FABRICATION OF URANIUM DIOXIDE FUEL PELLETS IN SUPPORT OF A SLOWPOKE-2 RESEARCH REACTOR HEU TO LEU CORE CONVERSION

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Abstract

The International Centre for Environmental and Nuclear Sciences (ICENS) at the University of the West Indies in Jamaica operates a SLOWPOKE-2 research reactor that is currently fuelled with highly-enriched uranium (HEU). As part of the Global Threat Reduction Initiative, Atomic Energy of Canada Ltd. has been subcontracted to fabricate low-enriched uranium (LEU) fuel for the ICENS SLOWPOKE-2. The low enriched uranium core consists of a fuel cage containing uranium dioxide fuelled elements. This paper describes the fabrication of the low-enriched uranium dioxide fuel pellets for the SLOWPOKE-2 core conversion.

1. Introduction

The SLOWPOKE-2 (Safe LOW Power Kritical Experiment) is an Atomic Energy of Canada Ltd. (AECL) designed research reactor with a maximum thermal power of 20 kW and a maximum neutron flux in an inner irradiation site of 1×10^{12} $cm^{-2}s^{-1}$. A schematic of the reactor core is shown in Figure 1. The reactors are mainly used for neutron activation analysis, neutron radiography (where equipped), and nuclear education [1]. Of the eight SLOWPOKE-2 reactors built, five remain in operation [2]. The original cores for most of the reactors were fuelled with highly-enriched uranium (HEU) alloyed with, and clad in, aluminum. One of the SLOWPOKE-2 reactors has since been converted to a low-enriched uranium (LEU) core consisting of uranium dioxide (UO_2) fuel pellets clad in Zircaloy-4, while the remainder of the reactor cores (with the exception of the reactor at the Royal Military College in Kingston, Ontario, fuelled with LEU) are of the original design [1]. The SLOWPOKE-2 reactor located at the International Centre for Environmental and Nuclear Science (ICENS) at the University of the West Indies in Jamaica is one of the remaining reactors with an HEU fuelled core.

This paper presents details of the fabrication of the UO_2 fuel pellets for the core conversion.

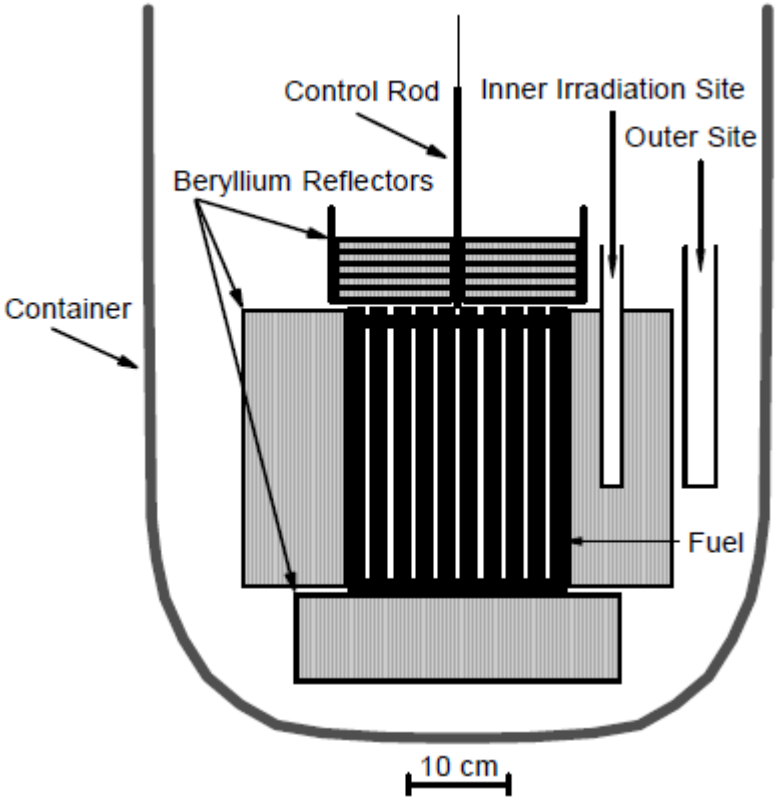

Figure 1 Schematic of a SLOWPOKE-2 Reactor Core [1]

2. Materials

Pellets were fabricated from customer-supplied UO_2 powder. The oxygen-to-metal (O/M) ratio of the powder was determined upon receipt using a thermogravimetric analyzer. The measured O/M for the powder was 2.06±0.01, within the specifications defined by [**3**]. Zinc stearate was used as a lubricant during final pressing of the UO_2 powder granules. The zinc stearate was mixed in with UO_2 powder granules in a double cone blender in the amount of 0.2 wt.%.

3. Preliminary Fabrication Trials

AECL was furnished with UO_2 powder to perform preliminary pellet fabrication trials. The as-received powder is pictured in Figure 2. The powder was in the form of free flowing granules with a green-brown colour. A series of fabrication trials were conducted to evaluate its sinterability.

In the first sinterability tests, the as-received powder was lubricated and loaded directly into the die. Green pellets were cold pressed to densities of 5.3-5.7 g/cm^3. The pellets were sintered in hydrogen at a temperature of 1650°C for two hours. Upon removal from the sintering furnace, the pellets were inspected visually and their densities were measured by geometric measurement and by immersion. Figure 3a shows the surface of one of the pellets after sintering. The surface of the pellet had a visibly rough finish and the pellet had randomly oriented surface cracks.

These observations were common to all of the sintered pellets. Figure 3b shows the surface of one of the sintered pellets after centerless grinding. The surface cracking appeared to penetrate into the bulk of the pellet. There also appeared to be regions in the pellet with a lighter colour. The measured sintered pellet geometric densities were between 10.0-10.3 g/cm^3 and the measured immersion densities were between 10.4-10.6 g/cm^3. The discrepancies between the measured geometric and immersion densities were likely due to absorption of water in the pellet surface cracks leading to artificially high immersed masses [4]. As such, the geometric densities better represented the true pellet densities. These densities were all below the pellet specification [**3**]. In an attempt to increase the sintered pellet density, an additional fabrication trial was completed in which the sintering temperature was raised to 1700°C. This did not yield a significant change in sintered pellet density and the pellet surface characteristics were the same. Therefore, different fabrication routes were pursued.

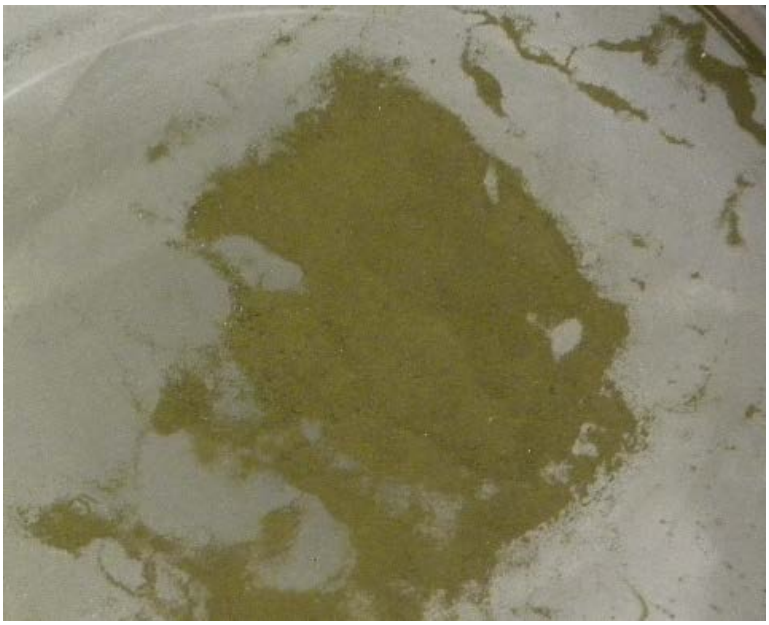

Figure 2 As-Received Powder

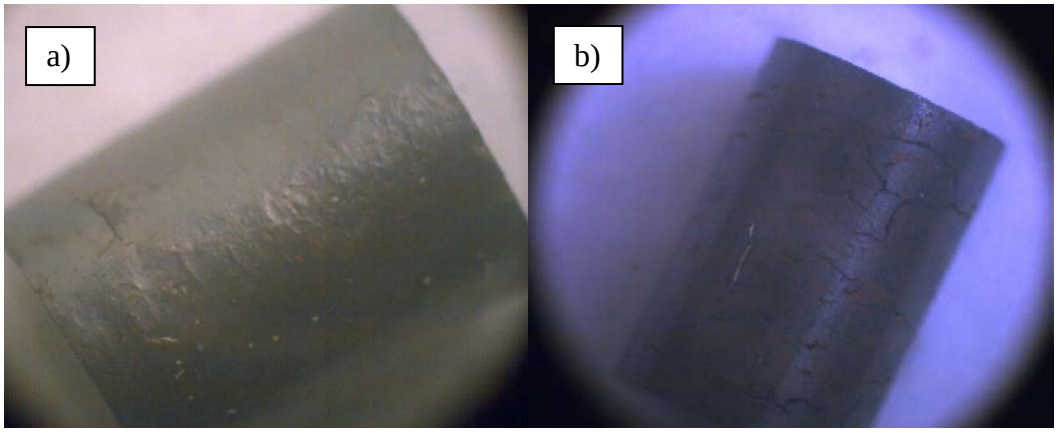

Figure 3 Picture of Sintered Pellet Surface a) before grinding b) after grinding

In the next fabrication trial, as-received powder was milled in a vibratory mill for 20 minutes and 40 minutes. The milled powders were lubricated with zinc stearate and cold pressed. The green pellets resulting from the powder milled for 20 minutes had green densities between 5.9-6.6 g/cm^3. The green pellets resulting from the powder milled for 40 minutes had green densities between 6.7-6.9 g/cm^3. All the green pellets were sintered together in hydrogen at a temperature of 1700°C for 2 hours. All of the sintered pellets had randomly oriented surface cracking upon removal from the sintering furnace similar to the previous tests. The measured geometric densities for the pellets made from powder vibratory milled for 20 minutes were 10.32 and 10.46 g/cm^3. The measured geometric densities for the pellets made from powder vibratory milled for 40 minutes were 9.73 and 9.82 g/cm^3. Again, these pellet densities were low. Low magnification, unetched microstructure pictures of two of the pellets are presented in Figure 1Figure 4. Both pellets had porous microstructures correlated to their low densities. The pellet produced from powder that had been milled for 40 minutes had greater porosity than the pellet that was produced from powder milled for 20 minutes. The low densities were due to oxidation of the powder during milling. The vibratory milling process generated heat due to the high intensity vibrations employed and the milling chamber contained oxygen from air. The mixture of UO_2 and higher oxide powder that resulted from the milling process produced pellets with low densities and high porosity as has been previously reported in literature [5]. Greater powder milling times resulted in a greater extent of powder oxidation and lower density, higher porosity sintered pellets. A small portion of the oxidized powder milled for 40 minutes was reduced and passivated using a thermogravimetric analyzer. This portion of powder was then pressed into a green pellet and sintered in hydrogen at 1650°C for 2 hours. The measured density for the pellet was 10.76 g/cm^3. This result suggested that vibratory milled powder could be used to produce high density pellets with an additional powder reduction and passivation step, however, this powder processing route was deemed to be too lengthy and complex for production purposes.

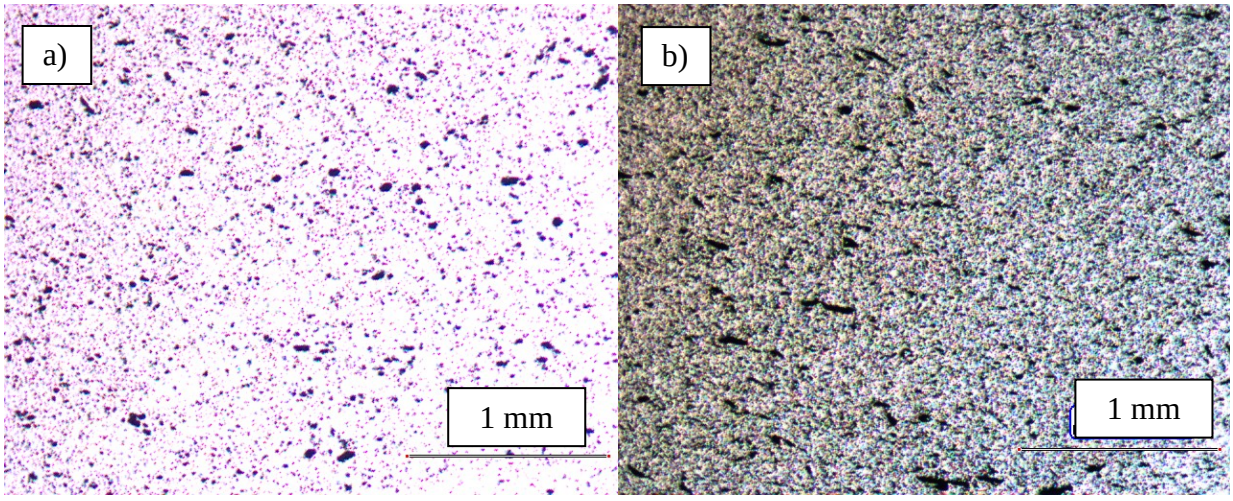

Figure 4 Unetched Micrographs of Pellets Produced from Powder Milled for a) 20 minutes b) 40 minutes

Another fabrication trial was conducted in which as-received powder was pre-pressed to form a compact with a density of 3.8 g/cm^3. This compact was then granulated by passing it through a sieve. The resulting granules were lubricated with zinc stearate and pellets were pressed with green densities between 5.3-5.6 g/cm^3. The green pellets were sintered in hydrogen at 1650°C for 2 hours. The measured sintered pellet densities were again low: 10.1-10.3 g/cm^3. A picture of a sectioned, mounted and polished pellet is provided in Figure 5. The picture shows light colour regions dispersed throughout the pellet. The nature of the lighter coloured regions was not investigated, however, they are believed to be low density regions. Similar light coloured regions were observed on the surface of a ground pellet in Figure 2b which had a similar density. This fabrication trial was unsuccessful in producing acceptable sintered pellets.

From the various preliminary fabrication trials that were conducted, it was evident that the as-received powders were not suitable to be used as-is for the production of high density sintered pellets without powder rework. Further powder rework options were investigated and a rework procedure was developed that produced a powder suitable for fabricating acceptable sintered pellets. The remainder of this paper will deal with the fabrication of UO_2 pellets to be used in the ICENS SLOWPOKE-2 LEU core conversion.

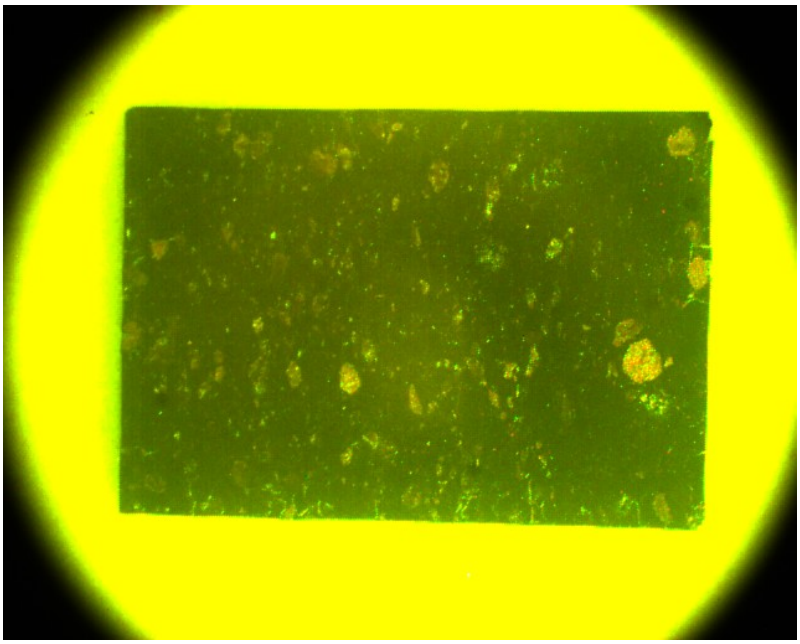

Figure 5 Sectioned, Mounted and Polished Pellet with Light Coloured Regions

4. Fabrication of UO_2 Pellets for ICENS SLOWPOKE-2 Core Conversion

Fabrication of the UO_2 fuel pellets followed the flow sheet in Figure 6. After the powder was reworked, the product of the rework procedure was passed through a sieve to form granules. Zinc stearate lubricant was added to the granules in the amount of 0.2 wt.% and mixed in a double cone blender. The lubricated granules served as the feed for final pressing. Green pellets

were pressed between 5.8-6.2 g/cm^3 using a hydraulically-compensated mechanical press and a custom made punch and die set. The green pellets were sintered in a hydrogen atmosphere at ~1650°C for four hours. A picture of as-sintered pellets from the first production batch of fuel pellets, with inlays illustrating improved pellet surface finish and microstructure, is shown in Figure 7. After sintering, the pellets were ground to final diameter using a centerless grinder. The pellets were then washed, dried, and inspected to ensure that they met the fuel specifications.

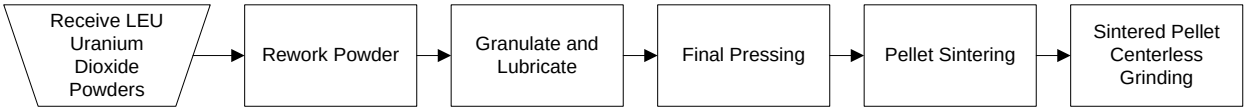

Figure 6 Pellet Fabrication Flow Sheet

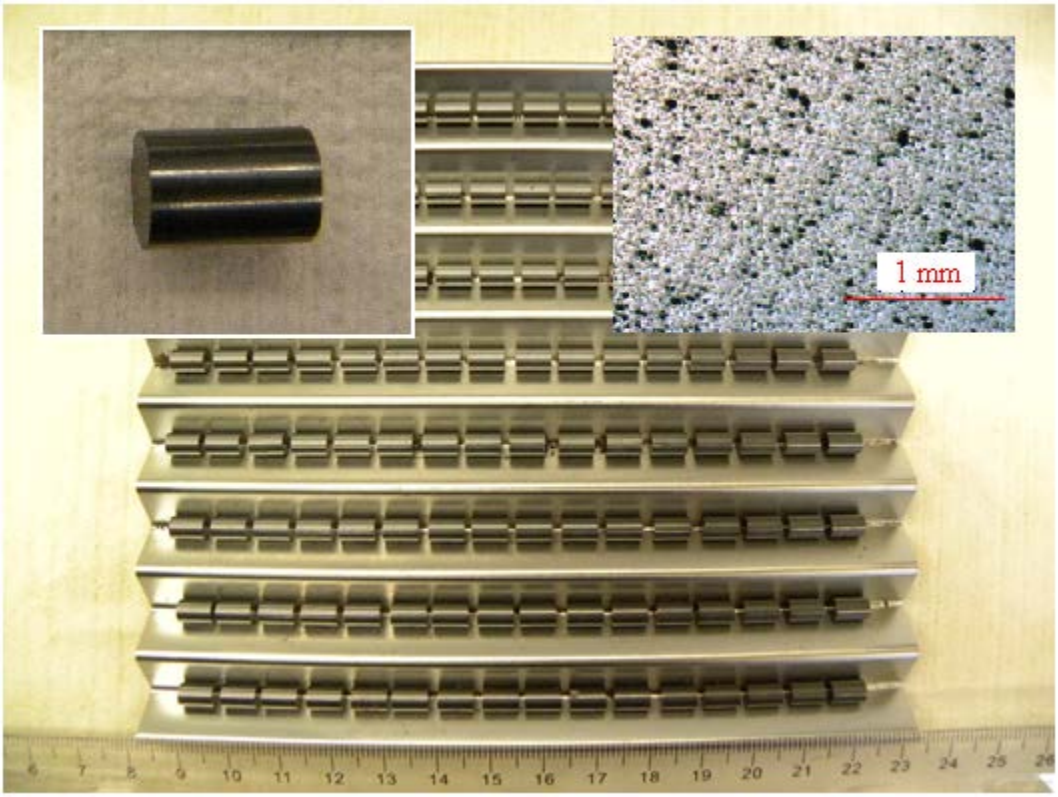

Figure 7 As-sintered Pellets from Production Batch #1

All pellets were visually inspected and pellets with non-conforming surface defects such as pits, cracks, and chips were removed and quarantined. A sample of the pellets that passed visual inspection was taken to inspect for pellet density, diameter, microstructure, end squareness, and surface roughness. The sample size for the pellet density and diameter inspections was determined using a sampling plan described in the Military Standard: Sampling Procedures and Tables for Inspection by Variables for Percent Defective [6]. For the remaining inspections, one pellet per batch was measured. Example inspection data from the first batch of production pellets are as follows. The measured pellet densities were 10.61±0.06 g/cm^3. The measured pellet diameters were 4.175±0.003 mm. For both characteristics, it was calculated statistically

using procedures outlined in [6] that the pellets met the acceptability criteria defined in [6] and in the technical specification [**3**]. The grain size, end squareness, and surface roughness were measured to be 7 μm, 0.006 mm, and 0.33 μm Ra, respectively. These measurements were all within specification as well.

5. Conclusion

ICENS at the University of the West Indies in Jamaica operates a SLOWPOKE-2 research reactor that is currently fuelled with HEU. AECL has been subcontracted to fabricate the LEU UO_2 pellets for its conversion to LEU fuel. Preliminary fabrication trials determined that the supplied UO_2 powder could not be used as-is to produce pellets that met the fuel specification. A powder rework procedure was developed at AECL so that acceptable finished pellets could be fabricated. Production pellet inspection data demonstrates that the fabrication process applied after powder rework produces pellets that meet the fuel specification.

6. References

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