Proceedings of the 12th Pacific Rim Conference on Ceramic and Glass Technology: Ceramic Transactions, Volume 264
Edited by Dileep Singh, Manabu Fukushima, Young-Wook Kim, Kiyoshi Shimamura, Nobuhito Imanaka, Tatsuki Ohji, Jake Amoroso and Michael Lanagan

NUCLEAR FUEL MODELLING AND PERSPECTIVES ON CANADIAN EFFORTS IN FUEL DEVELOPMENT

M.H.A. Piro[1,2], A. Prudil[1], M.J. Welland[1], W. Richmond[3], A. Bergeron[3], E. Torres[4], C. Maxwell[4], J. Pencer[4], N. Harrison[3], M. Floyd[3]

[1] Fuel & Fuel Channel Safety Branch, Canadian Nuclear Laboratories, Chalk River, Canada
[2] Faculty of Energy Systems and Nuclear Sciences, University of Ontario Institute of Technology, Oshawa, Canada
[3] Fuel Development Branch, Canadian Nuclear Laboratories, Chalk River, Canada
[4] Computational Physics Branch, Canadian Nuclear Laboratories, Chalk River, Canada

ABSTRACT

Significant progress has been made in recent years in developing a new generation of nuclear fuel models, simulations, and codes with the overall goal of enhancing predictive capabilities of nuclear fuel behaviour to improve performance and safety. Computational efforts span the entire multi-scale multi-physics spectrum, ranging from atomistic electronic structure and classical inter-atomic potential calculations, to meso-scale simulations of micro-structural evolution, to continuum scale thermo-mechanical simulations of fuel performance. These computational activities support performance and safety analyses for industry while also providing guidance to various experimental research programs. The aforementioned computational research has supported Canadian efforts in the development and testing of non-conventional fuels, including thoria-urania, thoria-plutonia, and urania-plutonia fuels. Advanced fuel cycle development activities at the Canadian Nuclear Laboratories include fuel fabrication, post-irradiation assessments, safety related testing, materials behaviour, etc. This paper will give a high-level overview of recent advances in nuclear fuel modelling and simulation activities in addition to providing some perspectives on efforts in nuclear fuel development in Canada.

INTRODUCTION

Understanding the behaviour of fuel in a nuclear power reactor is of paramount importance to maximize performance, reliability, and safety. A variety of scientific disciplines must be understood in regards to nuclear fuel behaviour, including heat transfer, structural and fracture mechanics, thermodynamics, materials science, and reactor physics. A great difficulty in capturing the interdependencies of the foregoing mechanisms is that it is impossible to reproduce the conditions inside of a nuclear reactor in the laboratory; therefore, one must ultimately perform experiments in a test reactor while delegating some experiments to the laboratory when possible. This has been the conventional approach to investigate nuclear fuel behaviour, whereby computational activities typically provide more of a supporting role.

Contemporary approaches are moving towards augmenting in-reactor testing in part through sophisticated modelling and simulation techniques since in-reactor testing is very expensive, time consuming, and requires additional procedures to adequately address health and safety risks. Significant progress has been made in a variety of computational approaches in recent years, which has been facilitated by the more recent availability of affordable high performance computing equipment. Indeed, one cannot rely entirely on computational methods

as a research and development tool; the intent is to augment and compliment experimental activities to minimize the number of unnecessary experiments.

Canadian interests in nuclear fuel research and development have been mainly focused on uranium dioxide ceramic fuels, which are used in CANDU nuclear generating stations, and to a lesser extent in metallic dispersion fuels, which are used in research and test reactors. Some other mixed oxide fuels have been investigated for use in current generation technologies and next generation reactor concept designs. The National Research Universal (NRU) reactor – located in Chalk River, ON – has played a critical role in virtually all research and development activities of nuclear fuels investigated in Canada. The role that the NRU has played in nuclear fuel research in addition to other experimental activities and computational research will be described in the following sections, which will conclude with some high-level perspectives of recent and anticipated developments.

NUCLEAR FUEL MODELLING

The following sub-sections discussing various aspects of nuclear fuel modelling are categorized with respect to their respective length and time scales, which is illustrated in Figure 1. First, continuum scale simulations are described, which are typically used to assess fuel performance from an engineering perspective. Second, thermodynamic modelling is used to predict phase equilibria. Third, mesoscale simulations generally apply to lower length scales to predict microstructural evolution. Finally, atomistic scale modelling is used to predict electronic structure and atomic scale behaviours, which can be used to predict fundamental material properties as input to higher scale models.

Continuum Scale Simulations

The fuel evolves significantly during irradiation as a result of strong radiation fields, high temperatures, large temperature gradients, and fission product accumulation. Continuum scale fuel performance codes are used to predict important macroscopic fuel performance parameters such as maximum fuel temperature, cladding strain, or fission gas release as a function of the fuel design and irradiation conditions. The ultimate goal of this type of analysis is to predict whether the fuel will remain within design and licensing limits. A unique challenge for Canadian fuel performance codes is the high volumetric heat generation rate compared to other reactor designs, leading to much higher temperatures during Normal Operating Conditions (NOC).

At the start of irradiation, the pellet size increases as a results of fuel thermal expansion and thermal stress-induced cracking. The high temperatures then gradually reduce the fuel porosity as it continues to sinter during operation (known as densification). Eventually, the fuel swells as a result of the accumulation of fission products and comes into mechanical contact with the cladding, called Pellet-Cladding Mechanical Interaction. This contact introduces tensile stresses in the cladding that contribute to Stress-Corrosion Cracking of the cladding by volatile fission products, thereby limiting both the operating power of the fuel as well as the maximum rate of change of the fuel power. If through-wall cracks develop in the cladding, coolant water may enter the fuel leading to fuel oxidation and further cladding degradation due to secondary hydriding from the zirconium-steam interaction inside the cladding. Cladding failure may release fission products to the primary coolant system, which increases plant maintenance costs and can force reactor shutdowns if radioactivity in the coolant reach license limits.

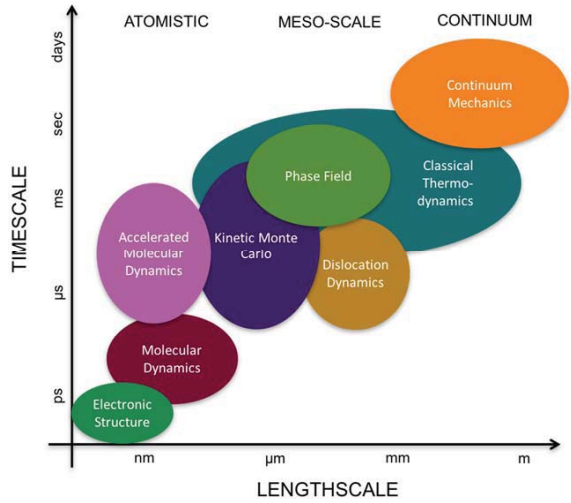

Figure 1. The variation in spatial and temporal domains of different modelling approaches are illustrated[1].

Fuel performance models attempt to account for the macroscopic strains resulting from thermal-expansion, densification, cracking, elastic and plastic deformations as well as intra-and-intergranular swelling in order to estimate the mechanical contact stresses experienced by the cladding. This is further complicated by time-varying power generation and coolant conditions. The uncertainty in fuel performance code predictions is largely a result of material property uncertainties, stochastic effects (e.g., cracking patterns), manufacturing variability (e.g., microstructure, defects) and irradiation condition uncertainty (e.g., power generation rate and coolant conditions).

The primary Canadian continuum scale fuel codes developed from the late 1960s to the 1990s are ELESIM[2]/ELESTRES[3] for NOC, and the ELOCA[4] code for accident transients. These codes are primarily one dimensional and based heavily on semi-empirical approaches. They are intended to provide design-centred fuel performance predictions for UO_2 ceramic fuel based on a large database of fuel irradiations and experiments resulting from NRX, NRU and Whiteshell Laboratory test loop irradiations and power reactor irradiations. In the early 2000s, considerable effort was spent documenting, validating, and verifying these codes to provide industry standard fuel modelling codes for licensing analysis purposes[5-8]. These codes are well suited for this purpose; however, they rely heavily on empirical correlations derived from experimental results and have not capitalized on recent advancements in computer hardware and software. Most recently, the focus has been on the development of multidimensional mechanistic codes that better capture the physics and offer greater flexibility at the cost of greater computational expense[9-11]. The multi-physics approach is also being done by coupling fuel performance codes to reactor physics and thermal hydraulic codes to predict irradiation conditions[12,13]. For this application, the Canadian Nuclear Laboratories (CNL) is evaluating the BISON fuel code being

developed at Idaho National Laboratories[14], based on the MOOSE finite-element multi-physics framework[15].

Models for alternate fuel cycles are also being considered in order to improve the sustainability of nuclear power by incorporating thorium and/or recycled plutonium to increase supply and reduce waste[16-19]. These elements are usually mixed as oxides (i.e., $(Th,U)O_2$, $(Th,Pu)O_2$ and $(U,Pu)O_2$) prior to pressing into pellets and sintering. One of the greatest challenges for modelling the performance of these fuels is determining the composition-dependent material properties. In the past, this was done empirically; however, as a result of the high cost of in-reactor experiments and/or handling radioactive material it is being augmented by computer modelling. The manufacturing process may also introduce other complications, such as heterogeneity due to uneven mixing of the powders, resulting in fuel microstructures that may lead to increased fission gas release[20]. The ability to simulate and rapidly virtually prototype composition-dependent material properties utilizing the multidimensional mechanistic codes mentioned earlier offers a greater degree of flexibility and capability for modelling new fuels in comparison to the earlier fuel codes developed and used for modelling conventional power reactor fuels. An example of a temperature distribution in a fuel element obtained using the FAST code[21] is shown in Figure 2 along with the finite-element mesh and labels indicating the components of the geometry.

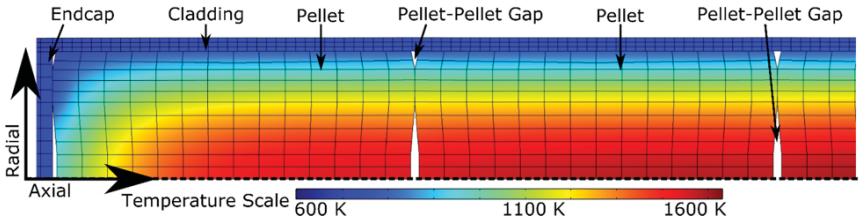

Figure 2. Example temperature distribution calculated using the FAST code[21] with finite-element mesh and labels overlaid.

Thermodynamic Modelling

A concerted effort has been put forth to developing a thermodynamic treatment of irradiated uranium dioxide nuclear fuel[22]. In addition to a model of non-stoichiometric UO_{2+x} (as shown in Figure 3a), which provides the foundation of the treatment, subsequent models have been added to capture the thermochemical behaviour of fission and activation products. The complete treatment includes approximately two dozen chemical elements, a dozen liquid and solid solution phase models, and numerous stoichiometric solid phases and gaseous species. This treatment has been used to predict the behaviour of simulated irradiated fuel (SIMFUEL)[23], irradiated fuel chemistry[24], and fuel melting[25].

In addition to thermodynamic modelling of power reactor fuels, work has also been performed to analyze the behaviour of research reactor fuels. In particular, thermodynamic

analyzes have been performed of U-Mo/Mg fuels as a candidate replacement design for the NRU reactor, which is intended to offer a high degree of reliability in-reactor while also addressing proliferation concerns through the use of low enriched uranium. In this design, U-Mo fuel particles are dispersed in a Mg matrix. Figure 3b shows the U-Mo phase diagram with the U-7Mo and U-10Mo fuel compositions highlighted. The main purpose of thermodynamic modelling of this fuel design was to compute material properties as input to heat transfer calculations and to also confirm chemical stability between the U-Mo fuel particles and surrounding matrix.

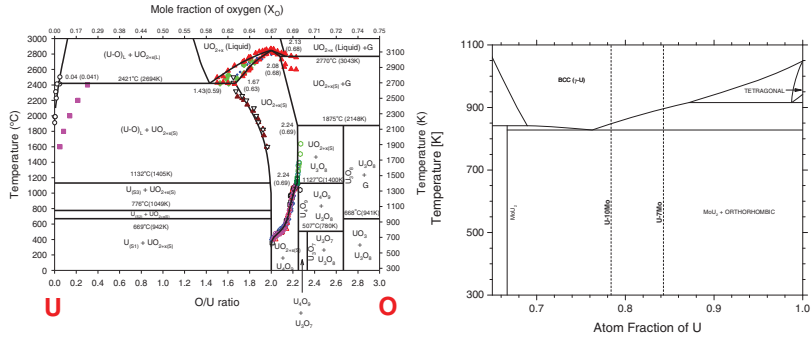

Figure 3. A) The U-O phase diagram is illustrated on the left with experimentally measured phase transitions superimposed[22]. B) The U-Mo phase diagram is shown on the right with pertinent fuel compositions highlighted[26].

Meso-Scale Models

Mesoscale phenomena occur at a length scale where behaviour is dominated by interfacial energy and effects including microstructure evolution, such as grain growth, and the behaviour of nano- to micro-scopic bubbles. In nuclear fuels, gaseous fission products (e.g., Xe, Kr) are continually generated during irradiation and accumulate within the solid matrix. Depending on how this gas is accommodated in the fuel's microstructure, it can alter the macroscopic behaviour by decreasing the local thermal conductivity, contributing to swelling of the fuel, and degrading heat-transfer across the fuel-to-cladding gap.

Fission gasses have a very low solubility in the solid matrix and are driven to migrate towards nearby voids which, in the bulk material, occur as intragranular bubbles. Despite accumulation of gas in the bubble, their growth is limited by the arrival of vacancies. Uranium is known to migrate via the interchange mechanism and that the self-diffusion coefficient is slow, and therefore cation vacancies are kinetically limited[27]. Bubbles may therefore over-pressurise compared to the equilibrium Laplace pressure of bubbles of their size, which exerts a stress field surrounding them, drawing in more vacancies[28].

This process was simulated by a thermodynamically self-consistent multicomponent phase-field model, which incorporated thermodynamic potentials directly via a quadratic approximation[28]. The model is capable of exploring non-equilibrium processes such as the

kinetically limiting diffusion and effects of interfacial energy. The formulation allows for variable local density and correctly reproduces increase in gaseous density in the bubble as a function of bubble size in agreement with the Young-Laplace equation.

Since vacancies are generated at grain boundaries and the bubble grows by vacancy adsorption, the bubbles grow preferentially towards the grain boundary while maintaining their spherical shape by interfacial energy effects as shown in Figure 4. The bubbles essentially migrate towards the grain boundary and as a result of the grain-grain interfacial energy the bubble becomes lenticular. This migration may contribute to the 'denuded' zone where the region adjacent to the grain boundary is devoid of bubbles.

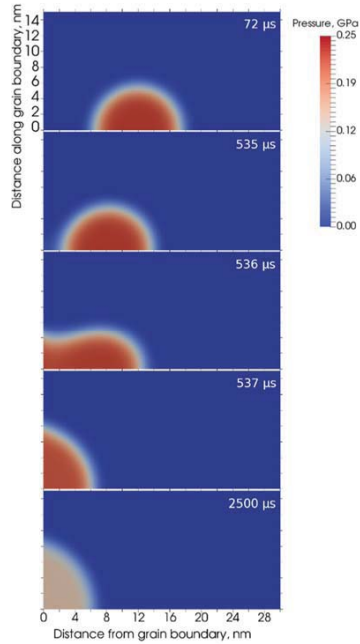

Figure 4. Snapshots of the time evolution of the intragranular bubble migrating and merging with the grain boundary, colored according to pressure. The simulation is azimuthally symmetric about the bottom boundary.

Bubbles on the grain boundary may interconnect, resulting in a long range, labyrinthine of tunnels along the grain boundary network, as shown in Figure 5. Once the interconnected network reaches a crack or surface, the gas in the tunnel network vents to the free element volume and the tunnel may collapse. The microstructure is thus altered by these events, and may contribute significantly to the macroscopic swelling of the fuel and the heat transport across the fuel-cladding gap. While phase-field models are demonstrated to be useful for simulating

bubbles mesoscale phenomena, they can be computationally expensive due to the requirement of resolving the interface in 3D. To reduce the computational expense, a new shell-porosity model has been developed recently which projects the surface of the intergranular porosity onto the 2D grain boundary network[29].

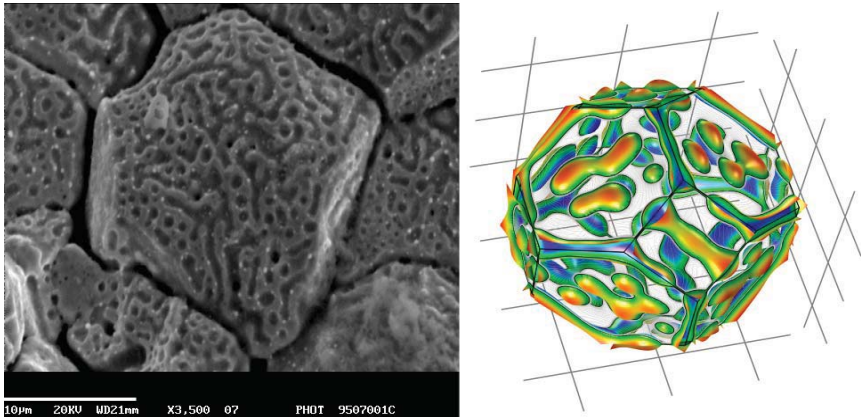

Figure 5. Interconnected intergranular porosity which accomodates fission gas within the solid fuel matrix. Left: example Scanning Electron Microscope images of advanced stages of grain boundary porosity formation[30,31]. Right: The newly developed shell porosity model which qualitatively captures porosity morphology[29].

The shell-porosity model derives from an energy functional of the system accounting for the shape of the intergranular bubbles. Time evolution proceeds to reduce the system's energy while conserving the quantity of species within the bubbles, in accordance with the Theory of Irreversible Processes. The predictions were verified against classical contact-angle theory to good agreement, and the sensitivity of the model to numerical parameters is discussed[29].

The shell-porosity model captures the 3D behaviour of the intergranular bubbles on a 2D surface, and therefore is a 2D simulation embedded in 3D space. This drastically reduces the computational expense of these simulations, enabling larger simulations to be run that more realistically capture the long-range behaviour of this phenomenon. A qualitative example is shown in Figure 5b, which shows a simulation result with interlinked intergranular bubbles. As a consequence of the projection of the interface on to the grain boundary, the technique is limited to contact angles of less than 90°.

Atomistic Scale Modelling

Atomistic scale simulations have become an increasingly valuable complement to experiments in the understanding of the behavior and properties of nuclear fuel materials. The combination of theoretical, computational, and experimental efforts can provide fundamental information about the properties of nuclear fuels relating atomic scale phenomena to

macroscopic behavior[32]. Atomistic simulations enable the modelling of essential phenomena, such as defect trapping and path of migration, that govern the time-evolution of void formation, fission gas diffusion in the bulk and at grain boundaries, and the onset of bubble formation. The effects of single atoms, clusters and bubbles on the basic properties such as elasticity or thermal conductivity can also be investigated. In cases where experimental data is scarce, such as with new or novel fuel material compositions, atomistic simulations can provide input parameters for larger scale simulations. The modelling of materials at atomistic scale requires an accurate description of the interactions between atoms, which is a crucial component in atomistic simulations. Properties of systems considering a few hundred atoms can be accurately determined from electronic structure calculations[33-35]. On the other hand, large systems or time-scales can be modelled using classic mechanical approaches, therefore requiring empirical functional models to describe interactions between atoms[36].

Theoretical calculations using standard Density Functional Theory (DFT) can accurately reproduce experimentally measured material properties. For instance, DFT can reasonably reproduce lattice parameters, bulk modulus, and cohesive energy of nuclear fuels. However, DFT alone does not accurately describe the strongly correlated behavior of f-electrons in actinoid elements. To accurately account for strong correlation effects due the localized f-electrons, simulations of actinoid based materials can be performed within the Hubbard corrected DFT method (DFT+U)[37,38]. Calculations of actinoid oxide materials, using the DFT+U approach, can accurately reproduce the magnetic ground state, band gap, and predict the relative stability of impurity atoms[39,40]. Simulations based on the DFT+U approximation have been performed to study the properties of mixed oxide nuclear fuels and the behavior of fission products gases[32-34]. Predictions from the DFT+U method can provide direct input to theory-based engineering models, such as diffusion constants for point defects and impurities. Furthermore, energies and forces evaluated with DFT+U can be used to derive classical interatomic potentials for use in higher scale modelling, where the computational cost associated with the larger system sizes precludes use of the DFT+U approach alone.

In classical mechanics simulations, usually known as Molecular Dynamics (MD), the time evolution of each atom in the system is numerically integrated and the equilibrium bulk properties of the system are obtained from statistical mechanics evaluations. Many-body potentials, such as the Embedded Atom Method (EAM) and the Modified Embedded Atom Method (MEAM) have been successfully used for calculations of equilibrium energy and thermal properties of nuclear materials[41,42]. However, the explicit consideration of charge transfer is still a challenge in MD simulations. Earlier nuclear fuel simulations based on empirically derived interatomic potentials have been successful in predicting some nuclear fuel properties, such as thermal conductivities of fuel[43]. Such models are less reliable in their description of point defect interactions and fission gas diffusion, in part because of the lack of experimental data upon which to tune interatomic potentials to reproduce point defect and impurity migration energies. An accurate representation of such point defect and migration energies can be derived theoretically and reliably through first principles methods such as DFT or DFT+U. These DFT or DFT+U-based results, in turn, can be used to derive interatomic potential parameters appropriate for large scale simulations of point defect or impurity diffusion, such as for helium in nickel[44] and xenon in uranium[45]. CNL is currently working on building on existing studies to derive interatomic potentials suitable for large scale simulations to predict thermal conductivity and fission gas diffusion under various conditions for various candidate advanced nuclear fuel compositions.

NUCLEAR FUEL EXPERIMENTS

Fuel Fabrication

Fuel fabrication capabilities are key to enabling fuel development and modelling activities. The fabrication of high quality fuel to within design specifications and the data generated from testing of these fuels provides valuable input for designers and modelers. CNL (formerly Atomic Energy of Canada Ltd.) has had a long history of fabricating a variety of fuel types including: UO_2 and UO_2-based fuels (i.e., UO_2 containing neutron poisons such as Dy and Gd, SIMulated irradiated FUEL- SIMFUEL mimicking the chemical composition of irradiated fuel, UO_2 with additives for enhanced grain growth, etc.), ThO_2 and ThO_2-based fuels (including SIMFUEL), mixed oxide fuels (UO_2-ThO_2, UO_2-PuO_2, ThO_2-PuO_2), and zirconia pellets containing burnable neutron absorbers. Fuel has been fabricated at laboratory and pilot scales with fuel fabrication activities performed under a Quality Assurance Plan compliant to Z299.2 and ISO 9001:2008 quality programs.

CNL is building on its past fuel fabrication experience to support the development of next generation nuclear fuels. Advanced nuclear fuel concepts are being proposed that offer higher performance, proliferation resistance, increased safety, and accident tolerance. These fuel designs are often based on advanced materials, have inhomogeneous structures, or have complex geometries. Many of the next generation nuclear fuels cannot be manufactured via conventional processes applied to traditional UO_2 based nuclear fuels. Therefore, new methods are required for their fabrication. Current research programs at CNL are focusing on modern manufacturing technologies such as spark plasma sintering and additive manufacturing. If these technologies can be successfully applied to the fabrication of new fuel concepts, experimentation and modelling can be performed to advance their development.

In-Reactor Testing

Canada has a rich history of conducting in-reactor tests to support nuclear fuel development. These tests have been essential to producing data that can be used in the development and validation of models that predict fuel behaviour. Canadian in-reactor fuel tests have been primarily conducted in the NRU and NRX reactors (located at Chalk River, Ontario), NPD (Rolphton, Ontario) and WR-1 reactors (Whiteshell, Manitoba). Instrumented fuel tests were performed primarily in NRX, but also in NRU. These tests have primarily supported Pressurized Heavy Water Reactor (PHWR) fuel development and included natural U, slightly enriched U, (U, Pu)O_2 (MOX) and various thoria-based fuels, including ThO_2, (Th, U)O_2 and (Th, Pu)O_2. Synergies with spent LWR fuels have also been explored, as have fuels containing burnable neutron absorbers.

Post-Irradiation Assessments

Hot cell facilities at Chalk River, Ontario and Pinawa, Manitoba have enabled Canadian post-irradiation fuel performance assessments for more than 50 years. These assessments have included experimental fuels irradiated in NRU, NRX, NPD and WR-1, as well as fuels irradiated in commercial power reactors (primarily Canadian-based PHWRs). These assessments have

produced data for key fuel performance parameters that are calculated by fuel models, including fission-gas release, clad strain and pellet grain growth. This data has been foundational to the development and validation of nuclear fuel models.

PERSPECTIVES ON CANADIAN EFFORTS IN FUEL DEVELOPMENT

Canada has a rich heritage of nuclear fuel development that has primarily focused on support for ceramic-based fuels associated with PHWR technology. This includes major efforts in the areas of fabrication development, materials properties measurements, in-reactor testing, and post-irradiation performance assessment. Fuel types have included natural uranium, slightly-enriched uranium, mixed-oxide (U-Pu), and thoria-based (including Th-U and Th-Pu) designs. Work in these areas has laid the ground work for the development and validation of fuel computational models for various fuels over a wide range of operating conditions. These models are key to enabling Canada and the world to understand and predict fuel behaviour in support of diverse initiatives including fuel design, operations, safety, and licensing.

CONCLUSION

Significant efforts have been put forth to establish a combined experimental, computational, and theoretical understanding of ceramic nuclear fuel behaviour for Canadian purposes. Nuclear fuel modelling efforts span the entire multi-scale paradigm, ranging from fundamental material property predictions based on quantum mechanical effects to continuum scale simulations that are of direct use for engineering purposes. Experimental facilities and capabilities have been well established in fuel fabrication, in-reactor testing, and post-irradiation assessment to support nuclear fuel performance and safety.

ACKNOWLEDGEMENTS

This research was undertaken, in part, thanks to funding from the Canada Research Chairs program.

REFERENCES

[1] Stan, M. (2009). Discovery and Design of Nuclear Fuels. *Materials Today*, Volume 12, Issue 11, 20-28.
[2] (1975). ELESIM II - A Computer Program for Modelling CANDU Fuel Element Performance. Atomic Energy of Canada Limited.
[3] Tayal, M. (1987). Modelling CANDU Fuel Under Normal Operating Conditions: ELESTRES Code Description. Atomic Energy of Canada Limited.
[4] Sills, H.E. (1979). ELOCA: Fuel Element Behavior During High-Temperature Transients. Atomic Energy of Canada Limited.
[5] Moeck, E.O., Luxat, J.C., Simpson, L.A., Petrilli, M.A., & Thompson, P.D. (1996). Validation of Computer Codes used in Safety Analyses of CANDU Power Plants. *Proceedings of Canadian Nuclear Society*, Fredericton, Canada.
[6] Luxat, J.C., Kupferschmidt, W., Thompson, P.D., & Petrilli, M.A. (2000). The Industry Standard Toolset (IST) of Codes for CANDU Safety Analysis. in: Barcelona, Spain.

[7] Sim, K.-S., Chassie, G.G., Xu, Z., Tayal, M., & Westbye, C. (2001). Progress in Qualifying ELESTRES-IST 10 code: Verification and Interim Results of Validation. *Proceedings of Canadian Nuclear Society*, Kingston, Canada.
[8] Popov, N., Wren, D., Snell, V., White, A., & Boczar, P. (2003). Development and Qualification of AECL Computer Codes for ACR Safety Analysis Applications. *Proceedings of Canadian Nuclear Society*, Toronto, Canada.
[9] Prudil, A., Lewis, B.J., Chan, P.K., & Baschuk, J.J. (2015) Development and testing of the FAST fuel performance code: Normal operating conditions (Part 1), *Nuclear Engineering and Design*, Volume282, 158–168.
[10] Chassie, G., & Shams, M. (2016). 3D Modelling Capability for CANDU Fuel Performance. *Proceedings of International Conference on CANDU Fuel*, Kingston, Canada.
[11] Williams, A., Yatabe, S., & Belov, A. (2016). Finite Element Simulations of a Fuel Element Simulator Sagging Experiment. *Proceedings of International Conference on CANDU Fuel*, Kingston, Canada.
[12] Liu, Y., Xu, T., & Seydaliev, M. (2016). Multi-physics Reactor Transient Simulations Through a SALOME-Backbone Joint Platform, *Proceedings in International Conference on CANDU Fuel*, Kingston, Canada.
[13] Liu, Y., Nishimura, M., Seydaliev, M., & Piro, M. (2016). Backbone: A Multiphysics Framework for Coupling Nuclear Codes Based on CORBA and MPI, *Journal of Nuclear Engineering and Radiation Science*, Volume 3, 11020-11030.
[14] Hales, J.D., Novascone, S.R., Spencer, B.W., Williamson, R.L., Pastore, G., & Perez, D.M. (2014). Verification of the BISON Fuel Performance Code, *Annals of Nuclear Energy*, Volume 71, 81–90.
[15] Gaston, D., Newman, C., Hansen, G., & Lebrun-Grandié, D. (2009). MOOSE: A Parallel Computational Framework for Coupled Systems of Nonlinear Equations, *Nuclear Engineering and Design*, Volume 239, 1768–1778.
[16] Bell, J.S., Prudil, A., Chan, P.K., Lewis, B.J., & Bonin, H.W. (2013). Development of a Fuel Performance Model in Support of the Canadian SCWR fuel, *Proceedings in International Symposium Supercritical Water-Cooled Reactor*, Shenzhen, China.
[17] Bell, J.S., & Chan, P.K. (2013). Development of a Fuel Performance Model for Evaluating Conceptual Th-based Canadian SCWR Fuel Designs, *Proceedings in International Conference CANDU Fuel*, Kingston, Canada.
[18] Prudil, A., Welland, M., Richmond, W., & Yatabe, S. (2016). Preliminary Modelling of U-Pu MOX Fuel in CANDU Geometries using BISON, *Proceedings in International Conference of CANDU Fuel*, Kingston, Canada.
[19] Chassie, G., Jiang, A., Gill, A., & Shams, M. (2016). MOX Fuel Performance Modelling of CANDU Fuel, *Proceedings in International Conference of CANDU Fuel*, Kingston, Canada.
[20] Dimayuga, F.C., Floyd, M.R., & Cox, D.S. (1999). Fabrication and Performance Testing of CANDU Mixed-Oxide Fuel, Atomic Energy of Canada Limited.
[21] Prudil, A., Lewis, B.J., Chan, P.K., Baschuk, J.J., & Wowk, D. (2015). Development and Testing of the FAST Fuel Performance Code: Transient Conditions (Part 2). *Nuclear Engineering and Design*, Volume 282, 169–177.
[22] Thompson, W.T., Lewis, B.J., Corcoran, E.C., Kaye, M.H., White, S.J., Akbari, F., He., Z., Verrall, R., Higgs, J.D., Thompson, D.M., Besmann, T.M., Vogel, S.C. (2007). Thermodynamic

Treatment of Uranium Dioxide Based Nuclear Fuel. *International Journal of Materials Research*, Volume 98, Issue 10, 1004-1011.
[23] Corcoran, E.C., Lewis, B.J., Thompson, W.T., Mouris, J. & He, Z. (2011). Controlled Oxidation Experiments of Simulated Irradiated UO_2 Fuel in Relation to Thermochemical Modelling. *Journal of Nuclear Materials*, Volume 414, 73-82.
[24] Piro, M.H.A., Banfield, J., Clarno, K.T., Simunovic, S., Besmann, T.M., Lewis, B.J., & Thompson, W.T. (2013). Coupled Thermochemical, Isotopic Evolution and Heat Transfer Simulations in Highly Irradiated UO_2 Nuclear Fuel. *Journal of Nuclear Materials*, Volume 441, 240-251.
[25] Welland, M.J., Thompson, W.T., Lewis, B.J., & Manara, D. (2009). Computer Simulations of Non-Congruent Melting of Hyperstoichiometric Uranium Dioxide. *Journal of Nuclear Materials*, Volume 385, 358-363.
[26] Kulakov, M., Saoudi, M., Piro, M.H.A., & Donaberger, R.L. (2017). Structure and Thermal Properties of As-Fabricated U-7Mo/Mg and U-10Mo/Mg Low-Enriched Uranium Research Reactor Fuels, *Journal of Nuclear Materials*, Volume 484, 288-296.
[27] Yakub, E., Ronchi, C., & Staicu, D. (2007). Molecular Dynamics Simulation of Premelting and Melting Phase Transitions in Stoichiometric Uranium Dioxide, *Journal of Chemical Physics*, Volume 127, Number 9, 094508.
[28] Welland, M.J., Tenuta, E., & Prudil, A.A. A Linearization-Based Method for Solving a Multicomponent Diffusion Phase-Field Model with Arbitrary Solution Thermodynamics. In review.
[29] Prudil, A.A., & Welland, M.J. (2017). A Novel Model of Third Phase Inclusions on Two Phase Boundaries", *Materials Theory*, In Press.
[30] White, R.J. (2004). The Development of Grain-Face Porosity in Irradiated Oxide Fuel. *Journal of Nuclear Materials*, Volume 325, 61–77.
[31] White, R.J., Corcoran, R.C., & Barnes, P.J. (2006). IFPE/CAGR-UOX-SWELL Fuel Swelling Data Obtained from the AGR/Halden Ramp Test Programme, Organization for Economic Cooperation and Development Nuclear Energy Agency, Paris, France.
[32] Samaras, M., Victoria, M., & Hoffelner, W. (2009). Nuclear Energy Materials Prediction: Application of the Multi-Scale Modelling Paradigm, *Nuclear Engineering and Technology*, Volume 41, 1-10.
[33] Hernandez, S.C., & Holby, E.F. (2016). DFT+U Study of Chemical Impurities in PuO_2, *Journal of Physical Chemistry C*, Volume 120, 13095−13102.
[34] Wang, B.-T., Zheng, J.-J., Qu, X., Li, W.-D., & Zhang, P, (2015). Thermal Conductivity of UO_2 and PuO_2 from First Principles, *Journal of Alloys and Compounds*, Volume 628, 267–271
[35] Wang, B.-T., Zhang, P., Lizárraga, R., Di Marco, I., & Eriksson, O. (2013). Phonon Spectrum, Thermodynamic Properties, and Pressure-Temperature Phase Diagram of Uranium Dioxide, *Physical Review B*, Volume 88, 104107.
[36] Arima, T., Yamasaki, S., Inagaki, Y., & Idemitsu, K. (2005). Evaluation of Thermal Properties of UO_2 and PuO_2 by Equilibrium Molecular Dynamics Simulations from 300 to 2000 K. *Journal of Alloys and Compounds*, Volume 400, 43–50
[37] Jomard, G., Amadon, B., Bottin, F., & Torrent, M. (2008). Structural, Thermodynamic, and Electronic Properties of Plutonium Oxides from First Principles. *Physical Review B*, Volume 78, 075125.

[38] Dorado, B., Amadon, B., Freyss, M., & Bertolus, M. (2009). DFT+U Calculations of the Ground State and Metastable States of Uranium Dioxide. *Physical Review B,* Volume 79, 235125.
[39] Dorado, B., Jomard, G., Freyss, M., & Bertolus, M. (2010). Stability of Oxygen Point Defects in UO_2 by First-Principles DFT+U Calculations: Occupation Matrix Control and Jahn-Teller Distortion. *Physical Review B,* Volume 82, 035114.
[40] Dorado, B., Freyss, M., Amadon, B., Bertolus, M., Jomard, G., & Garcia, P. (2013). Advances in First-Principles Modelling of Point Defects in UO_2: f Electron Correlations and the Issue of Local Energy Minima. *Journal of Physics: Condensed Matter*, Volume 25, 333201
[41] Cooper, M.W.D., Rushton, M.J.D., & Grimes, R.W. (2014). A Many-Body Potential Approach to Modelling the Thermomechanical Properties of Actinide Oxides, *Journal of Physics: Condensed Matter*, Volume 26, 105401
[42] Cooper, M.W.D., Murphy, S.T., Rushton, M.J.D., & Grimes, R.W. (2015). Thermophysical Properties and Oxygen Transport in the $(U_x,Pu_{1-x})O_2$ Lattice. *Journal of Nuclear Materials*, Volume 461, 206–214.
[43] Arima, T., Yamasaki, S., Idemitsu, K., & Inagaki, Y. (2008). Equilibrium and Nonequilibrium Molecular Dynamics Simulations of Heat Conduction in Uranium Oxide and Mixed Uranium–Plutonium Oxide. *Journal of Nuclear Materials*, Volume 376, 139–145.
[44] Torres, E., Pencer, J., & Radford, DD. (2016). Density Functional Theory-Based Derivation of an Interatomic Pair Potential for Helium Impurities in Nickel. *Journal of Nuclear Materials*, Volume 479, 240-248.
[45] Thompson, A.E., & Wolverton, C. (2013). Pathway and Energetics of Xenon Migration in Uranium Dioxide. *Physical Review B*, Volume 87, 104105.