Fuel Engineering Chemistry Research at the Royal Military College of Canada

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ABSTRACT

The authors conduct and direct computational and experimental research in nuclear fuel engineering that combines physical chemistry focused on matters of phase stability in multi-element systems coupled to, where possible, heat and mass transfer modelling associated with reactor operation and safety issues. Recently, the research program has expanded as a consequence of collaboration between the authors, the CANDU Owners Group, Chalk River Laboratories of Atomic Energy of Canada Limited, and the Natural Sciences and Engineering Research Council of Canada. This has also opened the way to research programs with other organizations in the nuclear industry in Canada and the United States.

The presentation will highlight research projects presently underway as well as those concluded in the recent past that led to the current collaboration and, with examples, show the way the authors connect their specialties to commercial requirements. The presentation will conclude with observations on the linkage between academic engineering research and the Canadian nuclear industry and how, in their opinion, this collaboration can be further improved and advanced.

1. INTRODUCTION

Over many years, nuclear fuel research has focused on improvement of fuel performance with the current CANDU bundle design and, more recently, in the strategic development of new advanced fuel designs for enhanced performance, economics and reactor safety. For example, an advanced fuel bundle design has been proposed for the low-void reactivity fuel (LVRF) project for Bruce Power and the Advanced CANDU Reactor (ACR), which employs as the central fuel element UO_2 doped with Dy_2O_3 in order to reduce the positive void reactivity.[1,2,3] In the manufacturing process of advanced fuels, it is important to understand bulk diffusion processes in order to homogenize the fuel material. One must further understand complications that could potentially arise with the occurrence of different phases. This understanding is particularly important in order to avoid any potential for fuel cracking in the doped-fuel manufacturing process. During operation with doped fuel, oxidation products unknown to conventional fuel may form such as (UDy_6O_{12}), which could affect the fuel loss and fission product release characteristics during operation with defected fuel. Research at the Royal Military College of Canada (RMC) is now focusing on such key issues in support of the manufacture and operation of such fuels.

Even with conventional UO_2 fuel, a better understanding of the behaviour of defected fuel is required to enhance safety margins and for economic reliability. For instance, fission products and fuel debris can escape into the primary heat transport system, which can increase the occupational exposure.[4] Over the last number of years, the I-131 action limits, which could lead to the premature shutdown of the reactor, have been reduced significantly. Thus, a better ability to predict the fission product release behaviour from defected fuel during steady power operation as well as reactor startup, shutdown and bundle shifting operations is required.[5] It is also important to develop techniques to characterize defected fuel and uranium contamination during reactor operation, and particularly locate any defected bundle(s) for quick removal. The presence of heavy-water vapour in the fuel-to-sheath gap can lead to a degradation in the element thermal performance since the gap heat transfer coefficient may be reduced. Moreover, the fuel oxidation process can lead to a degraded thermal conductivity in the hyperstoichiometric fuel, a lower incipient melting temperature and an enhanced fission-product release with a greater fission-product mobility in the hyperstoichiometric fuel matrix.[6,7,8] In order to assess the impact of in-reactor fuel oxidation on the fuel thermal performance, it is necessary to determine the relative importance of in-bay oxidation effects, especially when post-irradiation data are used for validation of fuel-oxidation models.[9] The development of models for defect fuel for fuel performance codes could be useful to assess the impact of defected fuel operation in safety and licensing analysis.

Fundamental property measurements are required to describe the thermal and mechanical behaviour of the fuel. Of particular importance for the time-dependent heat conduction equation is the thermal conductivity, heat capacity and density/thermal expansion of the fuel. Although these properties are generally well established for unirradiated uranium dioxide, such properties are less well known (particularly, at high temperatures) for irradiated fuel (containing fission products), and for fuel which may be doped in advanced designs and/or for fuel which has been oxidized due to operation in a defected state. Extrapolated thermal properties have a much larger uncertainty as the temperature increases. One must therefore rely increasingly on theoretical means to acquire the necessary high-temperature properties.

Further understanding is needed to ascertain the high-temperature behaviour of the fuel material and associated fission product release under different atmospheric (hydrogen/steam) environments as a result of transient conditions.[10] For example, release models are particularly needed to describe the behaviour of low-volatile fission products under different atmospheric conditions.[11] Such models have been developed using equilibrium thermodynamics to assess the chemical form (i.e., partial pressure) of the fission product species and mass transfer theory to determine their ability for release.[12] In addition, it is important to assess the volatility of the uranium dioxide fuel at high temperature and the incipient melting behaviour of the hyperstoichiometric fuel.[13,14] This type of investigation requires the development of thermodynamic treatments for the uranium-oxygen system, which can be enhanced for multi-component analysis with the addition of fission products and dopant materials.[8,15,16]

Research at the RMC to address such issues on fuel behaviour has been undertaken through a Collaborative Research and Development (CRD) grant in partnership with the Natural Sciences and Engineering Research Council (NSERC) and the CANDU Owner's Group (with experimental support from Atomic Energy of Canada Limited (AECL)). Examples of research projects being conducted at RMC as part of this university/industrial collaboration is discussed in Section 2 and those issues that have arisen as a result of this collaboration are discussed in Section 3.

2. COLLABORATIVE RESEARCH WORK AT RMC

The collaborative projects at RMC involving multi-disciplinary research on defected fuel behaviour include: (i) fuel oxidation kinetics modelling during normal operation and with the possible occurrence of molten fuel during upset conditions; (ii) fission product release for defected fuel including fuel-failure monitoring techniques, tagging methods for defect location and xenon solubility analysis for grab sample monitoring; (iii) thermodynamic analysis to determine phase appearance with fuel burnup, oxidation of Ln-doped fuel and thermochemistry of fuel in contact with the liquid water; and (iv) neutron diffraction studies on U_4O_9 precipitation from UO_{2+x} on cooling.

2.1 Fuel Oxidation

A model has been developed to describe the fuel oxidation kinetics in operating defected elements.[9] This treatment accounts for: (i) steam/hydrogen transport in the fuel-to-sheath gap for estimation of the hydrogen-to-steam partial pressure ratio in the gap; (ii) multi-phase transport including both normal diffusion and thermodiffusion for interstitial oxygen migration in the solid fuel matrix and gas-phase transport of hydrogen/steam in the fuel cracks; (iii) heat conduction in the fuel with feedback for a reduced thermal conductivity in the hyperstoichiometric fuel; and (iv) a thermodynamic treatment for estimation of the equilibrium state of the oxidized fuel (Section 2.3). The coupled governing equations are solved using a finite-element technique with FEMLAB. The model is able to successfully determine both the radial and axial profile of the oxygen-to-uranium ratio in defected fuel, which has been benchmarked against measured oxygen-to-metal profiles obtained at the Chalk River Laboratories with a coulometric titration method for pellet samples taken from spent defected elements (see Figure 1).

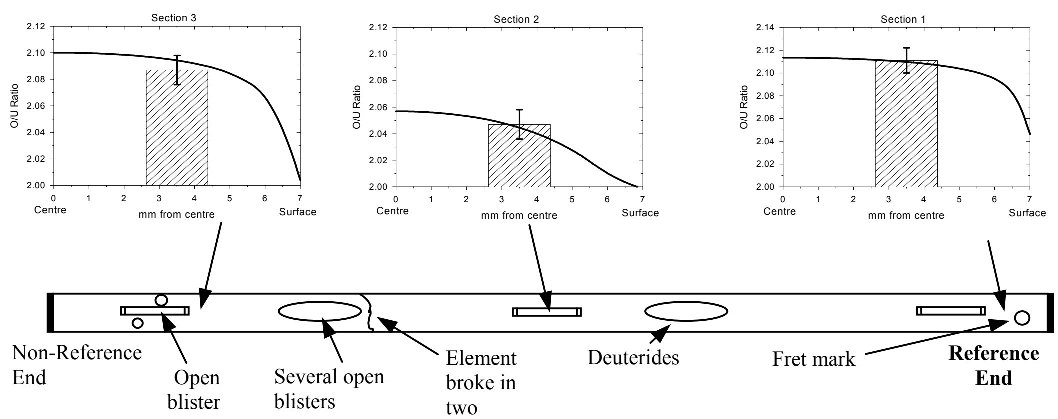

Figure 1. Fuel oxidation model predictions and measured oxygen-to-metal profile for element M86661Z-11.

The model is also being extended to account for fuel containing a molten core. This treatment involves the solution of a Stefan problem for both heat conduction and mass transfer (i.e., oxygen diffusion in the solid and liquid state) as a result of a moving boundary at the discontinuity for the solid/liquid interface.[14] This theoretical analysis further requires as boundary conditions the liquidus and solidus lines for the uranium-oxygen phase diagram, which follow from a thermodynamic analysis (see Section 2.3).

A semi-empirical model to predict the fuel oxidation kinetics is also being developed for possible use as a stand-alone algorithm in fuel performance codes.

2.2 <u>Defected Fuel Behaviour</u>

The Defective Element/Tramp Estimate Characterization Tool (Visual_DETECT) provides a methodology to characterize fuel failures and the amount of uranium contamination based on a coolant activity analysis.[17] The model is able to provide an estimate of the number of fuel failures and defect size (see Figure 2). The Visual_DETECT code could also be interfaced to a gamma-ray spectrometer and commercial nuclide identification software so that this tool could potentially be used in real-time as an on-line "intelligent" defective fuel monitor. This analysis, however, is strictly applicable to steady-state conditions and implicitly assumes that a constant source release from defected fuel and steady-state coolant activity conditions have been achieved.

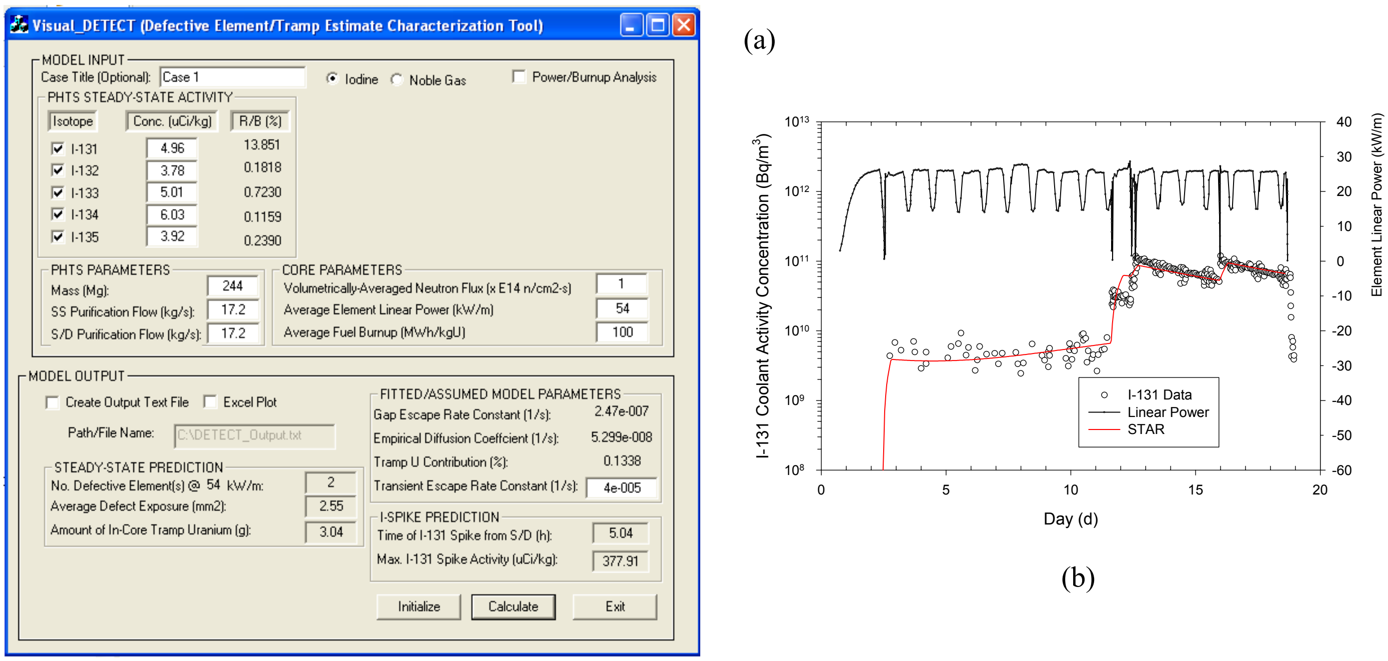

Figure 2. Defected fuel modelling tools. (a) Graphical user interface for Visual_DETECT and (b) comparison between measured and predicted coolant activity concentrations in experiment FFO-110 using STAR.

Action limits for the coolant activity concentrations of I-131 have continually decreased over the years and are currently quite restrictive. With the lack of a delayed-neutron location system (or with the possible degradation of an existing system), it may be difficult to locate and remove defected fuel from the core. With just a few (high-powered) defected elements in core, a transient iodine spike could force the shutdown of a reactor. Once the reactor is shutdown, the identification of these defects is even more difficult to achieve. Hence, it may be useful to distinguish the iodine release that occurs from transient operations (i.e., as an artifact of iodine spiking) versus increased iodine levels that actually result from fuel-element degradation or with the creation of new defects. A time-dependent model has therefore been developed to estimate, in a prognostic fashion, the coolant activity levels as a function of the given reactor power level and purification flow (i.e., to provide an indication of how long the reactor operator has in order to discharge defected fuel before a regulatory action limit is reached). Thus, the Steady-state and Transient Activity Release (STAR) code has been developed for iodine and noble gas analysis to predict the activity in both the fuel-to-sheath gap and primary coolant for defected fuel during steady reactor operation, as well as for transient conditions of reactor shutdown, startup and bundle-shifting operations.[5] Similar to Visual_DETECT, the fission product transport model is based on solid-state diffusion in the fuel matrix and first-order kinetics in the fuel-to-sheath gap. The effects of precursor diffusion and neutron absorption, as well as losses due to radioactive decay and coolant purification (i.e., ion exchange and degassing operations), are considered in the treatment. The code has been benchmarked against well-characterized in-reactor experiments with defected elements conducted in the X-2 defect loop facility at the Chalk River Laboratories (CRL) and several defect occurrences in the commercial reactor.

As previously discussed, it is generally of interest to remove defected fuel as soon as possible. The ability to locate defected fuel, however, is sometimes difficult depending on the given capability at the commercial station. Hence, several novel bundle-tagging methods are being investigated at RMC. The possibility of adding a passive substance of known properties, which could be added to the fuel element during fabrication, so that in the event of failure the "tag" would be released for detection, is being researched. Two possible tagging methods are currently being investigated.[18] The first method involves a nanocrystal tag where, with a careful material and size selection for the "quantum dot", the emitted wavelength from a nanocrystal can be predicted with extreme precision as an individual tag. The second method involves the addition of a specific ratio of noble gas during fabrication, i.e., ^{78}K, ^{80}K, ^{82}K, ^{126}Xe and ^{129}Xe, which could be monitored using mass spectrometry. Current research efforts, however, are trying to address the following issues. A ligand-capping method is required to protect the dot for stability in the hostile environment (i.e., for an aqueous medium at high temperature in the presence of an intense radiation field). It is also not clear, if losses by ion-exchange purification and/or plate out will affect the threshold capability for detection as well as chemical composition changes, which can arise due to neutron transmutation effects. The noble gas tagging method is expected to involve the addition of a ~1 mL (STP) tag as proposed for LWR-type rods; the applicability of this technique to the CANDU fuel bundle design (which has a small free void volume) is not clear.

Several commercial CANDU plants typically employ: (i) on-line gaseous fission product (GFP) and/or (ii) grab sampling techniques for coolant activity monitoring. The grab sampling technique uses an unpressurized sample bomb. Hence, thermodynamic analyses are currently underway to help determine the solubility of xenon with coolant depressurization using solubility data measured at RMC.[19]

2.3 Chemical Thermodynamic Modelling

A generalized thermodynamic model has been advancing in order to determine the equilibrium state of oxidized fuel closely representative of the actual fuel chemistry. This first-principles model takes into consideration all known phases in the U-O binary phase diagram, as well as a treatment for each of the fission products in order to account for the effects of burnup on the oxygen potential in the fuel.[16] The equilibrium thermodynamics model is able to replace previous correlations for calculation of the equilibrium state of the fuel (which do not account for saturation with other oxide phases). Pourbaix diagrams, depicting the ionic equilibria in contact with water, have been further computed for the uranium and oxygen system to investigate possible effects of fuel oxidation during post-irradiation conditions, including spent defective fuel storage in the fuel bays.[20]

Thermodynamic computations are also being undertaken to better understand doped-fuel properties based on the modelling of the trending of Lanthanide Sesquioxides (i.e., Ln_2O_3). In particular, this work is addressing crystal structure polymorphism, mixtures of doped materials with nuclear fuels and fission products, and determination of density and heat capacity for the doped fuel. The modelling is specifically focused on phase-equilibrium modelling of various UO_2-Ln_2O_3 systems in order to develop a likely UO_2–Dy_2O_3 phase diagram to better understand the use of Dy_2O_3 as an additive in advanced CANDU fuel designs.[15] The predicted phase diagram is expected to provide the basis for confirmatory experimental work.

2.4 Neutron Diffraction Studies

A generalized thermodynamic model has been advanced to improve the placement of the UO_{2+x}/U_4O_9 phase boundary, including the development of a non-stoichiometric model for U_4O_{9-y}.[8,15] An invited investigation using neutron diffraction was performed at the Lujan Neutron Scattering Center at the Los Alamos National Laboratory to specifically investigate the precipitation of U_4O_9 from UO_{2+x}. The intent of this opportunity for a collaborative experiment was to validate our thermodynamic model and to shed light on the rate of U_4O_9 appearance in the cooling of oxidized fuel. As shown in Figure 3, this information is obtained using a Rietveld refinement analysis of the raw diffraction data.

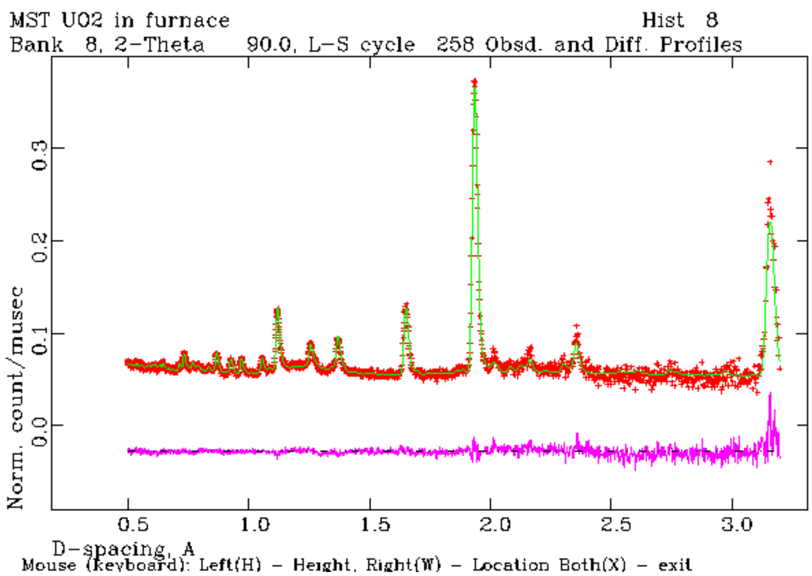

Figure 3. Comparison of the measured count rate (red dots) and predicted Rietveld refinement (green solid line) versus diffraction angle for a mixture of U_4O_9/UO_2 based on collaborative neutron diffraction experiments at LANL.

3. PROMISES AND CONCERNS

In the course of carrying out computational and experimental research in nuclear fuel engineering at the university in partnership with industry (see Section 2), the following issues have been identified. These issues can be categorized as: (i) promises made to both the university and industry; (ii) concerns that have arisen in such a collaboration; and (iii) possible ways to move ahead and particularly improve the collaboration, as well as (iv) the need for the development of a future vision.

(i) The Promises

The "promises" that are possible with a collaborative effort between the university and industry are:

- A synergy of multidisciplinary research among professors *within the university*;

- The sharing of resources between various organizations, including: proprietary data, research techniques, sophisticated equipment, software and personnel;
- Promotion and a greater visibility for the nuclear industry;
- The training of future personnel with a nuclear science and engineering foundation;
- The ability to leverage funding (NSERC-CRD, NSERC Chairs, UNENE, CFI);
- An enhanced safety environment for students and a reassurance to universities in conducting experimental work (i.e., following a safe practice based on experience and knowledge gained in industrial practice);
- Exposure of students to industry professionals;
- The ability to conduct "trial balloon" or exploratory projects at universities for research that could not be easily justified at a national laboratory as they may not be mainstream or have an immediate payback.

(ii) The Concerns

The following items that are of possible concern, which have arisen in setting up a collaborative research effort in nuclear fuel research, are identified below. However, there is reason for optimism as these issues have been or are currently being addressed.

- Access of university personnel to secure laboratory sites (e.g., co-operation with AECL, Cameco, and LANL);
- Ownership of the Intellectual Property derived from the collaborative research. This requires a Memorandum of Understanding between the various organizations. It is realized that perhaps the university research should emphasize the science more than the technology;
- The freedom to publish and present the research work at conferences for peer review, visibility, and to establish a means for further networking with other organizations;
- The importance of developing a multiplicity of independent and competing organizations to carry out research in Canada (with possibly a funding passage through COG for all organizations);
- A need for a quality assurance (QA) policy conceived before university collaboration is undertaken, i.e., it may be possible to have a lower level of QA at the university (e.g., idea stage), which is more appropriate to the university environment;
- There appears to be a declining "cutting-edge" experimental capability in Canada with continued funding cutbacks in the nuclear industry. Unfortunately, there is a tendency for research to be driven more by (immediate) operational requirements rather than for the longer vision of pushing the technological edge via pioneering (and indeed more risky) research;
- The need for administrative support in funding leveraging (e.g., an NSERC funding office at COG).

(iii) Improving the Collaboration

The university/industry collaboration can be improved perhaps through the following activities:

- Workshops in specific research areas;
- The placement of Industrial Fellows at the Universities (honorary);

- Industrial personnel appointed as Adjunct Professors (honorary);
- Industrial secondments as part of graduate studies;
- Sabbatical leave for industrial researchers;
- The establishment of short unfounded project proposals from industry for final-year students (e.g., these projects could include: literature synopsis, databasing, trial balloon tests, simple modeling that could mature into independently pursued industrial projects or into expanded graduate student projects);

(IV) GRANDER VISION OF THE FUTURE IN FUEL ADVANCEMENT

A grander vision is needed to generate and advance new ideas as a partnership between the university and industry:

- The establishment of Industry/University Laboratory Centres of Excellence (in partnership with existing laboratory capability in Canada);
- Stronger university liaison with organizations in the United States and overseas (e.g., LANL, ORNL, ITU);
- A way to promote innovative research in Canada (e.g, the possibility of developing a Canada Foundation for Innovation (CFI) grant application to promote high-temperature material property research on both unirradiated and irradiated fuel material, e.g., to address current licensing requirements with improved operational and safety margins, and to support the improvement and/or development of new fuel designs including the low void reactivity fuel (LVRF) project (e.g., at Bruce Power), Advanced CANDU reactor (e.g., at AECL) and Generation IV reactors.

4.0 CONCLUSIONS

Experimental and theoretical work conducted at RMC is reviewed. This work has focused on fuel oxidation kinetics modelling, the development of fission product release models for fuel-failure monitoring activities, and equilibrium thermodynamic assessments for multi-component systems involving fuel, fission products and burnable poisons.

This university/industry collaboration in computational and experimental research in nuclear fuel engineering has identified: concerns that have arisen in such a collaboration; possible suggestions to enhance and promote the collaboration, and the need for a future vision.

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