THE PARALLEX PROJECT: CANDU MOX FUEL TESTING WITH WEAPONS-DERIVED PLUTONIUM

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ABSTRACT

The Parallex Project consists of a parallel experiment in which weapons-derived plutonium (WPu) from the United States and from the Russian Federation will be tested as mixed-oxide (MOX) CANDU® fuel in the National Research Universal (NRU) reactor at the Chalk River Laboratories in Canada. Plutonium derived from excess weapons will be fabricated into CANDU MOX fuel at the A.A. Bochvar Institute in Moscow and at the Los Alamos National Laboratory in the United States. The MOX fuel will be transported to CRL, where it will be characterized, assembled into fuel bundles and then irradiated in the NRU reactor. Following irradiation, the fuel will be examined in hot cells to assess its irradiation performance. This paper describes the scope, rationale and current status of the Parallex Project.

BACKGROUND AND INTRODUCTION

The end of the Cold War has created hundreds of tonnes of surplus weapons-usable fissile materials in both the United States and the Russian Federation. In 1994, the US National Academy of Sciences (NAS) warned that the vast excess stocks of fissile material (highly enriched uranium and plutonium) pose a "clear and present danger" to international security and non-proliferation because of risk of theft or diversion [1]. The NAS recommended achieving as-soon-as-possible disposition of the excess plutonium (Pu) to a "spent fuel standard". To achieve this standard, the Pu must be placed in a form in which it is at least as difficult to recover for weapons use as is the Pu in spent fuel from commercial reactors.

The US Department of Energy (USDOE) has been evaluating several options for the disposition of excess Pu. One of the options being considered is to incorporate the Pu into mixed-oxide (MOX) fuel for utilization in commercial power reactors. The Canadian government with Atomic Energy of Canada Limited (AECL) and Ontario

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Hydro have agreed to consider the CANDU reactors at the Bruce A Nuclear Generating Station as the reference for this mission. The feasibility of the CANDU MOX fuel option was established by AECL and Ontario Hydro in a study sponsored by the USDOE in 1994. This feasibility study covered technical and strategic issues, schedule, and cost-related parameters, with the objective of identifying an arrangement permitting consumption of 50 t of weapons-derived Pu as MOX fuel in CANDU reactors over a 25-year period [2]. It was concluded that MOX fuel could be fabricated in the United States, transported to Canada, and used as fuel for Bruce A reactors without significant modification to the reactors, and only small changes to the fuel handling. An extension to this study in 1996 further concluded that the duration of the Pu dispositioning mission could be reduced to 15 years.

A parallel study in 1996 was conducted by Canada and Russia, to assess the feasibility of fabricating CANDU MOX fuel to Canadian specifications in Russia and safely transporting the fuel to the Bruce site in Canada [3]. The feasibility of utilizing Russian MOX fuel was established, with consideration of technical, environmental, regulatory and economic issues.

On 1997 January 14, the USDOE issued its Record of Decision [4], which announced a two-track program to disposition surplus Pu. Most of the Pu will be converted to MOX fuel for use in commercial power reactors (known as the MOX option), in parallel with an immobilization option - encasing the Pu with radioactive waste in glass blocks and consignment to an underground repository.

Within the MOX option, both light-water reactors (LWRs) and CANDU reactors are under consideration. The use of CANDU reactors in the Pu dispositioning mission has been retained in the United States as an option, in the event that multilateral agreements are reached by the United States, the Russian Federation and Canada for joint disposition of some portion of the American and Russian Pu.

The Parallex Project was initiated in 1996 May and represents the first step towards demonstrating the feasibility of the CANDU MOX concept with weapons-derived Pu. The test program builds on existing CANDU MOX fuel experience that has been acquired over 30 years of R&D, related to MOX fuel irradiation performance and fabrication development [5-7].

PROJECT SCOPE

The scope of the Parallex test covers fabrication, irradiation, and examination of one experimental bundle of CANDU MOX fuel containing plutonium from weapons disassembled in the United States and the Russian Federation. The conversion of the plutonium from weapons components (pits) to PuO_2 is not a part of the test but is a prerequisite. The fabrication of MOX fuel pellets meeting the specifications of AECL will be accomplished through blending the PuO_2 with depleted UO_2, pressing, sintering, and grinding. The loading of the pellets into elements and closure welding are to be done at the Los Alamos National Laboratory (LANL) for the US fuel and at Bochvar for the RF fuel. The assembly of elements into bundles and necessary tests and characterization before irradiation will be done at the Chalk River Laboratories (CRL). In addition, CRL will procure or fabricate the hardware and other elements to complete the bundles and do the necessary analyses to support the irradiation. Irradiation will be done in the pressurized loops of the National Research Universal (NRU) reactor at CRL for a period of about 16 months. Then, the fuel will be examined in the hot cells at CRL to assess its performance.

The original scope was for 4 bundles of joint US–RF fuel to be tested. The current plan covers one bundle and retains an option for 3 additional bundles, contingent upon the outlook of US–RF–Canada negotiations on the CANDU option, funding for additional MOX fuel fabrication at LANL, and availability of facilities at LANL. Thus the *first bundle* refers to the currently planned 1-bundle test, the *full test* refers to the original 4-bundle test matrix, and *contingency bundles* refers to the 3 additional bundles. The first bundle will contain about 10.6 kg MOX (280 g Pu), and the full test program includes 54 kg MOX (about 1 kg Pu).

Although the data from the Parallex Project will represent a significant first step, much additional work will be required to qualify MOX fuel for use in the Bruce reactors including additional irradiations of the final fuel designs in the NRU reactor, zero-power reactor physics measurements, critical heat flux measurements, and irradiation of prototype bundles in the Bruce reactors.

PROJECT OBJECTIVES

The objectives of the Parallex Project are twofold:

- to contribute to the database that would eventually qualify MOX fuel for use in CANDU reactors, and
- to demonstrate the feasibility of the infrastructure involved in the disposition of excess weapons plutonium as MOX fuel in reactors.

The first objective is met through careful design of both the irradiation test conditions and the fabrication processes that will be used to manufacture the test fuel. The test conditions in the NRU reactor will bracket those expected in the Bruce reactors and provide meaningful data on performance of the MOX fuel. The test will also produce data showing how production and processing variables, as well as the detailed design of the pellets themselves, affect the performance of the CANDU MOX fuel. These comparisons will also be used to optimize the MOX fuel specifications and fabrication methods.

Although only laboratory quantities of MOX fuel are being tested, and a research reactor is being used for the irradiation, most elements of the infrastructure required to utilize the excess weapons plutonium as MOX reactor fuel will be demonstrated as part of the Parallex Project. This satisfies the second objective. Completion of this test will demonstrate the conversion of Pu metal from weapon components into oxide, fabrication of MOX pellets, loading into fuel elements, welding, transportation of MOX fuel to Canada, assembly of bundles, irradiation, discharge, and storage in an interim facility prior to eventual emplacement in a repository as spent fuel.

ROLES AND RESPONSIBILITIES

The Parallex test program is being funded by the USDOE. The Oak Ridge National Laboratory (ORNL) is managing the program on behalf of the USDOE, and will coordinate and oversee the activities of the other parties to ensure success of the project.

AECL, as the design authority for CANDU MOX fuel, has the principal responsibility of ensuring that the irradiation test program is designed and conducted to confirm the feasibility of the CANDU MOX option. AECL will provide specifications, drawings and fabrication guidance to LANL and Bochvar and will inspect the finished fuel pellets and accompanying data reports to ensure that they meet the quality assurance requirements of the test. AECL will supply depleted UO_2, Zircaloy components and some inspection equipment to both fabricators, and procure or manufacture other components for the MOX test bundles. As operator of the NRU reactor where the test will be conducted, AECL is responsible for ensuring that the test is designed and operated in compliance with all the safety and regulatory requirements of the Atomic Energy Control Board and other agencies. Conduct of the NRU irradiation, post-irradiation examination, storage and disposal of the waste materials and reporting the test results is the responsibility of AECL.

The Bochvar Institute will be responsible, under contract to AECL, to develop process parameters and fabricate the RF MOX fuel, using weapons-derived Pu provided by MINATOM. The fabrication of MOX pellets and fuel elements will be done in accordance with AECL technical specifications, drawings and stipulated quality assurance requirements. Bochvar, MINATOM and AECL are jointly responsible for transportation of the finished RF MOX fuel to Canada.

The LANL is responsible for the following: to develop the process parameters and produce the US test MOX fuel in accordance with AECL product specifications and drawings; to secure a source of weapons-derived PuO_2 feed stock, including the available technical data showing its processing and characteristics; and to provide the required data on the characteristics of the finished fuel pellets and elements, as specified in the AECL requirements. In addition, the LANL will be jointly responsible with AECL for the packaging, transportation, safeguards, and security of the finished fuel transfer to CRL.

TEST DESCRIPTION

The Parallex test fuel is based on the reference design of CANDU MOX fuel, which is a 37-element bundle containing depleted UO_2 as the matrix throughout the bundle. The depleted uranium is mixed with 15% dysprosium (Dy, a burnable neutron absorber) in the central 7 elements, with 3.1% Pu in the third ring of 12 elements, and with 1.6% Pu in the outer 18 elements. With a bundle average burnup of 10.0 MW·d/kg HE, this fuel design would disposition about 1.5 t of weapons grade Pu per year per reactor.

The NRU reactor has 2 pressurized test loops that provide 3 test sections, each of which has 6 vertical positions for testing CANDU-sized bundles: positions 1 and 6 on the ends are lower flux, positions 2 and 5 are intermediate flux, and positions 3 and 4 adjacent to the centreline are the highest flux. The centre element in each bundle is removed to accommodate a guide tube for assembling the bundles vertically in the NRU test loops.

An objective of the Parallex irradiations is to test the reference MOX fuel composition (3.1% and 1.6% Pu in U) at heat ratings that bracket the range calculated for operation in the Bruce reactors. However, the conditions in the NRU test loops are such that a 37-element bundle of the reference design (containing 3.1% and 1.6% Pu in the intermediate and outer rings) would operate at overly high linear powers. Accordingly, special bundle configurations were devised for the Parallex tests.

The full Parallex test matrix consists of 4 bundles: two are standard 37-element bundles with the central element removed (termed "fixed" bundles), and two are special "demountable" bundles in which the outer 18 elements are mechanically attached and may be removed and replaced between reactor cycles. The fixed bundles will be used for testing the 3.1% Pu elements with the MOX elements located in the centre 2 rings to lower the heat ratings. The outer ring will contain natural UO_2 in one case, and depleted UO_2 mixed with 1% Dy to lower the power further in the second case. These 2 bundles are named BD-1 and BD-2 respectively, and are shown schematically in Figure 1. The BD-1 bundle is identified as the first bundle for testing.

The demountable bundles will be used for the 1.6% Pu elements with the test elements in the outer detachable ring. The inner 2 rings will contain natural UO_2 elements in one case, and depleted UO_2 mixed with 5% Dy to lower the power in the second case. The cross sections of the demountable bundles, named DM-1 and DM-2, are shown in Figure 2.

Each bundle will contain 2 fuel elements containing UO_2 with low-enriched-uranium (LEU) so that the power rating is approximately the same as the power rating of the surrounding MOX elements. These UO_2 "controls" will provide a standard of comparison to the large UO_2 database.

The demountable bundles will be removed to the spent fuel bays and eight of the demountable elements from each bundle will be replaced after about 100 d in reactor. Visual examinations will be conducted during these handling operations in the bays. Throughout the test, data will be acquired on loop power and reactor conditions and fuel bundle powers will be calculated and documented for each irradiation period. At the completion of irradiation, elements will be cooled for about 3 months in the spent fuel bays before being sent to hot cells for a post-irradiation examination (PIE).

TEST MATRIX

The intent of the Parallex tests is to fabricate and irradiate CANDU MOX fuel with a limited number of parameters controlled within specific ranges. The variables for the test include the 2 loadings of Pu proposed for the initial Bruce A fuel (1.6% and 3.1%), 2 levels of PuO_2 homogeneity, and a range of linear heat ratings to cover the complete envelope expected for MOX fuel in the Bruce reactors. Other fuel characteristics that could be expected to affect the fuel performance will be carefully controlled or monitored, or both, but will not be intentionally varied.

The level of PuO_2 homogeneity attainable by the various commercial MOX fabrication processes has historically been a basis for claiming superiority of one process or another. The primary incentive for attaining a very finely dispersed PuO_2 mix in LWRs, the ability to dissolve the fuel easily in reprocessing, does not apply here; however, below some level of homogeneity it is believed that the PuO_2 distribution could affect CANDU MOX fuel performance. Although the PuO_2 homogeneity within the range produced by the current commercial MOX processes does not appear to affect the LWR fuel performance, the peak linear heat ratings for the CANDU fuel in the Bruce A reactors are about 55 kW/m compared with typical peak ratings of about 30 kW/m in LWRs. Large PuO_2-rich areas could affect the burnup threshold where increasing gas release begins or could produce hot spots on the cladding. On the other hand, arbitrarily setting a very tight specification on homogeneity could possibly eliminate one of the existing commercial processes from consideration or could add unnecessarily to MOX powder processing costs.

The degree of PuO_2 homogenization in the UO_2 matrix will be intentionally varied during fuel fabrication by the method of blending and milling the fuel mix. This can be accomplished in both laboratories with existing equipment. In both levels of homogeneity, a master mix containing 10% PuO_2 will be blended and ball-milled. This master mix will then be blended down to the required compositions of 1.6% and 3.1% Pu in ball mills (for a short time) and a conventional V-blender. This technique is expected to produce a relatively good PuO_2 homogeneity (termed intermediate homogeneity in this test). For the high homogeneity pellets, the entire mix will be given a final high intensity milling following the dilution and V-blending. The high intensity milling method is vibratory milling with steel balls at LANL and magnetically oscillated steel needles at Bochvar. The homogeneity produced in each case will be characterized with the best available techniques. These data will be used later in establishing specifications for the mission fuel.

The test matrix, shown in Table 1, thus requires that 4 batches of fuel be produced from 1 master mix from each fabricator. Table 2 shows the numbers of elements of each type and the total quantity of MOX from each fabricator. Eight elements will be removed from each of the demountable bundles after about 5 MW·d/kg HE burnup and replaced with fresh elements. The remainder of the test will be irradiated to the peak burnup of about 15 MW·d/kg HE which is expected in the Bruce A reactors. The removal and replacement will give intermediate burnup data on the 1.6 % Pu elements at about 5 and 10 MW·d/kg HE.

Other fuel characteristics that may affect performance but which will not be intentionally varied include pellet surface finish, impurity content, sintered density, pore size and distribution, and oxygen-to-metal ratio (O/M ratio). The density, pore distribution, O/M ratio and other parameters are not expected to be affected greatly by the PuO_2 additions, and thus the normal UO_2 CANDU pellet specification will be imposed. The surface finish may be affected because dry centreless grinding is desirable for the MOX pellets, whereas wet centreless grinding is normally used for the CANDU pellets. The surface finish is important because it influences heat transfer from the pellet to the cladding. It is hoped that the dry grinding will produce a finish that meets the usual specification; however, this requirement may be relaxed if necessary. The impurity content is of concern primarily because of the gallium that is present as an alloying agent in the WPu. There is no experience to indicate whether gallium would be a problem. It has the potential to affect in-reactor performance of the fuel through mechanisms such as stress-corosion cracking of the cladding, or the microstructural evolution of the fuel. Preliminary evidence shows that much of the gallium is removed during normal fabrication during lubricant removal and sintering. Gallium content will be carefully monitored in this test, but will not be an intentional variable. Because the Pu feedstocks will be from weapons material, the only other major impurity is expected to be iron pickup from the milling media. This and other impurities will be monitored.

POST-IRRADIATION EXAMINATION

An important output from the PIE tasks will be a summary of the Parallex MOX fuel performance as a function of the homogeneity levels and the various power levels during the NRU irradiation. Conclusions will be drawn about performance that would be expected under conditions in the Bruce A reactors.

All the PIE data will be compared to pre-irradiation characterizations of the elements and data for archive samples to quantify changes during irradiation, and to assess whether these changes are consistent with the expected changes based on prior knowledge of CANDU fuel performance. The data on irradiation conditions will be used to perform computer code fuel performance simulations to predict measured parameters such as gas release, strain, void volumes, and grain size. The examination will include

- Visual Examination and Profilometry: Each element will be examined visually along its length with a stereo microscope. Element diameters and profiles will be measured over the entire length at 3 orientations.
- Axial Gamma Scans: Fuel elements from each bundle will be axially scanned for gross gamma activity and isotopic activities to check for flux peaking and pellet-to-pellet gaps.
- Element Puncture and Fission-Gas Analysis: Fuel elements will be punctured to determine gas volumes and end-of-life internal pressures. A sample of the gas from 5 of the 10 elements will be analyzed by mass spectrometry to measure the fission-gas composition and the Xe and Kr isotopic composition.
- Ceramographic and Metallographic Examination: Samples will be cut from the axial midplane of fuel elements from each bundle for optical microscope examination of the fuel and sheath. Samples will also be cut from both ends of some of the elements. Oxide thickness will be measured, and fuel will be chemically etched to examine the grain growth, porosity, and condition of the fuel sheath interface. Finally, the sheath will be etched to examine the microstructure and hydride distribution. Alpha and beta autoradiography will be applied.
- Burnup Analysis: Samples will be cut from the peak flux position for burnup measurement. The fuel is chemically dissolved and burnup is determined by high-performance liquid chromatography, which uses ^{139}La as a fission monitor.
- Sheathing Hydrogen Analysis: Samples will be cut for analysis of the hydrogen content in the sheathing. The fuel is removed using a combination of chemical and mechanical methods. The hydrogen content of each sample is determined by vacuum extraction mass spectrometry.
- Scanning Electron Microscope (SEM) Examinations: Fuel cross sections from 5 elements will be examined in a shielded SEM to check for Pu homogeneity in the fuel matrix. Image analysis will be used to quantify the Pu homogeneity in the irradiated fuel.
- O/M and Microdensity Measurement: Thin cross sections of fuel from 2 elements will be cut into small samples at several radial positions using a numerically controlled saw. These samples will have their densities determined using an immersion technique with a high-precision micro-balance to yield information on densification and swelling. O/M ratios will be measured on these samples using coulometric titration.

STATUS

Detailed design of the NRU irradiation tests has been completed, including physics assessments of the fuel element powers and burnups in the NRU reactor. Drawings and specifications were prepared by AECL and transferred to LANL and Bochvar.

Procurement of Zircaloy hardware and equipment is mostly complete. Canadian import and export permits were obtained for shipments related to the project. The MOX fabrication processes to be used were defined, and manufacturing and inspection plans were prepared. MOX fabrication activities at LANL have produced intermediate homogeneity fuel for the first bundle (BD-1). Following approval of the US export permit, this fuel is expected to be shipped to Canada in early 1998. Contractual negotiations with Russian organizations were completed with the signing of the AECL-Bochvar contract on September 23, 1997. The commencement of MOX fabrication at Bochvar is expected near the end of 1997, with delivery of MOX to CRL in mid-1998. The irradiation tests will commence shortly after receiving the required MOX from LANL and Bochvar, in mid-1998.

SUMMARY

The Parallex Project is an important first step towards demonstrating the feasibility of dispositioning weapons-derived Pu as MOX fuel in CANDU reactors. Many aspects of a large-scale plutonium dispositioning mission are being demonstrated as the project proceeds. It is expected that irradiation testing of the US and RF MOX fuel will begin in the NRU reactor at CRL in mid-1998. The performance data from these tests will be applicable to qualifying MOX fuel for use in CANDU reactors.

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Table 1. Parallex Test Matrix

Bundle	Pu , wt %	Homogeneity	Power Rating[a]
BD-1	3.1	high, intermediate	low, intermediate, high
BD-2	3.1	high, intermediate	low, high
DM-1	1.6	high, intermediate	high
DM-2	1.6	high, intermediate	intermediate

[a] The power ratings are classified as "low" (35-45 kW/m), "intermediate" (45-55 kW/m) and "high" (55-65 kW/m).

Table 2. Quantity of Elements and MOX From Each Fabricator

MOX Type	Number of Elements	kg MOX
1.6 % Pu, intermediate homogeneity	14 (8+4+2)[a]	8.2
1.6 % Pu, high homogeneity	14 (8+4+2)	8.2
3.1 % Pu, intermediate homogeneity	9 (8+1)[b]	5.3
3.1 % Pu, high homogeneity	9 (8+1)	5.3
Totals	46	27

[a] 8 inserted originally, 4 replacements, and 2 archives.

[b] 8 inserted plus 1 archive.

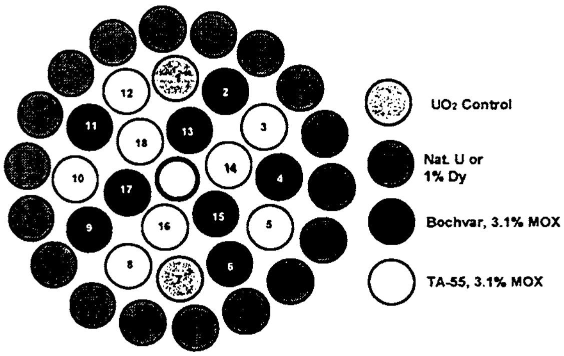

Figure 1. Schematic cross section of a fixed bundle for testing in the NRU reactor. The central tube is the guide tube and the next two rings are the experimental elements. Bundle BD-1 will contain natural UO_2 in the outer elements and BD-2 will contain depleted UO_2 with 1% dysprosium.

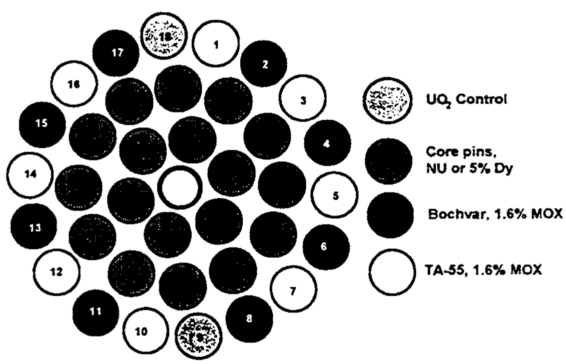

Figure 2. Schematic cross section of a demountable bundle. The central tube is the guide tube, the core elements are the next 2 rings, and the outer ring contains the 18 removable elements. DM-1 is to contain natural UO_2 core elements and DM-2 is to contain depleted UO_2 with 5% dysprosium. See Table 1 for definition of DM-1 and -2.