power-reactor operating conditions. Subsequent to the NPD irradiation, several bundles were power-ramped in NRU.

Fuel Design and Fabrication

The six bundles were of the standard 19-element design, but contained two different types of non-standard fuel pellets from two different manufacturers. Three of the bundles had annular pellets (3.33 wt.% Pu in natural uranium; density = 10.5 Mg/m^3) contained in thin-walled, collapsible Zircaloy-4 sheaths; the other three bundles had lower-density solid pellets (3.00 wt.% Pu in natural uranium; density = 10.2 Mg/m^3) contained in thick-walled, free-standing Zircaloy-4 sheaths (Table 1). The pellets were fabricated by two manufacturers using standard methods (i.e., mixing and blending, pre-pressing, granulation, final pressing, sintering and grinding).

Zircaloy-4 sub-assemblies (i.e., appendaged sheaths with one end cap welded in place and with graphite CANLUB coating on the sheath inside surface) were supplied by General Electric Canada (GEC). Loading of measured stacks of pellets and plenum inserts into the sub-assemblies and TIG end-closure welding were performed inside glove boxes at the Chalk River Laboratories (CRL). Bundle assembly welding was conducted by GEC. The bundles were inspected at CRL prior to irradiation in NPD.

Irradiation History

The bundles were loaded into NPD in 1973. Most of the bundles achieved outer-element discharge burnups in the range of 370 to 420 MWh/kgHE, with the exception of bundle KB, which achieved an outer-element burnup of 1180 MWh/kgHE (Table 2). Outer-element linear powers (OELP) up to 50 kW/m were experienced at beginning-of-life, decreasing to about 15 to 25 kW/m at end-of-life.

Following the NPD irradiation, three bundles (one with normal-density pellets, bundle KA, and two with low-density pellets, bundles KE and KF) were power-ramped in NRU. After a few hours of low-power operation (OELP of about 20 kW/m), the bundles were ramped to an OELP of 50 kW/m. This power ramp produced some defects in bundle KA. Bundles KE and KF survived and operated at 50 kW/m for 10 days. A further increase in OELP to about 70 kW/m resulted in a defect in bundle KE.

Post-Irradiation Examination

All bundles were visually examined after the NPD irradiation. The bundles were disassembled, and several elements were measured for their dimensional changes. In general, the elements exhibited compressive midpellet strains; the maximum values were -0.7% for the thin-walled, collapsible sheaths and -0.3% for the thick-walled, free-standing sheaths. Following the power ramp in NRU, elements from bundle KA (ramped to 50 kW/m) still exhibited small compressive strains. Following the power ramp to 70 kW/m, outer elements of bundles KE and KF exhibited tensile strains of about 1%. The lower-powered inner elements of both bundles exhibited compressive strains.

Internal gases were extracted, measured and analyzed by mass spectrometry for all bundles, except KD. The non-ramped bundles (KB and KC) exhibited low fission-gas release (FGR) in the range from 2 to 4% (Table 3). Elements that were ramped to 70 kW/m (KE and KF) experienced high FGR, in the range of 45 to 50%. Bundle KA, which was ramped to 50 kW/m, exhibited outer-element FGR of about 11%.

Metallographic examination showed that the microstructures had isolated regions of porosity and metallic fission-product precipitates (Figure 1). These were attributed to the presence of high-fissile-content particles in the as-sintered pellet that resulted in zones of increased fission density and localized temperature increase during operation. It is postulated that most of the FGR was from these zones of initial high-fissile Pu content.

PIE measurements indicated that elements containing low-density pellets had thicker CANLUB coatings than those containing normal-density pellets. There is no clear reason for this, although it may be attributed to a difference in as-fabricated coating thickness (the bundle kits were fabricated at different periods during 1972). The thicker CANLUB coatings may have augmented the benefits of low-density pellets, resulting in the observed superior performance of these bundles during power-ramp tests in NRU.

Conclusions of NPD-40

This test demonstrates the excellent performance of a dry-blended MOX fuel bundle that experienced a declining power history from beginning-of-life power ratings up to 50 kW/m to very high burnup (outer element = 1180 MWh/kgHE). Dimensional changes and FGR were minimal, and there were no indications of defects during the irradiation. Two of three bundles that experienced power ramps at about 400 MWh/kgHE experienced failures; those with low-density pellets and thick-walled sheaths exhibited a higher failure threshold (~ 70 kW/m) than the bundle with normal-density pellets and thin-walled sheathing (~ 50 kW/m).

THE BDL-419 EXPERIMENT

Objectives

Like the NPD-40 experiment, BDL-419 is a multi-bundle demonstration irradiation; however, unlike NPD-40, BDL-419 involved Bruce-type fuel that was completely fabricated in Canada. The objectives of the experiment were to:

1. Demonstrate that (U, Pu)O_2 fuel fabricated in the Recycle Fuel Fabrication Laboratories (RFFL) at CRL is capable of sustaining the power history requirement for CANDU power reactors.

2. Examine the performance of Pu-enriched Bruce-type fuel to high burnup under CANDU power-reactor operating conditions.

Six BDL-419 bundles, ABA to ABF, were irradiated in NRU during the early 1980s. This irradiation was terminated when some bundles were inadvertently broken during handling in the irradiated fuel bays. This paper discusses the PIE results for bundles ABB and ABC that achieved the highest powers and burnups.

Fuel Design and Fabrication

A total of fifteen (U, Pu)O_2 Bruce-type fuel bundles were produced for BDL-419. These bundles were fabricated in the RFFL, the CRL facility comprising lines of glove boxes and fume hoods, designed and built to fabricate alpha-active fuels [2]. The (U, Pu)O_2 fuel pellets contained approximately 0.5 wt % fissile plutonium in natural uranium. A conventional dry-blending process (i.e., turbula blending of the starting fuel powders, followed by pre-pressing, granulation, final pressing, sintering and centreless grinding) was employed for the fabrication of the pellets, resulting in an average pellet density of 10.54 g/cm^3. The pellets were loaded into sheaths coated with either DAG-154, ES-242 or siloxane. The loaded elements were assembled into bundles by GEC in 1980. The bundles were essentially the same as 37-element Bruce power-reactor fuel bundles, except for the removal of the central element to facilitate insertion of a central guide tube for vertical irradiation in NRU. Other pertinent manufacturing data are listed in Table 4.

Irradiation History of Bundles ABB and ABC

Bundles ABB and ABC were first installed in NRU in 1980 March and discharged from the reactor in 1984 August with chemically determined outer-element burnups of 470 and 540 MWh/kgHE, respectively (Table 5). During irradiation, both bundles experienced declining power histories, with a sustained maximum OELP of 52 and 59 kW/m, respectively.

Post-Irradiation Examination

Using in-cell periscopes, bundles ABB and ABC were examined visually and were found to be in good condition. Diameters of elements from both bundles were measured using an in-cell profilometer. Residual midpellet strains were generally compressive; ABB and ABC outer elements exhibited average midpellet strains of -0.1% and 0%, respectively. These results are within the range of those observed in natural UO_2 bundles having similar pellet densities and power histories.

Elements from bundles ABB and ABC were selected for gas-puncture analysis. The calculated FGR for the outer elements of bundles ABB and ABC was 4% and 13%, respectively (Table 6). Figure 2 compares the FGR results from bundles ABB and ABC with those from other natural UO_2 bundles having similar declining power histories. The FGR of bundles ABB and ABC is generally consistent with that observed by Floyd et al. for natural UO_2 CANDU fuel that experienced similar power histories in Bruce-A [3,4].

Metallographic examinations revealed that significant central grain growth occurred in the outer-element pellets of bundles ABB and ABC, resulting in equiaxed grains greater than 100 μm (Figure 3). This extent of grain growth is not observed in natural UO_2 power reactor bundles, even for those that experienced similar power histories to burnups of about 700 MWh/kgU [3,4]. This significant difference in fuel restructuring may indicate that the central temperature of outer-element pellets from bundle ABB and ABC was higher than that of UO_2 fuel experiencing similar power histories, or that the MOX fuel has different grain-growth kinetics. The extensive microstructural changes in bundles ABB and ABC probably occurred in the early part of the irradiation, when peak powers were achieved.

Figure 4 shows the macrograph and alpha and beta/gamma autoradiographs for an outer element from bundle ABC. The higher-powered outer element exhibits evidence of central grain growth, Pu homogenization and fission-product migration. These features are not observed in the lower-powered intermediate elements. Metallic precipitates, similar to the metallic fission-product precipitates seen in the NPD-40 bundles and attributed to the presence of high-fissile-content particles, were also observed in these bundles.

Discussion of Bundle ABB/ABC Performance

Investigations have demonstrated that MOX fuel having high Pu concentrations (10%-30%) exhibit lower thermal conductivities than that of UO_2 [5]. Blanpain et al. demonstrated that MOX fuel containing ~ 5% Pu in depleted uranium experiences centreline temperatures 100°C greater than those for UO_2 at a power of 40 kW/m [6]. Although lower thermal conductivities (and higher operating temperatures) have been observed in MOX fuel, the Pu content of BDL-419 fuel is much lower than that used in these studies; i.e., 0.5% Pu in BDL-419 fuel vs. 5 - 30% Pu in the other fuels. Hence, thermal conductivity differences may not fully explain the observed grain growth. Another possible explanation is that this fuel may have experienced different grain-growth kinetics (e.g., a lower temperature threshold for columnar grain growth) compared to natural UO_2 fuel. Further studies are required to investigate the thermal conductivities and grain growth kinetics of MOX fuels having the appropriate range of Pu concentrations.

In spite of undergoing enhanced grain growth, bundles ABB and ABC did not exhibit FGR higher than that expected for comparable UO_2 fuel irradiated in Bruce-A. There are several possible reasons for this:

1. A difference in the point in the irradiation when peak power was achieved (40 MWh/kgHE for bundles ABB and ABC vs. 80 to 120 MWh/kgU for Bruce-A UO_2 bundles).
2. Lower pellet density (10.5 g/cm^3 for bundles ABB and ABC vs. 10.7 g/cm^3 for Bruce-A UO_2 bundles).
3. Recent studies indicate that, although local FGR from Pu-rich agglomerates is high due to high local burnup, the fission gas tends to be retained by the relatively cool, surrounding matrix, forming closed porosity [5,7].

Conclusions from the PIE of Bundles ABB and ABC

Bundles ABB and ABC have demonstrated that $(U, Pu)O_2$ fuel fabricated by AECL in the RFFL can be successfully operated with declining power histories at peak OELPs greater than 50 kW/m to burnups around 500 MWh/kgHE. In general, their performance was comparable to that of natural UO_2 power reactor fuel. Although more extensive grain restructuring was observed in the MOX fuel compared with similarly operated UO_2 fuel (attributed to higher centreline temperatures or different grain-growth kinetics), this had no deleterious effect on the overall performance of the bundles. In particular, the FGR of the MOX fuel was similar to that of comparable UO_2 fuel.

CURRENT MOX FUEL IRRADIATIONS AT AECL

Several bundles from the BDL-419 experiment are currently under irradiation in NRU. (Th, 1.4% Pu)O_2 bundles are also being irradiated under experiment BDL-422. Both experiments demonstrate the performance of fuel bundles fabricated by AECL in the RFFL.

The BDL-419 bundles currently under irradiation in NRU have reached outer-element burnups up to 500 MWh/kgHE. In light of the excellent performance of bundles ABB and ABC, the target burnup for the bundles currently under irradiation is 750 to 1000 MWh/kgHE.

The BDL-422 experiment involves the irradiation testing of six Bruce-type $(Th, Pu)O_2$ fuel bundles containing outer-element gas plenums in NRU. Outer-element burnups are presently at ~ 500 MWh/kgHE. PIE is planned on three of these bundles, with the remainder being irradiated to a target burnup of 750 to 1000 MWh/kgHE.

STRATEGIC IMPORTANCE OF MOX FUEL

There are several reasons for the continued strategic importance of MOX fuel utilization in CANDU power reactors. Worldwide experience in MOX fuel fabrication and in-reactor performance is growing, resulting in increased confidence in this fuel type. Plutonium from conventional reprocessing is currently mixed with depleted uranium to form MOX fuel, which is used in several pressurized water reactors (PWRs) in Europe. For example, in France, 16 PWRs are already licensed to operate with partial (30%) MOX cores, and plans to expand this to 28 PWRs are being considered [8]. MOX fuel has also been used to operate two PWRs in Switzerland for 17 years [9]. In Japan, the long-term nuclear plan has a target of about ten light-water reactors utilizing MOX fuel by the year 2000 [10]. The required infrastructure for MOX fuel utilization already exists in these parts of the world. The ability to utilize MOX fuel in CANDU power reactors contributes to the flexibility of CANDU technology in fuel-cycle utilization.

MOX utilization is an important factor in the synergism between CANDU and other reactor systems. In Korea, the CANDU/PWR two-reactor system offers the possibility of recycling the MOX fuel from reprocessed PWR fuel back into CANDU reactors. This would have potential advantages compared with recycling in a PWR. A full MOX core could be used in existing CANDU reactors. Although MOX fuel is much more expensive to manufacture compared to natural uranium, the relatively simpler design of the CANDU fuel bundle will result in less expensive MOX fuel-fabrication costs compared with PWR MOX. A high-burnup CANDU MOX fuel therefore has the potential of lower fuel-cycle costs than PWR MOX. Up to 50% more energy could be extracted from the plutonium as MOX fuel in CANDU reactors, compared with recycle in a PWR. This has important advantages in improving natural uranium utilization, reducing enrichment requirements, and in reducing the amount of spent fuel for ultimate disposal. Further possibilities exist of achieving the full potential of CANDU/PWR synergism with new fuel-recycling processes. An example is the TANDEM fuel cycle, where the uranium and plutonium from PWR spent fuel are co-precipitated without separation. Only the fission products, and higher actinide isotopes, are removed. This fuel cycle takes advantage of the fact that the fissile component in PWR spent fuel (about 1.5%) can be used directly in CANDU reactors, without readjustment of the enrichment. This process is potentially simpler, cheaper, and more easily safeguarded than conventional reprocessing.

More recently, disarmament efforts in the United States and the former Soviet Union have resulted in large inventories of weapons-derived plutonium that require dispositioning. One option being considered is to incorporate the plutonium into MOX CANDU fuel and utilize it in Bruce NGS-A reactors [11]. AECL's experience, resulting from 30 years of MOX fuel fabrication and irradiation testing, has contributed to the credibility of this option.

Thorium fuel cycles also offer a resource-efficient alternative for dispositioning weapons-derived nuclear material: either highly enriched uranium or plutonium. In the Pu/Th cycle, a high fraction of plutonium would be consumed (>90% of the fissile plutonium, or >75% of the total plutonium), while valuable ^{233}U would be produced. The used fuel would be stored until the ^{233}U could be recovered in a manner that was economical and highly safeguardable.

SUMMARY

Irradiation testing and PIE of MOX fuel is continuing at AECL; it has progressed from multi-element to multi-bundle demonstration testing of the 37-element design. PIE has confirmed the excellent performance of (U, Pu)O_2 bundles in the following cases:

1. Experiencing declining power histories from beginning-of-life powers up to 50 kW/m to burnups up to 1180 MWh/kgHE.

2. Experiencing declining power histories from beginning-of-life powers > 50 kW/m to burnups of about 500 MWh/kgHE.

In both cases, dimensional changes and FGR were comparable to that expected for UO_2. These irradiation performance assessments have demonstrated that MOX fuel remains a viable CANDU fuel-cycle option that strategically warrants continued development. Further irradiation testing is required to investigate high-burnup CANDU fuel design with appropriate Pu concentrations relevant to projected MOX fuel-cycle needs.

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Table 1. Fuel Design Data for Bundles of Experiment NPD-40 (Type I is Bundles KA, KB and KC; Type II is Bundles KD, KE and KF; Dimensions in mm unless otherwise stated)

Parameter	Type I Fuel	Type II Fuel
Bundle Design	19-Element Geometry	19-Element Geometry
Overall Length	495.0	495.0
Overall Diameter	81.6	81.6
Element Design		
Overall Length	492.6	492.6
Internal Diameter	14.44	13.77
Wall Thickness	0.38	0.65
Sheathing Material	Zircaloy-4	Zircaloy-4
Pellet Design		
Manufacturer	Aktiebolaget Atomenergi	Belgonucleaire
Density (Mg/m^3)	10.5	10.2
Outside Diameter	14.2	13.67
Central Hole Diameter	2.54	
Length	14.6	14.68
Enrichment	3.33 wt% Pu in (U + Pu)	3.0 wt% Pu in (U + Pu)

Table 2. NPD-40 Element Burnup (MWh/kgHE)

Bundle	Centre Element	Inner Element	Outer Element	Bundle Average
KA	170	220	420	340
KB	850	930	1180	1080
KC	170	230	420	350
KD	170	220	390	320
KE	170	210	370	310
KF	190	240	400	340

Table 3. Fission-Gas (Xenon) Release for Outer Elements of NPD-40 Bundles

Bundle	Ramp Power, kW/m	Range FGR, %	Average FGR, %
KB	non-ramped	2.0 - 4.3	3.4
KC	non-ramped	--	2.4
KA	50	11.0 - 11.1	11.1
KE	70	45.2 - 47.7	46.7
KF	70	47.1 - 48.5	47.8

Table 4. Manufacturing Data for BDL-419 Bundles

Parameter	Value
Bundle Design	37-Element (Bruce) Geometry
Overall Length	493.59 mm
Heavy Element Weight	17.69 kg
Element Data	
Stack Length	465-472 mm
Sheath Internal Diameter	12.22-12.24 mm
Sheath Outer Diameter	13.06 mm
Sheathing Material	Cold-worked Zircaloy-4
Pellet Data	
Manufacturer	RFFL, AECL
Density	10.54 (10.47-10.60) g/cm^3
Outside Diameter	12.1 mm
Length	12.1 (11.8-12.4) mm
Enrichment	0.49 wt% fissile Pu in (Pu + U)

Table 5. BDL-419 Bundles ABB and ABC Burnup and Power History

Bundle	Outer-Element Burnup (MWh/kgHE)	Power (kW/m)		
		Operating[a]	Sustained Max.[b]	Peak[c]
ABB	468	47	52	54
ABC	539	53	59	61

a. highest average power for a given burnup interval
b. highest operating power sustained for at least 12 h
c. highest power attained during the irradiation

Table 6. Fission-Gas (Xenon) Release for BDL-419 Bundles ABB and ABC

Bundle-Element Type	Calculated Volume of Xenon Produced (mL)	Avg. Measured Volume of Xenon Collected (mL)	Avg. Percentage of Xenon Released (%)
ABB-Outer	255.5	11.0	4.3
ABB-Intermediate	198.7	0.82	0.4
ABC-Outer	294.3	37.2	12.7
ABC-Intermediate	208.0	1.3	0.6

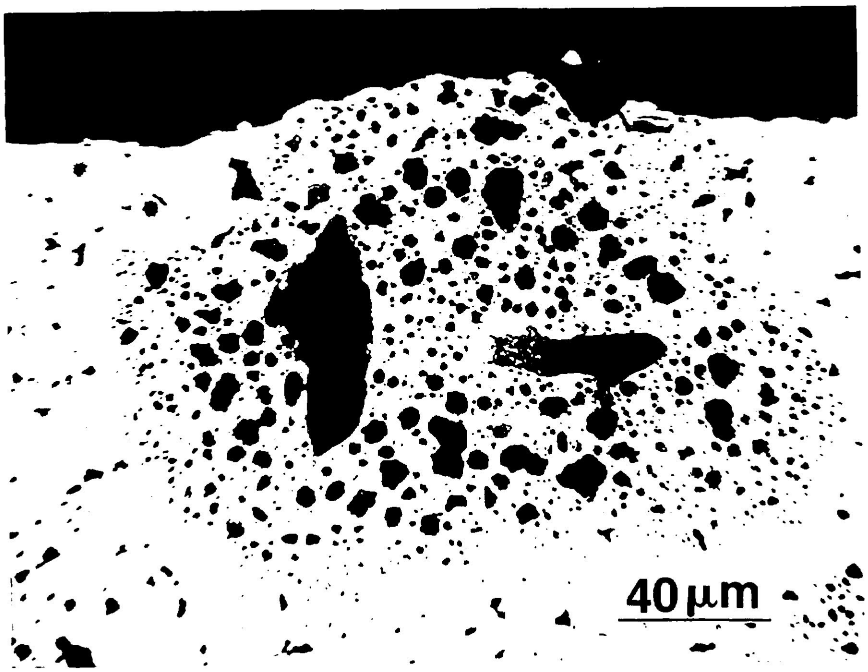


Figure 1. Microstructure of element KB-01 showing porous zone with metallic precipitates.

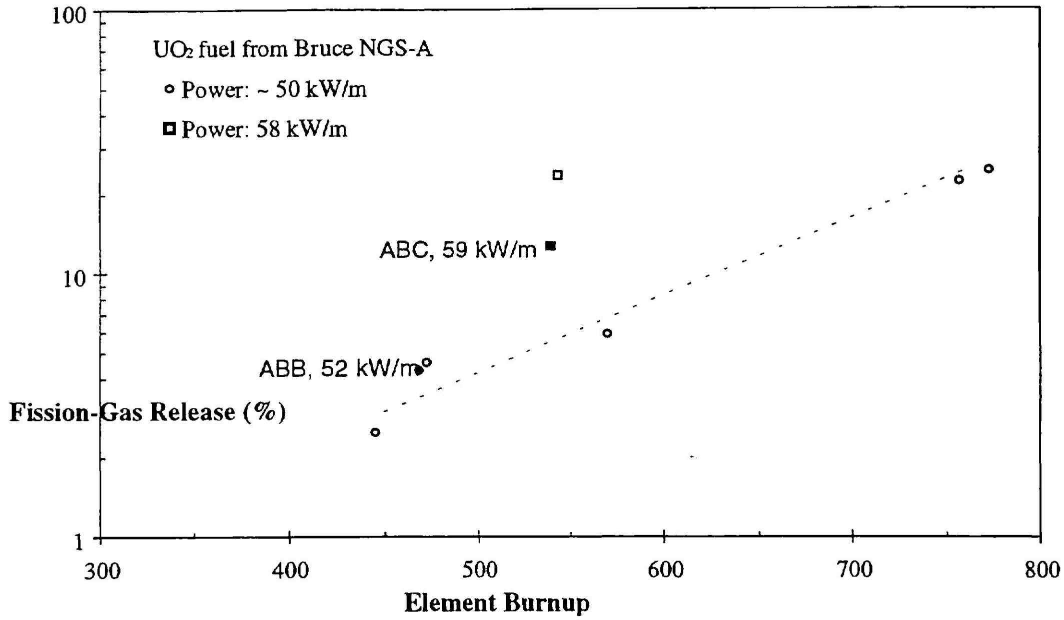


Figure 2. Fission-Gas Release versus Element Burnup for fuel experiencing declining power histories from beginning-of-life powers in the range of 50-59 kW/m (as shown).

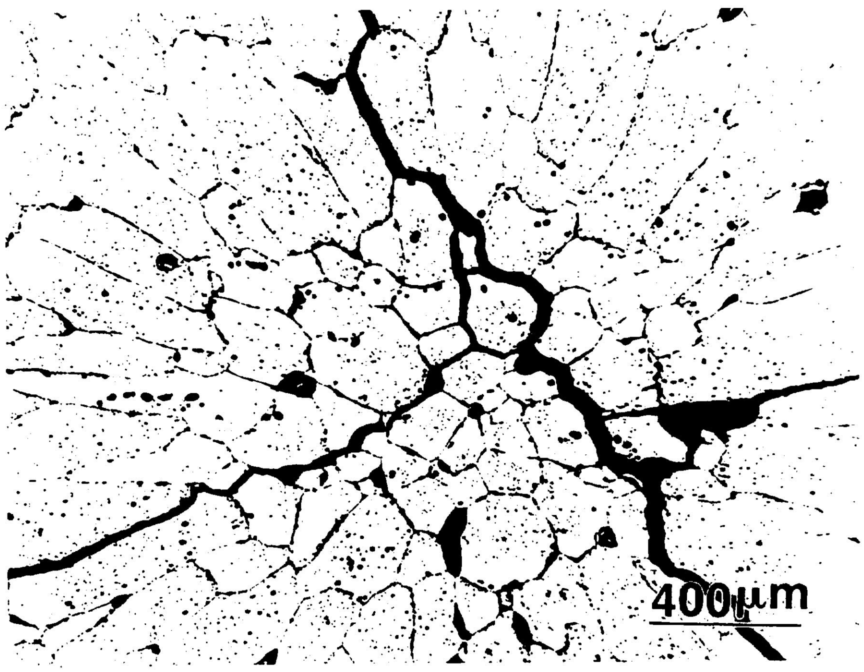
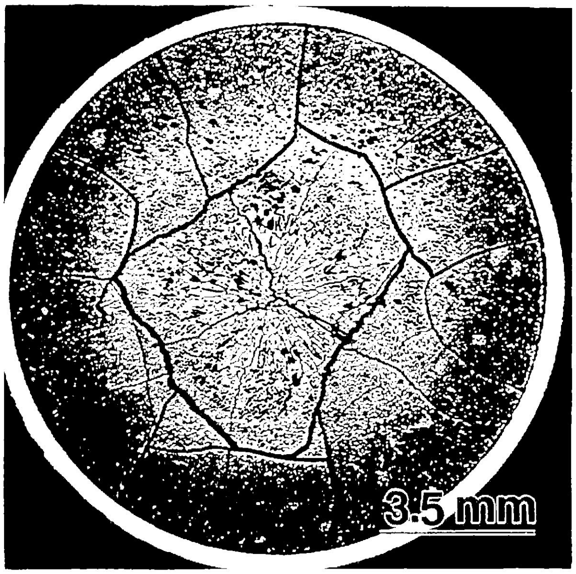
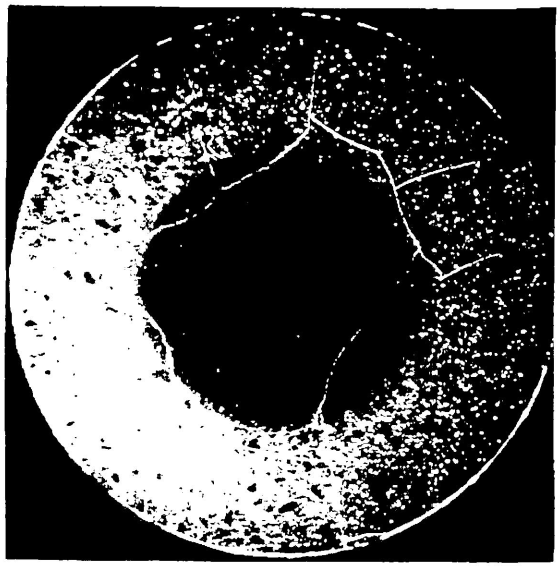


Figure 3. Micrograph showing grain morphology at pellet centre of an outer element from Bundle ABC. Note that the grains at pellet centres are larger than 100 μm.



(a)



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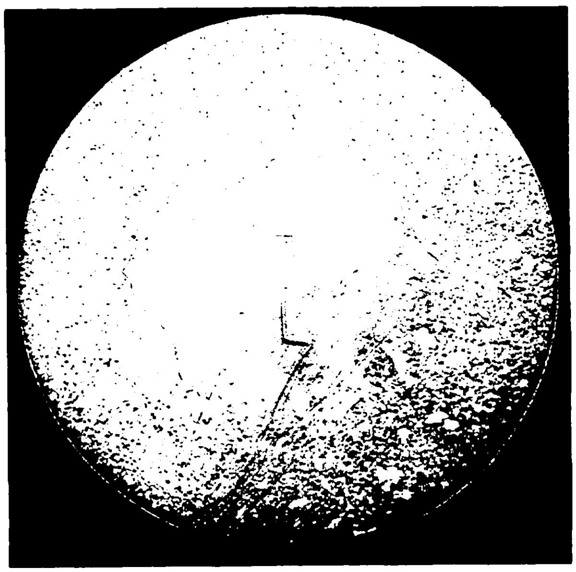

(c)

Figure 4. Outer element from bundle ABC: (a) macrograph showing fuel structure, (b) β/γ autoradiograph (white spots represent high fission-product regions and dark circle indicates fission-product depletion in the pellet centre), and (c) α autoradiograph (dark spots represent Pu-rich regions; note the Pu homogenization region in the pellet centre).