DEVELOPMENTS IN CANDU MOX FUEL FABRICATION

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ABSTRACT

As a strategic component of its advanced fuel cycle program, AECL continues to implement the MOX fuel development program involving MOX fuel fabrication and characterization, irradiation testing, post-irradiation examination, as well as reactor physics and fuel management studies. AECL performs its MOX fuel fabrication activities in the Recycle Fuel Fabrication Laboratories (RFFL) located at the Chalk River site. The RFFL facility is designed to handle alpha-active fuel material and produce experimental quantities of MOX fuel for reactor physics tests and demonstration irradiations.

From 1979 to 1988, several fabrication campaigns were conducted in the RFFL, producing close to two tonnes of MOX fuel with various compositions. RFFL operations were suspended from 1989 until 1994, at which time the facility was needed to fabricate MOX fuel for physics testing in the ZED-2 reactor. After completion of an extensive rehabilitation and re-commissioning of the RFFL, MOX operations were resumed in the facility in August 1996. An up-to-date description of the facility, including the fabrication process and the associated equipment, as well as the upgraded safety systems and laboratory services, is presented.

Since the resumption of MOX operations in the RFFL in 1996, several MOX fuel fabrication campaigns have been conducted in the facility; increasing the total amount of MOX fuel fabricated to-date in the RFFL to about three tonnes of MOX fuel. An overview of each of the fabrication campaigns is discussed. The fabrication processes used to manufacture the fuel from the starting powders to the finished elements are summarized. The various fabrication campaigns involved different technical requirements mainly due to the different intended uses of the fuel, i.e., test irradiations in NRU, physics tests in ZED-2, and dissolution experiments in support of the waste management program. Fabrication data including production throughputs and typical inspection results are discussed, including characterization techniques that were developed during the campaigns.

INTRODUCTION

Research and development activities on Pu-containing mixed-oxide (MOX) fuel have been conducted by Atomic Energy of Canada Limited (AECL) at its Chalk River Laboratories (CRL) site since 1960, and they remain a strategic part of AECL's advanced fuel-cycle program. The

program includes MOX fuel fabrication development, irradiation testing, post-irradiation examination (PIE), as well as reactor physics and fuel-management studies.

AECL's MOX fuel fabrication activities are performed in the Recycle Fuel Fabrication Laboratories (RFFL) at CRL. The RFFL facility is designed to produce experimental quantities of CANDU mixed-oxide fuel for reactor physics tests or for demonstration irradiations [1].

Since the start-up of the RFFL in the mid-1970s, a number of MOX fuel fabrication campaigns were conducted in the RFFL, producing various quantities of fuel with different compositions. After a stand-by period of about 5 years, a state where no fuel fabrication activities were conducted but the monitoring and ventilation systems in the facility were maintained, a project to rehabilitate the RFFL and bring it back into production was initiated in 1994. Rehabilitation and re-commissioning of the RFFL was completed in August 1996, and MOX operations were resumed in the facility with the production of thirty-seven $(U,Pu)O_2$ bundles destined for reactor physics tests [2].

To date, about 5000 individual fuel elements, equivalent to over 160 bundles and containing close to 3 t of MOX, have been fabricated in the RFFL (Table 1).

MOX FUEL FABRICATION

Commercial Fabrication Processes

The commercial processes employed in the production of MOX fuel follow conventional UO_2 practice, i.e., pelleting, sintering, grinding and loading into pins or rods with two main special considerations – one is due to the presence of Pu, i.e., the need to confine essentially all operations inside glove boxes, and the other is the need to blend the UO_2 and PuO_2 powders.

The two main commercial processes in use for the fabrication of MOX fuel are the MIMAS (MIcronizing MASter blend) process developed by Belgonucleaire and used by Belgonucleaire and Cogema, and the SBR (Short Binderless Route) process developed and used by BNFL [3]. Both processes are based on physical blending of UO_2 and PuO_2 powders, and both were developed to produce a homogeneous blend of mixed oxide powder that can be fabricated into sintered pellets that meet fuel specifications and that can be subsequently dissolved during reprocessing after irradiation.

The MIMAS process is characterized by a two-stage blending process [4]. The primary blend is achieved by micronizing or milling (typically using a ball mill) a high (about 20-30%) plutonium concentration master mix of UO_2 and PuO_2. The final desired plutonium concentration is achieved by blending the appropriate proportions of master mix and UO_2. In the SBR process [5], a single-stage blending process is achieved by attrition milling the UO_2 with the plutonium at the concentration desired in the final fuel. The mixed oxide powder is then spherodized for pressing into pellets.

Fabrication Process in the RFFL

Essentially, the processes employed in the RFFL are similar to the commercial fabrication processes. The fabrication process adopted in the RFFL is outlined in Figure 1. Depending on the requirements, the starting PuO_2 may first be conditioned either through a simple sieving operation to obtain a specific size distribution of PuO_2 particles or a milling operation using a vibratory mill. Blending of the PuO_2 and UO_2 powders is achieved through various combinations of three pieces of blending equipment – the vibratory mill, a high-intensity mixer, and a low-intensity (Turbula) blender (Figures 2-4). The high-intensity mixer can also be used as a low-intensity mixer for powder quantities up to a few hundred grams (Figure 3), whereas the Turbula blender is primarily used for kg quantities of powder. These options provide a high degree of flexibility in terms of the homogeneity that can be achieved in the fuel being fabricated, from a relatively inhomogeneous distribution of pure PuO_2 particles within the UO_2 matrix (resulting from a single-stage blending process using the low-intensity blender) to a homogeneous distribution of Pu in solid solution within the matrix (as achieved by vibratory milling the PuO_2 and UO_2 powders together).

After blending, the MOX powder is pre-pressed using an isostatic press, to convert the powder into compacts, which are, in turn, fed into a granulator. Zinc stearate (for die lubrication) is mixed in with the resulting free-flowing granules, which are then suitable for final pressing into green pellets using a single-cavity hydraulic press.

The green pellets are placed into molybdenum trays, and loaded into the furnace, where sintering is done in a dilute hydrogen cover gas (10% H_2 in N_2) at a temperature of 1700°C. Sintered pellets are then centreless ground to a specified diameter and surface finish. The pellets are washed and then dried in warm air. Acceptable pellets are formed into specified stack lengths, and loaded into sheaths. The end cap is welded to the loaded sheath using a gas tungsten arc welding (GTAW) system. The sealed elements are then helium leak-tested, scanned for surface alpha contamination, weighed, and visually and dimensionally inspected. The finished elements are shipped out of the facility or kept in temporary storage for subsequent use, e.g., for bundle assembly for an irradiation or physics test.

Fuel manufacturing entails a number of inspections to determine whether or not the product (i.e., fuel pellets, elements, and bundles) meet established technical and quality requirements. In some cases, these inspections are done in the same glove boxes where the fabrication activities are done, e.g., visual and dimensional examination of pellets, immersion density, etc. In other cases, the inspections are done in separate glove boxes or fumehoods in the RFFL, including ceramographic examination, measurement of oxygen-to-metal ratio of powders, alpha-autoradiography, and assay of samples for Pu content (by coulometry). Still, in other cases, some inspections are done outside the RFFL such as impurity and isotopic composition measurement of powders or pellets (although sample preparation for all chemical analysis is done in the RFFL), scanning electron microscopy of fuel samples, and X-radiography of welds. Actual inspections done on the fuel are based on customer requirements established before the fabrication campaign is started.

Occasionally, specialized operations are conducted in the facility such as loading and recovery of neutron-activated foils from fuel elements used in reactor physics testing, and inspection and repackaging of Pu-containing packages from CRL's storage facilities.

Production Throughput

In the RFFL, the batch-type fabrication process was originally designed to have a throughput of one 12 kg-batch of MOX fuel per day. During the fabrication campaign in 1996-1997 to produce thirty-seven bundles for reactor physics tests in ZED-2, production throughput averaged 0.6 batches per day (each batch weighing 11 kg MOX), with a peak throughput of 1.2 batches per day (about 13 kg MOX). Overall, 77 batches of MOX fuel totaling about 850 kg of MOX fuel pellets were fabricated and loaded into more than 1370 finished fuel elements over a period of 26 weeks.

Operations Quality Assurance

The RFFL is a Canadian Nuclear Safety Commission-licensed nuclear facility that comes under the AECL Nuclear Operations Quality Management Program. A facility-specific Conduct of Operations Manual was developed to meet the requirements of the CAN/CSA N286.5-95 standard, and the Manual describes the facility organization, responsibilities, processes, and control measures implemented to ensure operational safety. The Conduct of Operations Manual contains the full range of measures for management actions, performance and verification functions, program assessment and corrective functions, and documents and records control.

A separate QA Plan that describes the quality program used by the RFFL to comply with the requirements of the CAN3-Z299.2-85 and ISO 9002:1994 QA standards was established to ensure the quality of the MOX fuel elements and bundles. The QA program is supported by documented operating procedures, both for normal and emergency operations. Thus, the QA program, as a whole, describes the measures implemented in the RFFL to ensure both operational safety and product quality.

Operational Radiation Protection

The overarching principle in the management of radiological exposure is that personnel doses be kept as low as reasonably achievable, economic and social factors taken into account (ALARA principle). These doses should also be kept below the limits in applicable regulations.

The major components of the Radiation Protection Program as applied in the RFFL are as follows:

- Staff classification and training – staff members are classified into appropriate groupings based on their duties and responsibilities with respect to radiation protection, and they receive appropriate training to become fully qualified within their designated group.

- Zoning - the facility is divided into zones of different contamination hazard, with each zone having its associated controls such as access control, dosimetry, protective clothing, and radiation and contamination monitoring.
- Work Control – work in the facility is performed in accordance with authorized procedures, protocols, work plans or work permits.
- Workplace Air Monitoring - a system of alpha continuous air monitors (CAMs) is distributed through the facility and gives a prompt warning of any unexpected increase in airborne alpha activity in the workplace.
- Exposure Monitoring – external doses are monitored through the use of personal theromoluminescent dosimeter (TLD) photobadges. Additional dosimeters such as extremity TLDs, personal alarming dosimeters, or pocket ionization chambers are used when required. Internal doses are monitored through the use of Personal Air Samplers (PAS) for all staff doing work in the facility. This practice is combined with a bioassay program that involves annual whole body and lung counts and urine analysis. It is worth mentioning that the Pu-in-urine analysis by thermal ionization mass spectrometry (TIMS) currently done in CRL is capable of high sensitivity measurements of Pu-239 and Pu-240, down to femtogram quantities of Pu-239 or a few μBq of activity. This enables the dosimetry program to have the capability of detecting intakes of Pu at levels around one-twentieth of the annual limit of intake (ALI) or better [6].

RECENT FABRICATION CAMPAIGNS IN THE RFFL

ZED-2-96 Campaign

The ZED-2-96 fabrication campaign involved the production of thirty-seven MOX fuel bundles that simulate mid-burnup CANDU fuel. These bundles were used in physics tests in the ZED-2 reactor to validate the codes used to predict void reactivity. The simulation of mid-burnup natural uranium CANDU fuel required MOX fuel consisting of 0.30% Pu in depleted U (0.37% U-235) plus 0.05% dysprosium to simulate the fission products.

In accordance with the Manufacture, Inspection and Test Plan (MITP), the fabrication process adopted for the campaign started by sieving the PuO_2 through a 44-micron screen. The PuO_2 and the Dy_2O_3 was blended with the depleted UO_2 using a high-intensity mixer to produce a mastermix containing about 3 wt.% Pu. This mastermix was blended with more UO_2 using the low-intensity blender to arrive at the final concentration of the MOX powder. The MOX powder was then processed through the fabrication line using the standard operations of pelleting, sintering, centreless grinding, washing, drying, and loading into fuel sheaths.

Of prime importance in this campaign was the fuel composition - its actual value and the batch-to-batch consistency. To maintain control over this parameter, weights of the starting powders were strictly monitored and recorded. Chemical analysis (coulometry for Pu content; high performance liquid chromatography for Dy content) of the finished pellets was used to confirm the accuracy of the batch components. Of the 77 batches processed, there was one batch whose chemical analysis indicated a significant difference from the calculated values of Pu and

Dy contents. Further repeats of the chemical analysis confirmed the low Pu and Dy contents. A non-conformance was raised against this batch, and it was found during the ensuing investigation that a container of depleted UO_2 instead of recycled MOX was inadvertently added to the blend. This batch was not used in the experiments.

The two-stage blending process achieved sufficient homogeneity in the finished fuel required for the physics tests. The extent of homogeneity in the fuel was determined by ceramography and alpha autoradiography. As shown in Figure 5, a typical microstructure of the ZED-2-96 fuel consisted of a duplex microstructure, where Pu-rich areas (indicative of the original Pu particles) are randomly distributed within the matrix of uranium. The Pu-rich areas (ranging from 10 to 35 microns, with an average size of 20 microns) are characterized by an annular band of very fine grains and/or higher levels of porosity relative to the matrix. The random distribution of Pu particles within the uranium matrix was confirmed through alpha autoradiography (Figure 6).

^{238}Pu-Doped Pellet Campaign

Spent CANDU fuel contains approximately 0.4 wt% mixed Pu oxides, whose alpha (α) activity is due predominantly to ^{241}Am and $^{240/239/238}$Pu. However, spent fuel also contains high β/γ activities arising predominantly from fission products, which contribute to radiolytic processes that cannot be distinguished from α-processes. It was proposed to fabricate a series of unirradiated, depleted uranium CANDU fuel pellets containing various α activities that range from 1 to 100 times those found in irradiated CANDU fuel, which is conservatively assumed to be around 1 Ci/kg. This campaign involved the fabrication of nine depleted uranium pellets containing various amounts of ^{238}Pu to simulate various levels of α activity. These pellets were made into electrodes used to conduct electrochemical measurements. The purpose of doping the UO_2 with high α-activity is to investigate the effects of α-radiolysis on the oxidative dissolution properties of UO_2. This is necessary to ensure that the long-term dissolution processes of UO_2 are understood to enable models to be developed that will be used to conduct future performance assessment studies of a used-fuel disposal vault.

It was initially thought that pellets could be fabricated using the Pu oxide feedstock already available in the RFFL. However, given the calculated isotopic composition of the reactor-grade Pu, to fabricate UO_2 pellets containing the equivalent α dose rates of up to 100 times that of normal spent CANDU fuel would require 40 wt% Pu in MOX. This high PuO_2 content will likely compromise the experiment because of a significant change in the microstructure of the fuel. It was then decided that a high specific activity isotope, ^{238}Pu (17.1 Ci/g), be used as the additive. Three pellets at an α-activity level of 1 Ci/kg in CANDU fuel and 10 and 100 times this level were fabricated, for a total of 9 pellets at three additive levels. The corresponding Pu contents for each α-activity level were quite low - 0.00585, 0.0585 and 0.585 % ^{238}Pu in MOX.

The final UO_2 pellet was required to have a homogeneous distribution of Pu. The milling and blending processes employed in the RFFL for MOX fuel fabrication have been proven to result in a homogeneous distribution of Pu, i.e., a solid solution, for material quantities of several hundred grams. For this campaign, at 13 g per pellet, the total amount of material to be handled was

about 120 g, and the total amount of $^{238}PuO_2$ was only about 0.3 g. Each batch representing each dopant level comprised of only 40 g of MOX for 3 pellets. Given the small batch quantities for this fabrication campaign, utmost care was taken to address concerns with both homogenization of the mixture and material losses.

The first step of the manufacturing process was to weigh the as-received PuO_2 powder. The equivalent amount of UO_2 to obtain the required composition for the highest dopant level was weighed and blended with the as-received PuO_2 in the vibratory mill (Mix 1). A 40-g portion of Mix 1 was then obtained to press into three pellets. Additional UO_2 was put in the milling chamber with the remaining mix to make up Mix 2, which was subsequently milled. Again, a 40-g portion of Mix 2 was obtained to press into three pellets. More UO_2 was added to the remaining material to obtain the last mix, Mix 3, and then milled. All of the material in Mix 3 was then used to press three pellets. The green pellets were loaded into the furnace, where sintering is done in accordance to standard procedures. The sintered pellets were then cut into discs that served as electrodes for the electrochemical experiments.

Inspection activities consisted of weighing the as-received $^{238}PuO_2$, inspecting the green pellets after final pressing for defects as evidenced by the alcohol bubble test and for geometric pellet density, and inspecting the sintered pellets for weight, dimensions, and sintered density. The final inspection consisted of determining the suitability of the pellet discs as electrodes, which was based on the resistance measured during the Cu electroplating of the discs and the appearance of the Cu plating. In general, a resistance of 1 kΩ or lower and an evenly distributed and thin coating (thick coatings tend to delaminate) are indications of a good electrode.

Initial Cu plating results showed in many cases uneven or no evidence of plating, and resistances that were much higher than 1 kΩ (Figure 7). The low conductivity was possibly due to the slight hypostoichiometry of the pellets obtained during the reductive sintering process, which is promoted further by the presence of small amounts of Pu. It was decided to anneal the slices in the presence of a small amount of O_2 to induce a slight hyperstoichiometry and improve the conductivity. The annealed slices were then re-plated (the initial Cu plating was removed prior to annealing), and the results showed that resistances decreased to around 1 kΩ and the plating was more even (Figure 7), indicating that the slices were suitable for use as electrodes in the electrochemical experiments. These experiments are now being conducted in the facility, and are expected to yield data to enable better understanding of the dissolution processes of UO_2 that will be valuable in conducting performance assessment studies of a used fuel disposal vault.

BDL-446 Campaign

The BDL-446 experiment [7] was designed to investigate the effect of various levels of Pu homogeneity in the microstructure of MOX pellets on the irradiation performance of the fuel. Previous investigations conducted under LWR conditions, have shown only a slight difference in the performance of the fuel as a function of pellet microstructure [8-10]. CANDU fuel operates at higher linear element powers than LWR fuel, and this experiment is intended to reveal whether the higher powers magnify the effects of varying microstructure observed in previous studies.

The objective of pellet fabrication was to produce fuel pellets with three distinctly different microstructures. Three different fabrication routes were chosen:

- Method 1: To obtain a solid solution following sintering, UO_2 and PuO_2 powders were vibratory milled at the desired final plutonium concentration.
- Method 2: To obtain a MIMAS-like microstructure, a homogenous, high-plutonium concentration (30 wt.% Pu in HE) master mix was prepared by vibratory milling. The final desired plutonium concentration was achieved by blending an appropriate amount of master mix with depleted UO_2 powder using the high-intensity blender.
- Method 3: To obtain a distribution of pure PuO_2 particles within the UO_2 matrix, pure PuO_2 powder was vibratory milled. The final desired plutonium concentration was achieved by blending an appropriate amount of milled PuO_2 powder with depleted UO_2 powder using a low-intensity blender.

The remainder of the fabrication process was identical for all fuel types and consisted of the standard practices of pre-compaction, granulation, final pressing, sintering, and centreless grinding. Ground pellets were inspected, and acceptable pellets were loaded into zircaloy sheaths. The sheaths were end-closure welded using the gas tungsten arc welder.

Wavelength-dispersive X-ray (WDX) analysis was performed on the various fuel types, in order to quantify the plutonium distribution throughout the pellet. Plutonium mapping of the entire pellet surface was conducted, as well as microprobe analysis of plutonium-rich regions in samples where they were detected. The results indicate that:

- Method 1 has produced a microstructure in which the PuO_2 and UO_2 are in solid solution. No plutonium-rich regions could be detected (Figure 8).
- Method 2 resulted in a microstructure consisting of master-mix particles evenly distributed in a UO_2 matrix (Figure 8). The plutonium-rich regions range in size from 15 to 150 μm. Microprobe analysis indicated that the plutonium-rich regions contain up to 30 wt% plutonium, which was consistent with the master-mix concentration.
- As expected, Method 3 resulted in a microstructure consisting of pure PuO_2 particles evenly distributed in a UO_2 matrix (Figure 8). The PuO_2 regions range in size from 30 to 300 μm. Microprobe analysis indicated that the plutonium-rich regions are pure PuO_2.

The bundle containing the pellets is currently being irradiated in the NRU reactor at CRL. The irradiation conditions were designed to operate the fuel under relatively high powers, but not approach conditions that could result in columnar grain growth. In this way, any performance differences in the fuel types due to microstructural differences would not be erased by significant changes in the microstructure. At the time of writing, the mid-plane, outer-element burnup is estimated at 480 MWh/kgHE, and the bundle will achieve its target terminal burnup of 500 MWh/kgHE in November 2003. Post-irradiation examination of this fuel is expected to start in mid-2004.

Parallex Campaign

The Parallex Project is a parallel experiment demonstrating the use of U.S. and Russian weapons-derived plutonium (WPu) in CANDU MOX fuel [11]. As part of the Parallex Project, three bundles containing MOX fuel made with WPu in both the U.S. and Russia are undergoing irradiation testing in NRU.

The Russian MOX fuel contained in the Parallex bundles was made using the milling process developed at the Bochvar Institute. When Russia proceeds with the actual mission to utilize WPu, a different fabrication technique is being considered – MIMAS. To enable testing of fuel made by the MIMAS process, the project decided to use MOX fuel fabricated in the RFFL, using civilian Pu (civPu). Although the actual MIMAS process was not used, a fabrication process that produces a pellet microstructure comparable to MIMAS, i.e., particles of UO_2 and PuO_2 in solid solution homogeneously dispersed within a UO_2 matrix, was employed. The overall Pu concentration in the AECL-simulated MIMAS fuel was increased to account for the reduced concentration of fissile isotopes in civPu compared to WPu. This approach broadens the parallel nature of the program to include a direct comparison of the two candidate fabrication processes that have potential to be used in Russia.

The MOX fuel manufactured in the RFFL consisted of civilian PuO_2 and depleted UO_2 at a concentration of 5.3% PuO_2 in $(U+Pu)O_2$. The higher Pu content of the MOX fuel to be fabricated in the RFFL was chosen to match the fissile content of the MOX fuel from Russia containing 4.6% WPu (amount of fissile isotopes in WPu is higher than civilian Pu).

The blending method started with vibratory milling the mastermix containing equal amounts of UO_2 and PuO_2 powders. An appropriate amount of additional UO_2 powder was then added to the mastermix to achieve the desired final Pu concentration. To achieve a different size distribution of the Pu-rich areas, two blending methods were implemented during preparation of the final blend: one was low-intensity mixing, and the other was low-intensity mixing followed by a short period of high-intensity mixing. The latter was expected to achieve smaller-sized Pu-rich areas compared to the former.

The resulting distribution of the Pu-rich areas for the different types of MIMAS-simulated fuel is shown in Figure 9. As expected, the Pu-rich regions in the blend that was processed by low-intensity blending alone were larger than the ones processed by low-followed-by-high-intensity blending. Plutonium-rich regions in both types of fuel were limited to the mastermix concentration, as determined by microprobe analysis.

The MOX fuel fabricated in the RFFL was incorporated into two bundles configured such that MOX fuel made in Russia via the Bochvar process is tested side-by-side with the fuel made in the RFFL via the simulated MIMAS process. Both bundles are undergoing irradiation testing in NRU and are expected to achieve their target burnups in two years.

CONCLUSION

The RFFL has undergone an extensive rehabilitation resulting in upgraded safety systems, refurbished process equipment, up-to-date quality program and fully qualified technical staff. Several fabrication campaigns have been conducted in the facility since resumption of MOX operations in 1996, producing MOX fuel with various compositions and for various applications ranging from physics and irradiation testing to electrochemical dissolution for waste management studies. The facility's production capability is demonstrated to be highly flexible to satisfy widely differing requirements, from a high throughput production of several hundred kilograms of fuel to a very detailed, meticulous fabrication effort involving only a handful of pellets. A variety of inspection techniques have also been developed and implemented to characterize the fuel, including chemical and isotopic composition determination, SEM and microprobe analysis and alpha autoradiography to determine Pu homogeneity, and measurement of resistivity of pellet samples to name a few.

Preparations are now underway for the next fabrication campaign to produce 35 MOX fuel bundles for physics testing as part of the Advanced CANDU Reactor (ACR) program. The successful completion of several fabrication campaigns in the RFFL demonstrates the capability of this strategically important facility.

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REFERENCES

[1] CARTER, T.J., "The Recycle Fuel Fabrication Laboratory at Chalk River", Proc. Int. Conf. CANDU Fuel, October 6-8, 1986, Chalk River, Ontario, Canada, (I.J. Hastings, ed.), Canadian Nuclear Society, Toronto (1986), pp. 168-176.

[2] DIMAYUGA, F.C., "Fabrication of Simulated Mid-Burnup CANDU Fuel in the RFFL", Proc. Fifth Int. Conf. CANDU Fuel, vol. 1, September 21-25, 1997, Toronto, Ontario, Canada, (J.H. Lau, ed.), Canadian Nuclear Society, Toronto (1997), pp. 289-299.

[3] ROBERTS, A., "Progress in MOX Fuels Fabrication", Proc. Global '97 International Conference on Future Nuclear Systems, Yokohama, Japan, October 5-10, 1997.

[4] HAAS, D., "MOX Fuel Fabrication Experience at Belgonucleaire", in Recycling of Plutonium and Uranium in Water Reactor Fuel, Tech. Comm. Meeting, U.K., 1995, IAEA-TECDOC-941, pp. 77-89.

[5] EDWARDS, J. et al, "BNFL Supply of MOX Fuel Assemblies to the Beznau 1 PWR of NOK", in Recycling of Plutonium and Uranium in Water Reactor Fuel, Tech. Comm. Meeting, U.K., 1995, IAEA-TECDOC-941, pp. 57-67.

[6] LINAUSKAS, S.H. et al, "Performance Characteristics of a Bioassay Method for Plutonium Using TIMS", AECL-12157, 2003 January.

[7] HAMILTON, H. and DIMAYUGA, F.C., "Fabrication of Experimental MOX Fuel Pellets with Varying Microstructure", Proc. Seventh Int. Conf. CANDU Fuel, vol. 2, September 23-27, 2001, Kingston, Ontario, Canada, (B.J. Lewis, ed.), Canadian Nuclear Society, Toronto (2001), pp. 3C-11 – 3C-24.

[8] M. BILLAUX and J. VAN VLIET, "Impact of Fuel Heterogeneities on Fission Gas Release for LWR U-Pu Mixed Oxide Fuels", International Atomic Energy Agency, Proceedings of a specialists' meeting organized by the IAEA and held in Bowness-on-Windermere, United Kingdom, 1984 April 9-13, pp. 441-448, Dec. (1984).

[9] M.D. FRESHLEY, D.W.BRITE, J.L.DANIEL, P.E.HART and R.K.MARSHALL, "Plutonia Fuel Study Final Report", EPRI Report, EPRI-NP-637, January (1978).

[10] M. LIPPENS, C. VAN LOON and J. KETELS, "Ceramographic Techniques and Electron Probe Micro-Analysis Applied to Uranium-Plutonium Oxides", International Atomic Energy Agency, proceedings of a technical committee meeting held in Vienna, 1986 October 13-16, 135, pp. 69-76, May (1987).

[11] DIMAYUGA, F.C. et al, "Status of the Parallex Project – Testing CANDU MOX Fuel with Weapons-Derived Plutonium", Proc. Seventh Int. Conf. CANDU Fuel, vol. 2, September 23-27, 2001, Kingston, Ontario, Canada, (B.J. Lewis, ed.), Canadian Nuclear Society, Toronto (2001), pp. 4B-3 – 4B-15.

TABLE 1. Fuel Fabrication Campaigns Conducted In The RFFL

CAMPAIGN	DATE	FUEL TYPE	QUANTITY (kg MOX)
BDL-419	1979-80	(U, 0.5% Pu)O_2	15, 36-element bundles (320 kg)
BDL-422	1981-83	(Th, 1.75% Pu)O_2	6, 36-element bundles (120 kg)
BDL-430	1982	Natural ThO_2	1, 36-element bundle (20 kg)
WR1-1012	1982	(Th, 1.8% ^{235}U)O_2	2, 21-element bundles (20 kg)
WR1-1012	1982	(Th, 2.3% Pu)O_2	2, 21-element bundles (20 kg)
WR1-1010	1982-85	(Th, 2.3% Pu)O_2	1332 elements (650 kg)
BDL-432	1986-88	(Th, 1.4% ^{233}U)O_2	1350 elements (700 kg)
ZED2-96	1996-97	(U, 0.3% Pu)O_2	37, 37-element bundles (850 kg)
^{238}Pu-Doped	1999	(U, 0.006-0.6% Pu)O_2	9 pellets (120 g)
BDL-446	2000	(U, 1.35% Pu)O_2	1, 42-element bundle (23 kg)
Parallex	2001	(U, 5.3% Pu)O_2	20 elements (14 kg)

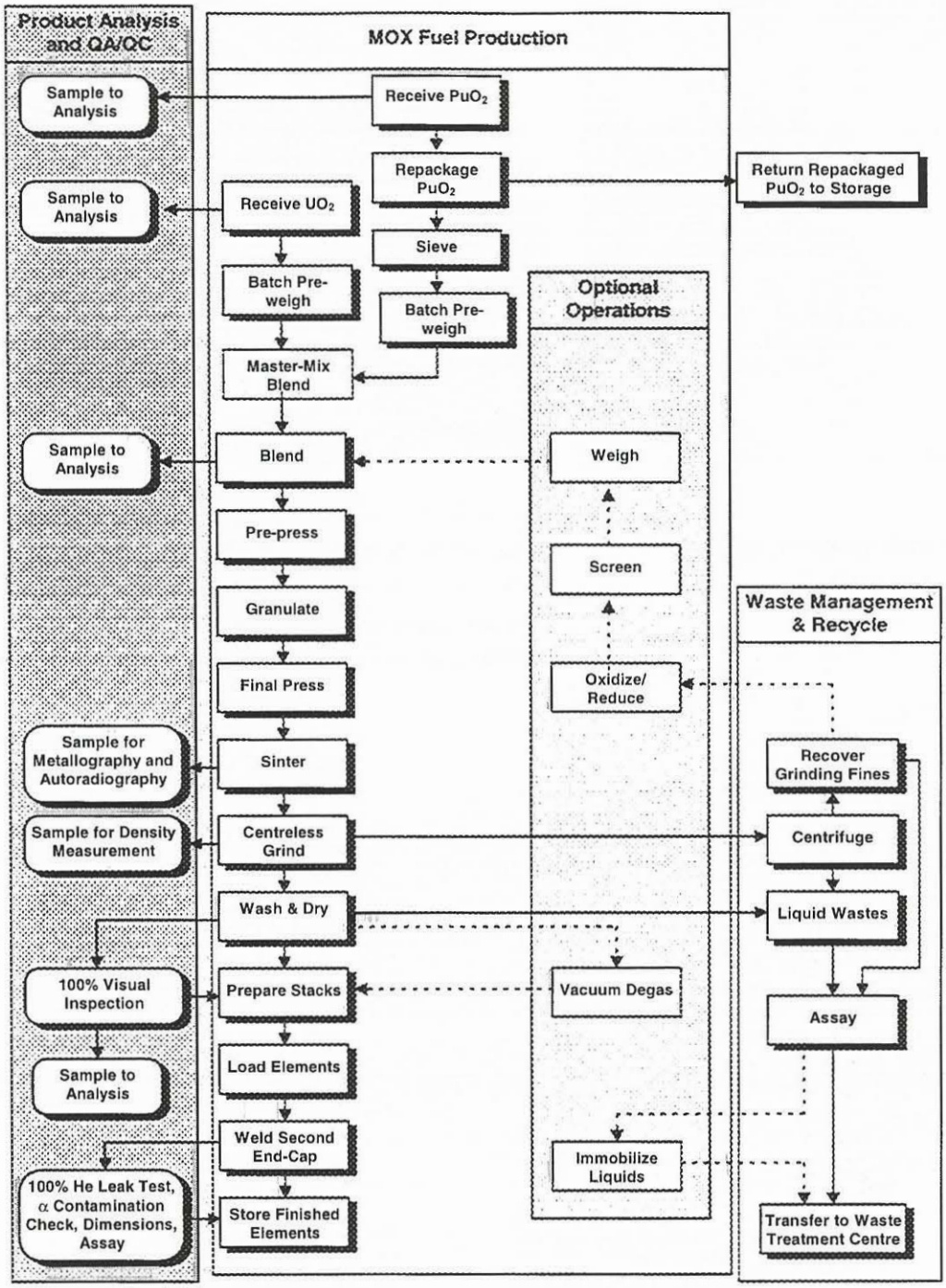

Figure 1. RFFL Fabrication Process Flowsheet

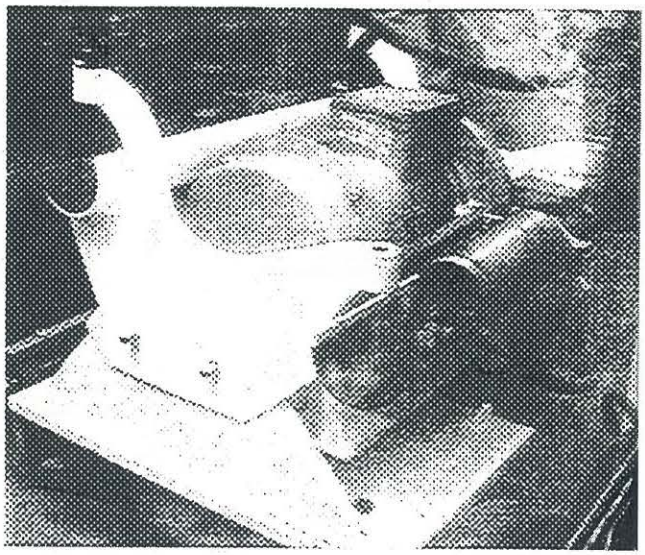

Figure 2: Vibratory mill showing milling chamber and grinding media.

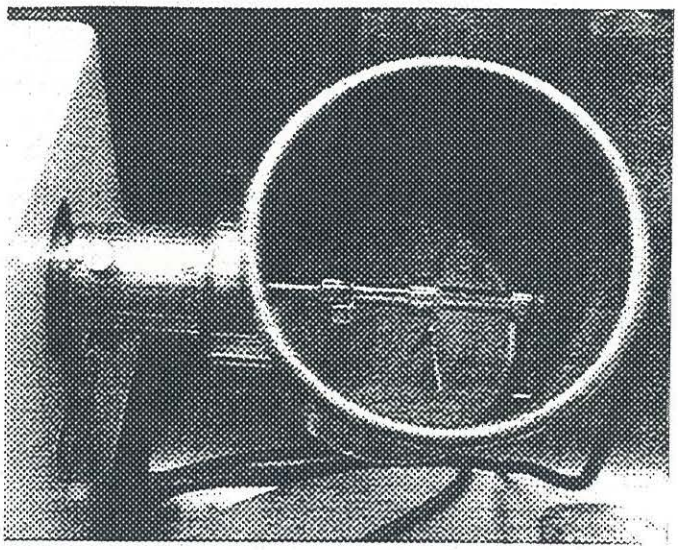

Figure 3. Blender used for high-intensity blending achieved by tumbling the powders while the intensifier blades are rotating at high speed. This blender could also be used for low intensity blending by not operating the intensifier blades.

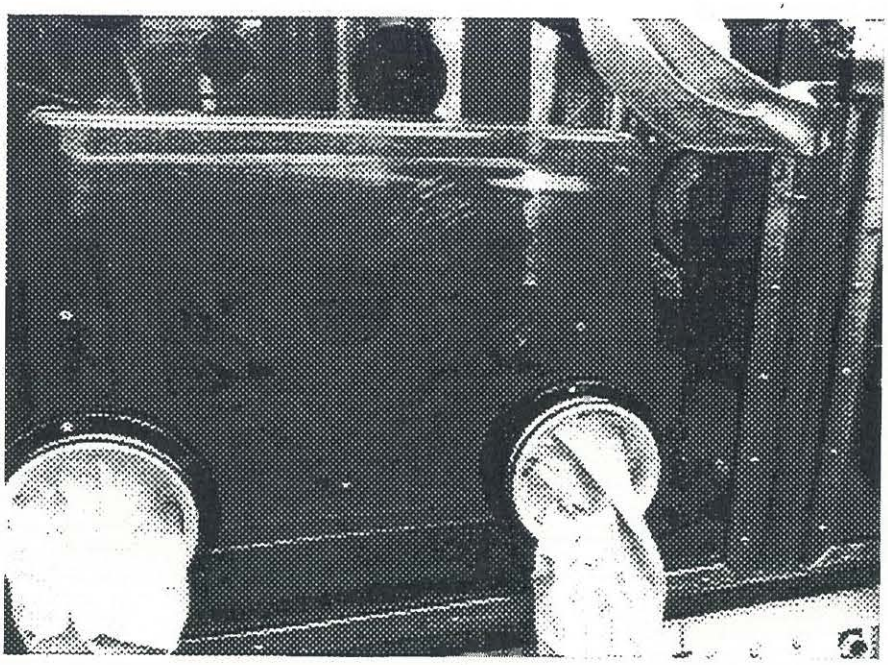

Figure 4. Turbula blender used for large-scale (up to 12 kg) low-intensity blending of fuel powders.

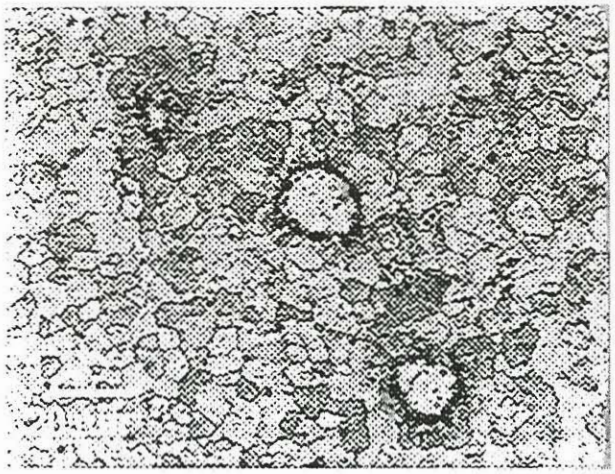

FIGURE 5. Micrograph showing a typical microstructure of the MOX fuel fabricated during the ZED-2-96 campaign. Note the Pu-rich areas characterized by an annular band of very fine grains and/or porosity

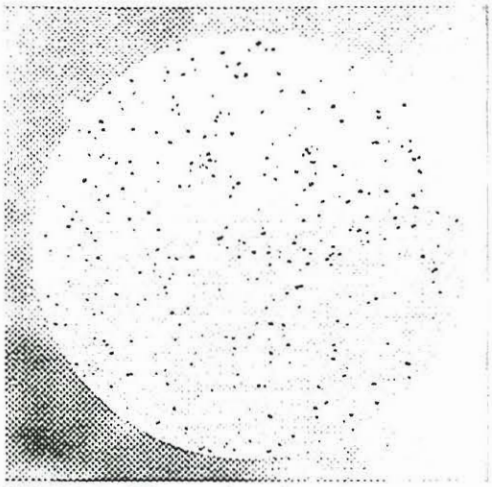

FIGURE 6. Photo showing the positive print of a typical alpha autoradiograph taken from fuel samples produced during the ZED-2-96 campaign. The black spots represent the Pu particles

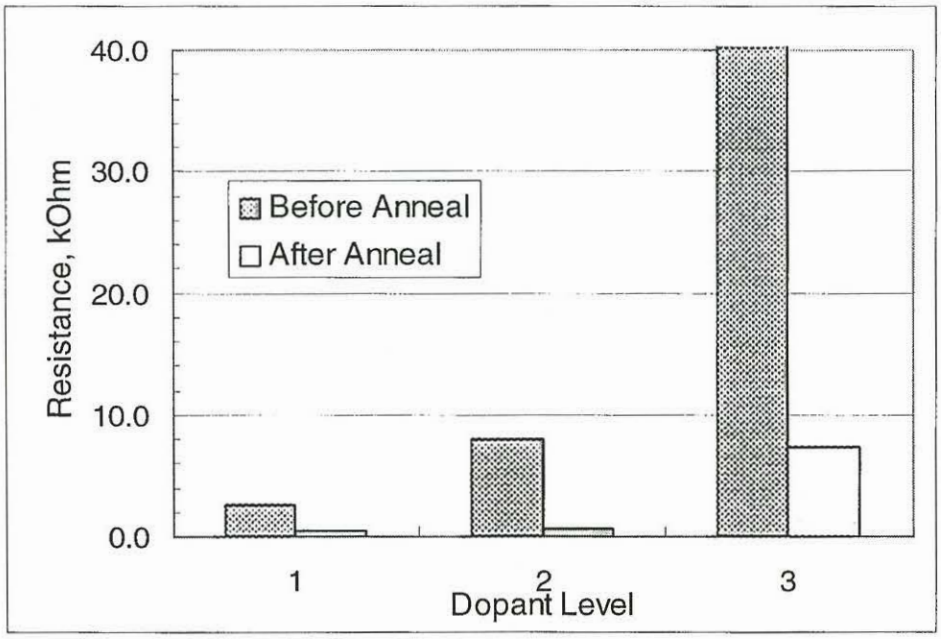

Figure 7. Plot of average resistances of ^{238}Pu-doped pellet slices before and after annealing in the presence of a small amount of O_2. Dopant levels 1-3 correspond to α-doses of 1-100 times the normal α-dose found in spent CANDU fuel. Note: the average resistance for dopant level 3 before annealing was actually off-scale at around 63 kΩ.

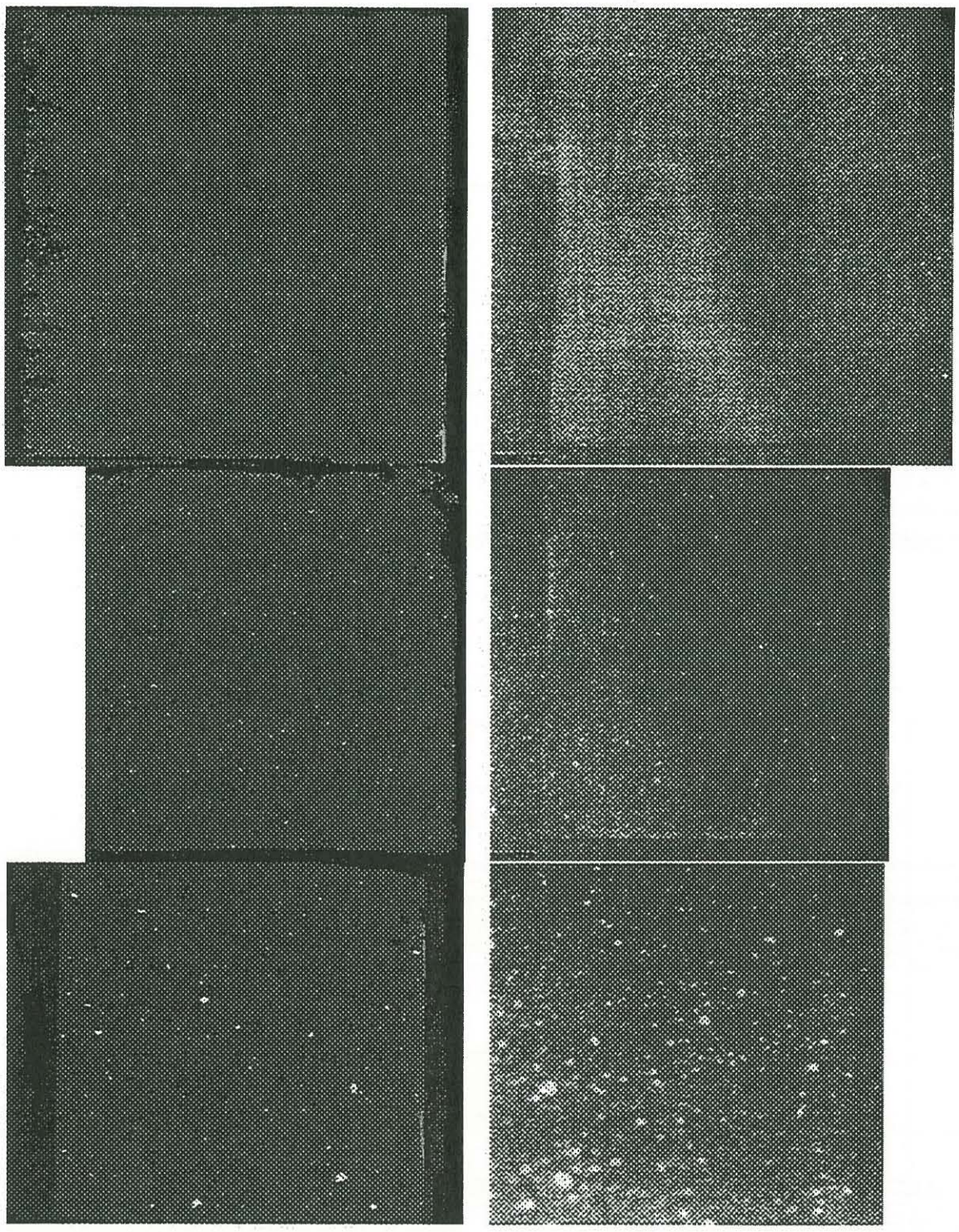

Figure 8. SEM plutonium mapping (left) and alpha-autoradiography (right) results for the three fuel types fabricated for the BDL-446 campaign. The light spots are the Pu-rich areas. Top-method 1, middle-method 2, and bottom-method 3.

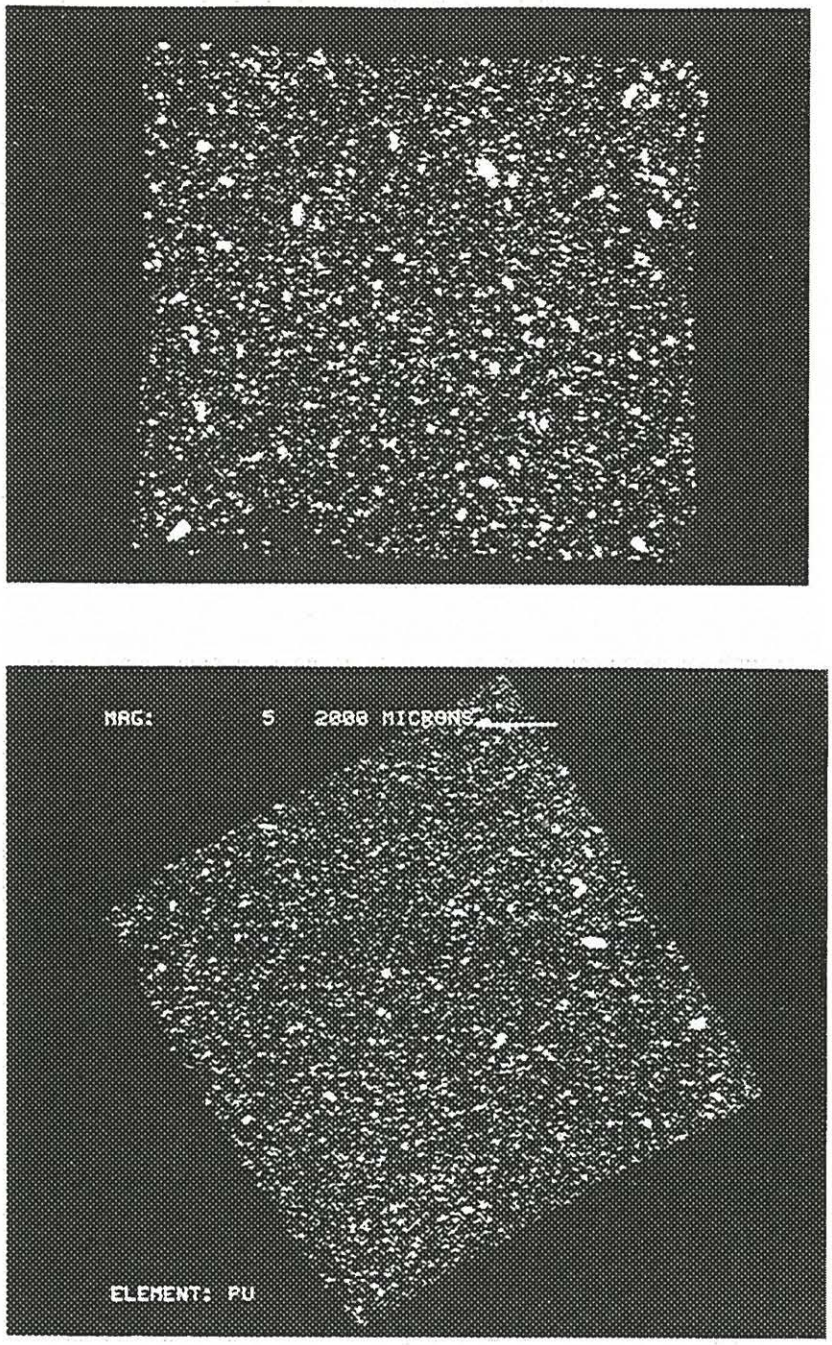

Figure 10. SEM plutonium mapping results for the two fuel types fabricated for the Parallex campaign. The light spots are the Pu-rich areas. Top – low-intensity blending alone, and bottom – low-then-high-intensity blending.