PROGRESS IN THE DEVELOPMENT OF THORIA FUEL SCIENCE & TECHNOLOGY

M.R. Floyd[1], R. Beier[1], J.S. Bell[2], A. Bergeron[1], E. Bulemela[1], G. Cota-Sanchez[1], R.S. Dickson[1], K. Leeder[1], S.J. Livingstone[1], A. Quastel[1] and M. Saoudi[1]

[1]Canadian Nuclear Laboratories, Chalk River, Ontario, Canada
(1-800-377-5995 ext. 43899, mark.floyd@cnl.ca)
[2]Royal Military College of Canada, Kingston, Ontario, Canada

ABSTRACT

This paper describes recent efforts at Canadian Nuclear Laboratories (formerly AECL) to address identified thoria fuel science and technology gaps that are of interest to the CANDU community, including the development of collaborations. Recent advances in pellet fabrication, ceramography, materials properties characterization, performance modelling, safety-related testing, defect behaviour and reprocessing are presented.

Introduction

Canadian Nuclear Laboratories (CNL; formerly AECL) has more than 55 years of experience in developing thoria-based fuel that supports pressurized heavy water reactor (PHWR) fuel technology [1, 2]. This includes several CANDU-type irradiation tests having 19, 28 and 37-element geometries and ThO_2 (thoria), $(Th, U)O_2$ and $(Th, Pu)O_2$ pellets [2-5].

In recent years, CNL's Thoria Roadmap Project has conducted a review of science and technology (S&T) requirements to support water-cooled reactor designs incorporating cylindrically-clad ceramic fuel pellets [1]. This review spanned eleven different S&T areas including fuel fabrication, irradiation testing and post-irradiation examination (PIE), materials properties, fuel modelling, fuel safety (including defected fuel behaviour), reactor physics, radiation protection and dosimetry, waste management, reprocessing, thorium supply (resources) and non-proliferation (safeguards). One of the primary outcomes of the project was the identification of knowledge "gaps" in each of the aforementioned S&T areas, and the development of a roadmap (plan) to address them. Collaboration with domestic and international organizations is integral to the on-going development and execution of the roadmap.

This paper describes recent CNL progress in addressing identified thoria fuel S&T gaps that are of interest to the CANDU community, including the development of collaborations. Recent advances in pellet fabrication, ceramography, materials properties characterization, performance modelling, safety-related testing, defect behaviour and reprocessing are presented.

1. Pellet Fabrication

CNL has significant experience in fabricating thoria-based fuels [6].The ability to produce ThO_2 and (Th, U)O_2fuel pellets having a high-quality, homogeneous microstructure and superior performance to UO_2at burnups of 360-930 MWh/kgHE(15-39 MWd/kgHE) has been demonstrated in the National Research Universal (NRU)reactor under the DME-221 irradiation test [3]. CNL intends to extend the burnup of DME-221 fuel to > 1000 MWh/kgHE (42 MWd/kgHE) prior to the planned closure of NRU in 2018.

Fabrication of (Th, Pu)O_2fuel for the BDL-422 irradiation test [4, 5] was performed in CNL's Recycle Fuel Fabrication Laboratories (RFFL) > 30 years ago (1983). This fuel contained 1.53 wt.% Pu in (Th, Pu)O_2 . The BDL-422 pellet fabrication process utilized a thoria powder that was wet-milled, dried and granulated. This process has been shown to result in granular (inhomogeneous) microstructures in (Th, U)O_2 fuels that exhibit higher fission-gas release (FGR) than pellets having homogeneous microstructures [2, 6].

At the time of fabrication of BDL-422 fuel, the pellets were not identified as having a granular microstructure. BDL-422 fuel had an initial (as-fabricated) average grain size of 3-4 μm (5-10 μm is considered normal for UO_2 fuel). During 2015, archived (unirradiated) BDL-422 pellets were subjected to detailed microstructural characterization. This investigation confirmed that the as-fabricated BDL-422pellets had a granular microstructure related to the fabrication process (Figure 1). The archived pellets were also subject to alpha-autoradiography to investigate Pu homogeneity; this identified Pu-rich areas having diameters of 2-158 μm (Figure 2). Previous investigations have shown that (U, Pu)O_2 fuels having similar Pu-rich areas exhibit higher FGR than those having more homogeneous Pu distributions [7].

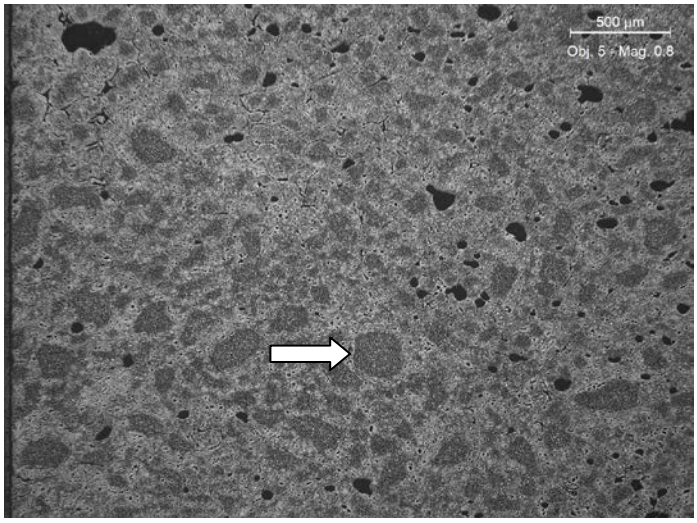

Figure 1 Ceramographic Image of Archived (Unirradiated)BDL-422 (Th, Pu)O_2 Pellet. Dark Areas (arrow) are Residual Granules that have not Homogenized during Final Pressing/Sintering.

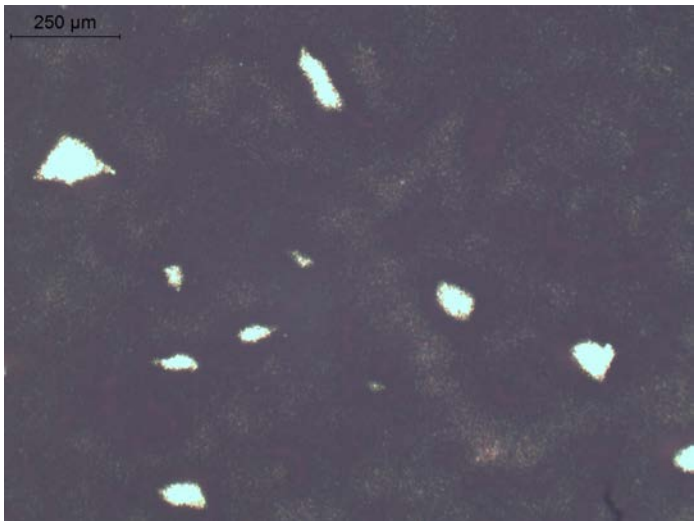

Figure 2 Alpha Autoradiograph of Archived Unirradiated BDL-422 (Th, Pu)O_2 Pellet. Light Areas are Pu-Rich Areas.

Six BDL-422 fuel bundles were irradiated in the NRU reactor during1984-2000. Notwithstanding its sub-optimal microstructure (granular microstructure, small average grain size and inhomogeneous Pu distribution), BDL-422fuel exhibited superior performance to UO_2 for burnups up to ~ 900 MWh/kgHE (37 MWd/kgHE), exhibiting FGR of only 1-5%. FGR was observed to increase to 20-33% at burnups > 1000 MWh/kgHE (42 MWd/kgHE) [8]. Recent modelling efforts identified that this FGR increase may be related to a power increase (< 10 kW/m) that occurred at 800-900 MWh/kgHE (33-37 MWd/kgHE); however, the sub-optimal pellet microstructure may also have also been a factor. It was concluded that further irradiation tests are required to determine the influence of pellet microstructure on the performance of (Th, Pu)O_2 fuel at high burnup.

During 2014, (Th, Pu)O_2 pellet fabrication trials were conducted in the RFFL; these were envisioned as prerequisite to fabricating fuel for a successor irradiation test to BDL-422. The fabrication trials sought to eliminate residual granules in the microstructure, in addition to producing pellets with various grain sizes ($\geq$ 5 µm average) and various degrees of Pu homogeneity (including PuO_2witha homogenous distribution within ThO_2).The experimental results showed that the final microstructure depends on both the blending method and sintering time. Fuel pellets pressedfrom ThO_2 and PuO_2 that were vibratory co-milled and sintered for8 hours exhibited a homogenous microstructure having an average grain size of 9 µm, and a density of 9.76 g/cm^3 (97.12 %TD; Figure 3). Fuel pellets fabricated using a 30% PuO_2 master-mix that was blended with ThO_2 using a low-intensity mixer (sintered for 12-16 hours), exhibited a slightly granular microstructure, characterized by a bimodal grain size distribution (average size of 4 µm) and a density of 9.78 g/cm^3 (97.27 %TD). Fuel pellets fabricated using a low-intensity blending method and sintering times of 4 hours exhibited distinct Pu particles, granular microstructure, very small grains anda density of 9.48 g/cm^3 (94.37 %TD).

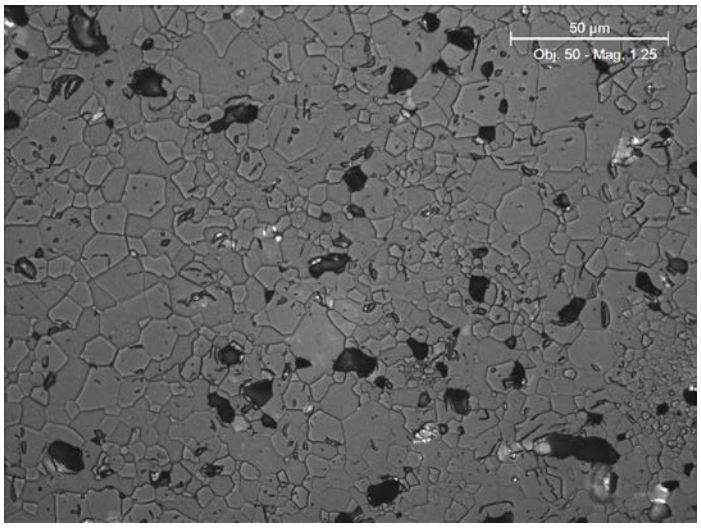

Figure 3 Ceramographic Image of a (Th, Pu)O_2 Pellet Produced UsingVibratory Co-Milled Blending (Note Homogeneous Pu Distribution and Absence of Bimodal Grain Size Distribution).

2. Ceramography

Prior to the post-irradiation examination(PIE) of BDL-422 fuel [4,5], CNL had limited experience with conducting ceramographic examination of (Th, Pu)O_2 fuel. During the BDL-422 PIE campaign, several challenges were encountered in producing high quality ceramographs that facilitated characterization of the irradiated (Th, Pu)O_2 pellets. It was found that ceramography techniques that were successfully used in the past for irradiated ThO_2 and (Th, U)O_2 fuels were sub-optimal for the BDL-422 (Th, Pu)O_2 fuel. It was concluded that developmental work was required to optimize thoria ceramographic techniques, particularly on irradiated (Th, Pu)O_2 pellets.

During 2014, a review of thoria ceramography techniques used by CNL facilities was conducted. These facilities included the RFFL (fabrication of (Th, Pu)O_2 pellets) and the Chalk River Advanced Fuel Technology Ceramics (CRAFT-C) laboratories (fabrication of ThO_2 and (Th, U)O_2 pellets). In addition, techniques used by the Fuels and Materials Hot Cells (FMC) facility were included in the review (ceramography of irradiated ThO_2, (Th, U)O_2 and (Th, Pu)O_2pellets). This work included experimentation on unirradiated (Th, Pu)O_2 pellets (fabricated in the RFFL) using various etching, grinding and polishing techniques, representative of those used in the RFFL, FMC and CRAFT-C. Improvements to ceramographic preparation techniques for unirradiated (Th, Pu)O_2 were identified; in addition, studies were recommended on irradiated (Th, Pu)O_2 to better understand the effect of irradiation on microstructural characterization.

During 2015, irradiated fuel from the BDL-422 (Th, Pu)O_2 test was selected for additional microstructural characterization in the FMC. These fuels covered a wide range of fuel powers and burnups (Table 1). Fuel samples were etched using a heated dilution of ammonium fluoride, using etching time as a variable. It was found that optimal etching times were dependent on the fuel irradiation history (Figure 4); longer etching times were required with increasing fuel power and/or burnup.

Table 1 Irradiated BDL-422 (Th, Pu)O_2 Fuel Data Relevant to Ceramography Study

Fuel Bundle Identity	Element Number (Bundle Ring)	Burnup – MWh/kgHE (MWd/kgHE)	Maximum Sustained Power – kW/m
ADA	2 (Outer)	1181 (49)	54
ADA	19 (Intermediate)	695 (29)	39
ADC	8 (Outer)	451 (19)	67

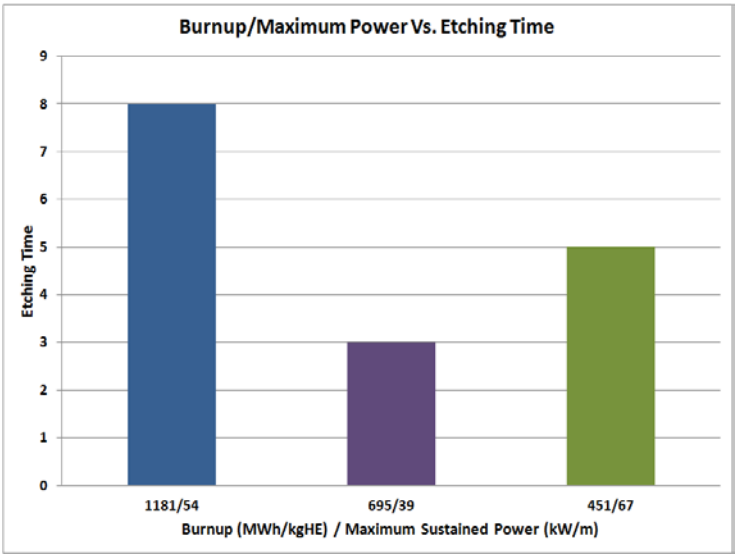

Figure 4 Optimized Etching Times for Irradiated BDL-422 (Th, Pu)O_2 Illustrating Dependence on Burnup and Power

3. Materials Properties Characterization

During 2013, CNL commissioned laser flash equipment for the purpose of thermal diffusivity measurements on unirradiated fuel. High density (Th, U)O_2 pellets were fabricated at CNL for the purpose of thermal diffusivity measurements having 0,1.5, 3, 8, 13, 30, 60 and 100 wt.% UO_2 (in total ThO_2 + UO_2). High density (Th,U)O_2pellets were also fabricated at the Joint Research Centre - Institute for Transuranium Elements (JRC-ITU; Karlsruhe, Germany) having 79 and 93 % wt.% UO_2.Laser flash thermal diffusivity measurements were conductedon the pellets at CNL and JRC-ITU [9].Thermal conductivity was calculated using pellet density data and specific heat data available in open literature.ThO_2 thermal conductivity is significantly higher than that for UO_2;however, (Th,U)O_2 thermal conductivity decreases rapidly with increasing UO_2 content, such that atconcentrations$\geq$ 60 %, it is close to that of pure UO_2 (Figure 5). Unlike UO_2, ThO_2 is a semi-transparent material; as a result, heat transfer is both conductive and radiative in nature. This effect is reduced during laser flash measurements by coating the front and rear surfaces of samples with carbon to facilitate the absorption of the laser energy on the sample surface of the front side and to enhance infrared emissivity at the rear surface. Thermal conductivity degradation is attributed to phonon scattering enhancement caused by lattice strain resulting from the introduction of uranium to the ThO_2 lattice. This

decrease may also be due to the semi-transparent nature of ThO_2;the contribution of radiative heat transfer decreases progressively with the addition of UO_2 such that it is lost at UO_2 concentrations $\geq$ 30 % .

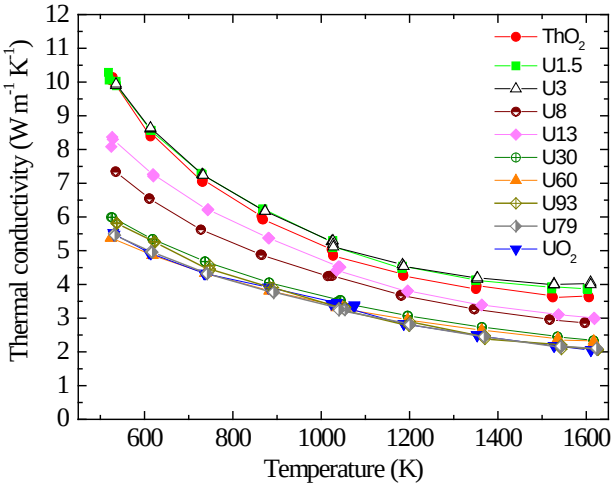

Figure 5 Variation of Unirradiated (Th, U)O_2 Thermal Conductivity with Temperature and UO_2 Content (0, 1.5 , 3, 8, 13, 30, 60, 79, 93 & 100 wt.%)

4. Performance Modelling

A major gap identified by CNL's Thoria Roadmap Project is the paucity of fuel performance models for thoria-based ceramic fuel, particularly for PHWR fuel [1]. To address this gap, the Royal Military College of Canada (RMCC) and CNL have collaborated to simulate ThO_2, (Th, U)O_2 and (Th, Pu)O_2 fuel behaviour using an existing COMSOL-based UO_2 model as a starting point (FAST; Fuel and Sheath Modelling Tool) [10]. This work implemented changes to the FAST UO_2 fuel model to input parameters specific to thoria-based fuel thermal-mechanical properties. The result of this work is the Multi-Purpose Material Fuel and Sheath Modelling Tool (MPM-FAST) [11,12]. During 2013/14, MPM-FAST was benchmarked against data from the DME-221 (ThO_2 and (Th, U)O_2) and BDL-422 (Th Pu)O_2irradiation tests [2-5,8].Modelling FGR forthoria-based fuel presents unique challenges because itsmaterials properties differ significantly from that of UO_2.Further work is required to better understand the mechanisms of fission product release in thoria-based fuels (in contrast to that of UO_2). The agreement between calculated and measured FGR is a good indicator of the performance of the model in calculating parameters such as fuel temperature and fission-product diffusion. Table 2 illustrates that MPM-FAST predicts realistic FGR values for DME-221 ThO_2 and (Th, U)O_2 fuels having burnups of 360-930 MWh/kgHE (15-39 MWd/kgHE). The calculated and measured FGR are also in reasonable agreement for BDL-422 (Th, Pu)O_2 fuel irradiated to burnups<900 MWh/kgHE (38 MWd/kgHE). Both the calculated and measured FGR values experience a large increase at burnups > 1000 MWh/kgHE (42 MWd/kgHE); however, the MPM-FAST calculated values are significantly less than those measured (14-16% vs. 20-33%). As discussed earlier in the paper, the observed FGR increase above 1000 MWh/kgHE is attributed to a power increase that occurred at 800-900 MWh/kgHE (33-37 MWd/kgHE), as well as the sub-optimal pellet microstructure (residual

granules, small initial grain size and inhomogeneous Pu distribution). Work is ongoing to improve MPM-FAST FGR calculations for BDL-422 fuel irradiated to >1000 MWh/kgHE.

Table 2 Comparison of Fission Gas Release – Calculated vs. Measured [3-5, 8, 11, 12]

Fuel Type (DME-221 & BDL-422)	**Discharge Burnup - MWh/kgHE (MWd/kgHE)**	**Maximum Sustained Power - kW/m**	**Calculated % FGR (MPM-FAST)**	**Measured % FGR**
ThO_2	361 (15)	41	< 0.1	0.05 - 0.1
ThO_2	594 (25)	41	0.2	0.05 - 0.1
(Th, U)O_2 – 1.0% U-235	499 (21)	46	< 0.1	0.06 - 1.2
(Th, U)O_2 – 1.5% U-235	594 (25)	55	0.2	0.08 - 2.8
(Th, U)O_2 – 1.5% U-235	929 (39)	55	3.3	0.08 - 2.8
(Th, Pu)O_2 – 1.53% Pu	451 (19)	67	3.5	5.3
(Th, Pu)O_2 – 1.53% Pu	597 (25)	64	2.9	1.2
(Th, Pu)O_2 – 1.53% Pu	856 (36)	52	5.6	2.8
(Th, Pu)O_2 – 1.53% Pu	1082 (45)	73	**14**	**20-27**
(Th, Pu)O_2 – 1.53% Pu	1181 (49)	54	**16**	**26-33**

5. Safety-Related Testing

The behaviour of thoria-based fuel pellets exposed to water and/or steam as a result of sheath failure is not completely understood. Thoria-based fuels may include U/Pu contents of 1-5% in currently operatingwater-cooled reactors and 10-15%in Generation IV concepts such as a super-critical water reactor. The oxygen potential ofnuclear fuel oxides is an important thermodynamic parameter. Oxidation of thoria-based fuels (leading to a change in oxygenpotential) can lead to increasedfuel restructuring, degradation of thermalconductivity, and increased fuel erosion. CNL has initiated work to better understand the behaviour of thoria-based fuels that experience sheath failure under normal operating and accident conditions. This includes understanding the effect of U/Pu addition to the ThO_2 matrix.

Work is in progress at CNL to better understand the diffusion of oxygen in (Th, U/Pu)O_2. This includes measurements (and modelling) of oxygen diffusion with varied concentrations of U/Pu at various temperatures. Oxygen diffusion in UO_2 has been measured in the past using several techniques, including electrical conductivity and gravimetric methods [13]. Surface profiling using ^{18}O has also been used to determine the oxygen concentration [13, 14]. Recent CNL oxygen chemical diffusion measurements have been conducted on unirradiated UO_2 samples at 650°C and 700 °C using thermal gravimetric analysisand CO_2/CO and H_2/Ar cover gases (reduction of the sample is oxygen diffusion controlled). Since the chemical diffusion coefficient in thoria-based fuel is lower than in UO_2 fuels at elevated temperatures [15],this method is expected to be adequate to obtain the desired oxygen diffusion measurements. Finite element modelling of the experiments is being conducted in parallel to numerically compute the oxygen diffusion coefficients.

During 2016, CNL anticipates the completion of a hot cell test to simulate the behaviour of irradiated thoria fuel under accident conditions. This test will include the trace re-irradiation of samples of ThO_2 fuel from the DME-221 test in NRU (to re-establish short-lived fission products),

followed by the exposure of the fuel samples to various gaseous atmospheres at high temperature in a furnace (inside a hot cell), to study the release of fission products.

6. Defect Behaviour

Very little data exists for defected thoria fuels with quantified fission product release to the coolant. One such dataset is associated with the FDO-680 test, performed in the X-2 loop of the NRX reactor ~ 40 years ago [2]. The FDO-680 test included a single CANDU-type (Th, U)O_2 element (4 wt% UO_2 in ThO_2) containing a single drilled defect in its sheath. The test included the collection of detailed fission product concentration data from the X-2 loop coolant. Analysis of the fission product data immediately following the test was minimal; it was only concluded that the observed fission product release was significantly less than that observed from a similarly defected/operated UO_2 element (FDO-681 test).[1] In 2013, an analysis of Xe-133 data generated by the FDO-680 and FDO-681 tests was published and concluded that the release of this long-lived gaseous fission product radioisotope was 10-100 times lower than that observed in similarly operated UO_2 fuel [2]. It was also noted that the FDO-680 fuel pellets experienced less erosion at the defect site than expected for defected UO_2 fuel, due to the chemical inertness of ThO_2.

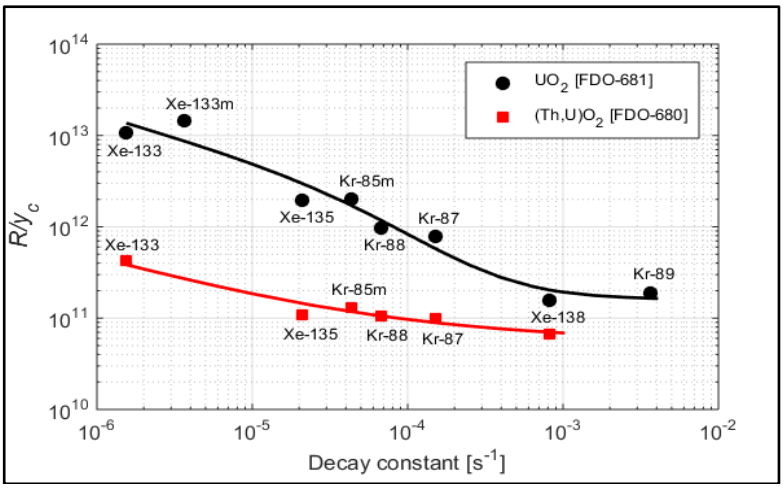

Figure 6 Yield-Corrected Noble Gas Release (R/y_c)as a Function of Decay Constant(λ)for (Th,U)O_2 (FDO-680) and UO_2 (FDO-681)

During 2014-2015, a more detailed analysis of coolant fission product data for the FDO-680 and FDO-681 experimentswas initiated. Figure 6 contrasts (Th, U)O_2 and UO_2 noble gas release to the coolant as a function of decay constant (λ; inversely proportional to half-life), where R [atom.s^{-1}] is the release from the defected element to the coolant, and y_c [atom.$fission^{-1}$] is the cumulative fission yield. R/y_c values were calculated from coolant activity measurements using the analytical tool developed by Livingstone [16], and the best fit lines follow the first order kinetic model developed by Lewis et al. [17]. The noble gas release from the (Th, U)O_2 fuelis approximately one order of magnitude less than that from the UO_2 (with the least difference for short-lived isotopes). Defected

[1]FDO-680 and FDO-681 are "sister" experiments that had identical drilled sheath defects (~ 1 mm^2 in area), having similar powers (~ 48 kW/m) and burnups (8-10 MWh/kgHE).

thoria-based fuels are expected to experience lower fission product release to the coolant (than UO_2 fuels) because the fuel matrix has superior fission product retention properties (higher thermal conductivity and melting point) and does not experience significant oxidation when exposed to the coolant.

7. Reprocessing

The thorium fuel reprocessing experiment (TFRE) was conducted at Whiteshell Laboratories (WL) during 1975-1985. Its mandate was to investigate a full range of technical and operating issues associated with the application of a modified THOREX process [18] for the separation and recovery of thorium, uranium and plutonium from thorium-based fuels. A review and summary of TFRE documentation was completed by CNL in 2015. TFRE investigations involved a multidisciplinary team of scientists, engineers, technologists, trades and operators. TFRE successfully demonstrated the overall viability of thorium fuel reprocessing, including significant progress in system design, material & component selection, fuel decladding & dissolution, underlying & supporting chemistry, process parameters, computer simulation and waste management. As a result of the 2015 review, key aspects of three reprocessing technologies (THOREX-based solvent extraction, fluoride volatility and pyroprocessing) were summarized and frameworks defined for further studies to be undertaken in the short, intermediate and long term. It was concluded that research in various S&T areas must be coordinated in order to develop thoria-based fuel cycles, including reprocessing, fuel fabrication, irradiation testing, PIE and waste management. This includes coordination of international collaboration on thoria fuel S&T.

CNL has conductedexperiments to investigate the dissolution of (Th, U)O_2 in trifluoromethanesulfonic ("triflic") acid solutions. Fragmentsof sintered (Th, U)O_2 and SIMFUEL[2]were dissolved in concentrated triflic acid solutions in order to evaluate reaction kinetics. Under reflux conditions,triflic acid solutions appear to dissolve sintered (Th,U)$O_2$and SIMFUEL more rapidly than nitric acid-hydrofluoric acid mixtures.During 2012, CNL conducted lab-scale experiments to investigate reprocessing thoria-based fuel using the fluoride volatility technique. Experiments were conducted on crushed, unirradiated (Th, U)O_2 pellets and on thoria SIMFUEL having a simulated burnup of 1440 MWh/kgHE (60 MWd/kgHE);98% of the elemental uranium was successfully separated from the thoria SIMFUEL [19]. More recently, the fluoride volatility technique was applied in a hot cell environment to crushed, irradiated DME-221 ThO_2 pellets having a burnup of 594 MWh/kgHE (25 MWd/kgHE). The results are currently being analyzed and will be reported at a later date.

8. Collaborations

CNL and the Royal Military College of Canada (RMCC, Kingston) havebenefited from collaborations on nuclear fuel initiatives for many years. Recent CNL/RMCC collaboration has been instrumental to the development of the MPM-FAST thoria fuel modelling tool, as described in this paper(and reference [12]).Under the framework of a Canada-Euratom agreement, CNL is collaborating with JRC-ITU to conduct (Th, U)O_2thermal property measurements (described in this paper).CNL and the University of Saskatchewan are

[2]SIMulated-irradiated FUEL with non-radioactive fission products.

collaborating to use synchrotron radiation techniques to investigate the oxidation behaviour of (Th, U)O_2 fuel with varying UO_2 contents.

Recently, CNL collaborated with the Organization for Economic Cooperation and Development Nuclear Energy Agency (NEA) and several other international contributors to conduct a review of thorium fuel cycles and their potential role in future nuclear energy production [20]. The report concluded that:

- Thorium fuel cycles complement uranium/plutonium fuel cycles and have the potential to improve the flexibility of nuclear energy in medium-term and its long-term sustainability.
- Options for the introduction of thorium into the nuclear fuel cycle include using it as a means to burn plutonium (and possibly other higher actinides).
- Thorium-based fuels have the potential to achieve higher conversion factors in thermal or epithermal neutron spectra, while recycling fissile material from spent fuels.
- Thoria-based fuel generally offers improved performance over current UO_2-based fuel designs because of its superior material properties.

Vanderbilt University (VU, Tennessee, USA) is conducting a study on thorium fuel cycle options under the US-DOE Nuclear Energy University Program. CNL is informally collaborating with VU to assist in the evaluation of thorium fuel cycle options, particularly those associated with PHWRs. VU provided valuble input to CNL's recent review of progress in thoria fuel S&T [1]. CNL is also working with VU to support meetings sponsored by the American Nuclear Society (ANS) and Electric Power Research Institute (EPRI) on thorium fuel S&T.

CNL reached an agreement in 2013 with the Nuclear Power Institute of China (NPIC) to collaborate on advanced reactor technology development. CNL is currently working with NPIC to develop scope for thoria fuel S&T collaboration in areas including fuel modelling, materials properties characterization and irradiation testing.In 2015, a joint proposal was approved betweenthe Bhabha Atomic Energy Research Instituteand CNL to apply various analytical techniques to characterize thoria-based fuels. These techniques include electron microscopy (SEM, TEM and EBSD), electron spectroscopy (EDS and WDS) and X-ray diffraction.

9. Conclusions

Nothwithstanding more than 55 years of thoria-based fuel technology development, CNL is continuing to identify and address thoria fuel S&T gaps. This work is being pursued in collaboration with both domestic and international partners and is key to improving the readiness of thoria fuel technology for nuclear power generation implementation. Recent advances under CNL's Thoria Roadmap Project include improvements in fuel pellet fabrication and ceramography. CNL's materials properties measurement capability has improved and contributed to a better understanding of the effect of UO_2 addition to ThO_2 on thermal conductivity. A model to simulate the performance of thoria-based fuels has been developed in collaboration with RMCC and benchmarked against CNL irradiation data. Safety-related tests are being conducted on unirradiated and irradiated thoria fuel to better understand its behaviour under normal operating and accident

conditions. Detailed analysis of fission-product release data has improved our understanding of defected thoria-based fuel behaviour (in contrast to that of UO_2). CNL has undertaken measures to preserve its expertise in thoria fuel reprocessing and to make advances in the development of reprocessing technologies.

10. Acknowledgements

Numerous CNL staff have contributed to progress in the development of thoria-based fuel S&T; the authors appreciate and acknowledge their contributions. Special thanks to Jeffrey Orman (formerly of CNL) who played a lead role in the detailed analysis of FDO-680 and FDO-681 fission-product data summarized in this paper.

11. References

[1] M.R. Floyd, J. Pencer and B.P. Bromley, "A Canadian Perspective on Progress on Thoria Fuel Science and Technology"*CNL Nuclear Review*,accepted for publication 2016 March (CNL report CW-124950-CONF-009).

[2] S.J. Livingstone and M.R. Floyd, "Thoria Irradiation and Post-Irradiation Examination Experience at AECL", Proceedings of the 12th International Conference on CANDU Fuel, Kingston, Ontario, Canada, 2013 September 15-18 (AECL report CW-124950-CONF-006).

[3] S. Corbett, M.R. Floyd, H. Hamilton, S. Livingstone and N.F. Harrison, "DME-221 Thoria Fuel: Fabrication, Irradiation Testing, and Post-Irradiation Examination", Proceedings of the 12th International Conference on CANDU Fuel, Kingston, Ontario, Canada, 2013 September 15-18 (AECL report CW-124950-CONF-007).

[4] M. Karam, F.C. Dimayuga and J. Montin, "Post-Irradiation Examination of CANDU Fuel Bundles Fuelled with (Th, Pu)O_2", Proceedings of the 11th International Conference on CANDU Fuel, Niagara Falls, Ontario, Canada, 2010 October 17-20 (AECL report CW-124950-CONF-003).

[5] M. Karam, F.C. Dimayuga and J. Montin, "Fission Gas Release of (Th, Pu)O_2 CANDU Fuel", Proceedings of the 10th International Conference on CANDU Fuel, Ottawa, Ontario, Canada, 2008 October 5-8 (AECL report CW-124950-CONF-002).

[6] H. Hamilton, S. Livingstone and B. Popov, "Thorium Fuel Fabrication and Testing at AECL", International Workshop on Thorium Utilization for Sustainable Development of Nuclear Energy (TU2007), Beijing, China, 2007 December 4-6 (AECL report CW-124950-CONF-001).

[7] N.F. Harrison, T. Janathasing and F.C. Dimayuga, "Post-Irradiation Examination of MOX Fuel with Varying Pu Homogeneity", Proceedings of the 11th International Conference on CANDU Fuel, Niagara Falls, Ontario, Canada,2010 October 17-20 (AECL report CW-124920-CONF-004).

[8] M.R. Floyd, "Advanced Fuel Cycle Development at Chalk River Laboratories", Proceedings of the International Conference on Future of Heavy Water Reactors, Ottawa, Canada, 2011 October 2-5 (AECL report CW-124900-CONF-002).

[9] M. Saoudi, J. Mouris, A. Bergeron, H. Hamilton, D. Freiss, M. Cologna and D. Staicu, "Thermal Diffusivity and Conductivity of Thorium - Uranium Mixed Oxides", Workshop on Research into Nuclear Fuel in Europe and Materials Modeling and Simulation for Nuclear Fuels, Karlsruhe, Germany, 2015 November 16-18.(http://mmsnf.org/print.html)

[10] A. Prudil, J.S. Bell, A. Oussoren, P. Chan, and B. Lewis, "FAST: A Combined NOC and Transient Fuel Performance Model Using a Commercial FEM Environment", Proceedings of the 19th Pacific Basin Nuclear Conference, Vancouver, British Columbia, Canada, 2014 August 24-28, 2014.

[11] J.S. Bell and P.K. Chan, "Development of a Fuel Performance Model for Evaluating Conceptual Th-Based Canadian SCWR Fuel Designs", Proceedings of the 12th International Conference on CANDU Fuel, Kingston, Ontario, Canada,2013September15-18.

[12] J.S. Bell and P.K. Chan, "Thorium Based Fuel Performance Modeling and the Development of Multi-Pellet Material Fuel and Sheath Modeling Tool (MPM-FAST)", Proceedings of the 13th International Conference on CANDU Fuel, Kingston, Ontario, Canada, 2016 August 15-18.

[13] K.W. Lay, "Oxygen Chemical Diffusion Coefficient of Uranium Dioxide",*Journal of American Ceramic Society*, Vol. 53, pp. 369-373, 1970.

[14] P. Garcia, M. Fraczkiewicz, C. Davoisne, G. Carlot, B. Pasquet, G. Baldinozzi, D. Siméone and C. Petot, "Oxygen Diffusion and Electrical Conductivity Measurements inUranium Dioxide", *Defect and Diffusion Forum*, Vol. 297-301, pp. 966-971, 2010 (Online: 2010-04-13(2010) Trans Tech Publications, Switzerland).

[15] T. Matsui and K. Naito, "Chemical Diffusion Coefficient of Oxygen in Thoria-Urania Mixed Oxide",*Nuclear Materials*, Vol. 135, pp. 149-154, 1985.

[16] S. Livingstone, "Development of an On-Line Fuel Failure Monitoring System for CANDU Reactors", PhD thesis, Royal Military College of Canada, 2012 March.

[17] B. J. Lewis, C. R. Phillips and M J. F. Notley, "A Model for the Release of Radioactive Krypton, Xenon, and Iodine from Defective UO_2 Fuel Elements", *Nuclear Technology*, Vol. 73, pp. 72-83, 1986.

[18] D. Das and S.R. Bharadwaj (editors), "Thoria-Based Nuclear Fuels", Green Energy and Technology Series, Springer-Verlag Publishing (London), pp. 279-333, 2013.

[19] H. Hamilton, "Removal of Uranium from Thorium –Uranium SIMFUEL by Fluoride Volatility", Proceedings of the Actinide and Fission Product Partitioning and Transmutation 12th Information Exchange Meeting, Prague, Czech Republic, 2012 September 24-27(AECL report CW-124950-CONF-005).

[20] Nuclear Energy Agency, "Introduction of Thorium in the Nuclear Fuel Cycle: Short- to Long-Term Considerations", NEA report no. 7224, 2015.