



Fabrication and characterization of (Th, Pu)O₂ fuel at Canadian Nuclear Laboratories

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HIGHLIGHTS

- Canadian Nuclear Laboratories (AECL) has been carrying out research on thorium-based fuels for more than 55 years.
- CNL has undertaken efforts to further characterize as-fabricated and irradiated (Th, Pu)O₂ fuel.
- CNL has conducted fabrication trials on (Th, Pu)O₂ fuel to better control and improve pellet microstructure.
- Autoradiography techniques have been developed and applied to irradiated fuels to better characterize fuel microstructural.

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ABSTRACT

Thorium and other advanced fuels are of interest to the nuclear industry because of their potential role in ensuring long-term sustainable nuclear energy. Thoria (ThO₂)-based fuel cycles are options for both conventional water-cooled reactors (including light water and heavy water reactors) as well as various advanced reactor concepts. Plutonium-bearing thorium-based fuels are an attractive option for consuming stockpiles of Pu, while breeding fissile U-233 and extracting energy from thorium. Canadian Nuclear Laboratories (CNL; formerly Atomic Energy of Canada Limited, AECL) has been carrying out research and developing thorium-based fuels, including (Th, Pu)O₂, for more than 55 years. The CNL irradiation test of (Th, Pu)O₂ fuel assemblies (BDL-422) in the National Research Universal (NRU) research reactor at the Chalk River Laboratories culminated in the publication of post-irradiation examination results during 2008–2013. More recently, CNL has undertaken efforts to further characterize as-fabricated and irradiated (Th, Pu)O₂ fuel by developing and implementing improved ceramographic techniques and conducting fabrication trials to better control and improve pellet microstructure. In addition, autoradiography techniques have been developed and applied to irradiated (Th, Pu)O₂ fuel to better characterize fuel microstructural behaviour in combination with scanning electron microscope (SEM) characterization. This work has resulted in improvements in the microstructural characterization and fabrication of (Th, Pu)O₂ fuels.

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1. Introduction

Thorium and other advanced fuels are of interest to the nuclear industry because of their potential role in ensuring long-term

sustainable nuclear energy. Thoria (ThO₂)-based fuel cycles are options for both conventional water-cooled reactors (including light water and heavy water reactors) as well as various advanced reactor concepts. Plutonium-bearing thorium-based fuels are an attractive option for consuming stockpiles of Pu, while breeding fissile U-233 and extracting energy from thorium.

The fabrication of fuel pellets having homogeneous microstructures is key to irradiation performance. Depending on the fabrication method, thorium-based fuels may exhibit heterogeneous microstructures characterized by regions of residual granules. Areas of fine granules (particles or grains) originate when the starting fuel powder is pre-pressed and granulated; these granules may remain during pellet pressing and/or sintering, resulting in

Abbreviations: AECL, Atomic Energy of Canada Limited; BSE, Back-Scattered Electron; CNL, Canadian Nuclear Laboratories; EDX, Energy Dispersive X-ray; FGR, Fission-Gas Release; MOX, Mixed Oxide Fuel; NRU, National Research Universal; PIE, Post-Irradiation Examination; POP, Powder Oxide Pellet; Pu, Plutonium; RFFL, Recycle Fuel Fabrication Laboratories; SEM, Scanning Electron Microscopy; Th, Thorium; U, Uranium; WDX, Wavelength Dispersive X-ray.

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residual granules¹ in the as-fabricated microstructure [1,2]. Fuel presenting residual granules experiences degraded thermal performance compared to homogeneous fuel [1].² In addition, a roughly bi-modal grain size variance is associated with areas of high plutonium concentration within the fuel matrix that cause abnormal grain growth during sintering; some grains grow rapidly in a matrix of finer grains, resulting in two distinct regions of different grain sizes [3].³ Conventional MOX fuels having residual granules in the as-fabricated microstructure exhibit slightly higher fission-gas release (FGR) than those with a non-granular microstructure [4].⁴ Thus, the ability to characterize unirradiated (as-fabricated) and irradiated thorium-based fuel microstructures and to fabricate thorium-based fuel pellets having homogeneous microstructures are important technologies required to achieve the potential benefits of thorium fuel cycles. This can be particularly challenging for (Th, Pu)O₂ fuels.

Canadian Nuclear Laboratories (CNL; formerly Atomic Energy of Canada Limited, AECL) has been carrying out research on and developing thorium-based fuels, including (Th, Pu)O₂, during a time spanning more than 55 years [1,5,6]. The most recently completed CNL irradiation test of (Th, Pu)O₂ fuel assemblies in Chalk River's NRU reactor (BDL-422) culminated in the publication of post-irradiation examination (PIE) results during 2008–2013 [1,7–9]. This previous work is summarized in this paper to provide context to more recent work on (Th, Pu)O₂ fuel, including measurements on BDL-422 fuels. The objective of the BDL-422 experiment was to study the fuel performance of (Th, Pu)O₂ fuel operating to high burnup [8]. The original BDL-422 fuel PIE included limited characterizations of pellet microstructure due to challenges encountered in performing ceramography – ceramographic preparation of (Th, Pu)O₂ differs significantly from that of ThO₂ and (Th, U)O₂, for which CNL has extensive experience.

Most recently, CNL has undertaken a review of ceramography techniques used in the examination of various thorium-based fuels with the objective of making improvements. This work, described in this paper, has resulted in improvements to the optical microscopy of (Th, Pu)O₂ fuels, including unirradiated and irradiated samples from the BDL-422 test.

Autoradiography is a valuable analytical tool, particularly in the characterization of fuels that are fabricated with heterogeneous distributions of Pu (i.e., distinct Pu particles or Pu-rich areas). High alpha emissions from Pu facilitate characterization of Pu distribution, both before and after irradiation. Although alpha autoradiography has been used by CNL in the characterization of conventional MOX (U, Pu)O₂ fuels [10–13], it was not used in the original characterization of irradiated BDL-422 fuel [1,7–9]. More recently, attempts to conduct alpha autoradiography on BDL-422 fuel produced results that were difficult to interpret. Preliminary results that were reported in 2016 for unirradiated BDL-422 fuel incorrectly identified light areas in the radiographs as Pu-rich areas [6]. Since that time, work has been carried out to further develop alpha autoradiography for (Th, Pu)O₂ fuel and to better characterize Pu distribution within the fuel, before and after irradiation. This

work is described in this paper.

BDL-422 fuel was heterogeneous in two aspects. Firstly, the ThO₂ and PuO₂ were blended using a low-intensity method that resulted in distinct Pu particles being uniformly distributed throughout the pellets. Secondly, the pellet fabrication method resulted in a granular microstructure characterized predominantly by very small grains (<5 µm) [2,6]. Although the BDL-422 fuel performed very well at high power to extended burnups, it was recognized that a more homogeneous microstructure would yield benefits in terms of fuel performance, especially at burnups >40 MWd/kgHE [6]. With this in mind, fabrication trials were recently conducted at CNL to produce (Th, Pu)O₂ fuels with superior microstructures to those of BDL-422 fuels. These fuels were not irradiated, but were characterized using techniques that had also been applied to BDL-422 fuel (ceramography, autoradiography and SEM characterization). This work is also discussed in this paper.

2. Bdl-422 fuel fabrication

The BDL-422 experiment, as part of AECL/CNL's recent effort to develop advanced fuels and fuel cycles utilizing thorium [8], involved the fabrication of six 37-element test fuel bundles fuelled with (Th, Pu)O₂ pellets in Zircaloy-4 sheathing (cladding). After the six bundles were irradiated in the National Research Universal (NRU) reactor, PIE was conducted on five bundles in the Chalk River hot cells (Table 1).

The six bundles were fabricated in AECL/CNL's Recycle Fuel Fabrication Laboratories (RFFL) in 1983 [6]. The pellets in the BDL-422 bundles contained 1.53 wt. % Pu in (Th, Pu)O₂ [8]. The plutonium concentration in the fuel was macroscopically uniform and the average fuel pellet density was 9.469 g/cm³ (95% of theoretical density). The thorium powder was blended with PuO₂ powder that was sieved to ensure the PuO₂ particle size was less than 150 µm. The pellets were fabricated in a process incorporating a thorium (ThO₂) powder that was wet-milled, dried and granulated to improve the sintered density of the fuel. This process resulted in heterogeneous microstructures having residual granules and small grains (3–4 µm) relative to typical UO₂ grains (5–10 µm) [6,7]. A plenum was inserted in one end of each fuel element to accommodate fission gas released during irradiation.

BDL-422 bundles had a similar geometry to bundles used in commercial PHWRs. Each bundle included three concentric element rings: an outer ring containing eighteen elements, an intermediate ring containing twelve elements, and an inner ring containing six elements. The centre element was removed to facilitate testing in the NRU loops. Elements are identified in this paper by the bundle it originated from, followed by a suffix representing the element. For example, the identity ADA-2 refers to element 2 from bundle ADA [8].

3. Bdl-422 irradiation

Six BDL-422 bundles were successfully irradiated in the U1 and U2 loops of the NRU reactor during the period of 1984–2000 to outer element burnups ranging from 19 MWd/kgHE to 45 MWd/kgHE (see Fig. 1). Table 1 lists the outer-element burnup and maximum sustained linear power for each bundle.

4. Bdl-422 fission-gas release

BDL-422 post-irradiation examination (PIE) was conducted on three bundles irradiated to burnups of 19–33 MWd/kgHE (ADC, ADE, ADF) and two bundles irradiated to burnups of >40 MWd/kgHE (ADA, ADD) [6,9]. The FGR results were of particular interest. FGR is a key fuel performance parameter, particularly at extended

¹ Compact particles resulting from pre-pressing and granulation that did not homogenize during pellet pressing and/or sintering. Examples of residual granules in thorium-based fuel are shown in Fig. 2.

² Thorium-based fuels with granular microstructures experience degraded thermal performance and exhibit similar FGR characteristics to conventional UO₂ fuels, while thorium-based fuels with homogeneous pellet microstructures exhibit superior performance to UO₂ fuels.

³ An example of a thorium-based fuel with bi-modal grain size is shown in Fig. 13.

⁴ The areas of high Pu concentration within the finer fuel matrix behave like fuel irradiated at higher powers, with increased grain restructuring and higher fission-gas release.

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