13th International Conference on CANDU Fuel
Holiday-Inn Waterfront Hotel
Kingston, Ontario, Canada, 2016August 15-18

CANDU FUEL:
Evolution towards Optimal Performance

Preliminary Modelling of U-Pu MOX Fuel in CANDU Geometries using BISON

A. Prudil*, M. Welland, W. Richmond and S. Yatabe
Canadian Nuclear Laboratories,
286 Plant Rd, Chalk River ON., K0J 1J0,Canada
*Tel. 1-613-584-3311ext.42604, Andrew.Prudil@cnl.ca

ABSTRACT – Preliminary models of Uranium-Plutonium Mixed Oxide (MOX) fuel elements from two experimental bundles irradiated in the National Research Universal (NRU) have been developed in the BISON fuel performance code. The code couples heat transport, thermomechanics, temperature and burnup-dependent material properties, fission product behaviour in the fuel, and cladding deformation within a Finite Element framework. Model predictions compare favourably with post-irradiation measurements of cladding deformation and fission gas release. The fuel was composed of 3.1%-5.3% Pu in depleted uranium and obtained peak linear powers of 41 kWm^{-1} and burnups of 375 $MWh(kgHE)^{-1}$. The samples represent two methods of MOX fabrication, which result in similar Pu homogenization in elements based on the 37-element bundle design.

Introduction

Nuclear fuels composed of mixtures of UO_2 and PuO_2, commonly referred to as Mixed-Oxide fuels, are an important alternative to the more common pure UO_2 fuel designs. MOX fuel cycles enable several important advantages over traditional UO_2 fuel, including:

- Provision of alternative source of fissile material, thereby reducing natural uranium and enrichment requirementsper unit energy produced.
- Consumption of excess plutonium stockpiles (military and civilian), thereby reducing proliferation risks,legacy liabilities, and meeting treaty objectives.
- Diverting waste streams, lowering disposal costs, and lessening environmental impact.

As a result of these benefits, MOX fuel has been of interest to the nuclear industry for many years. In some regions, such as France, MOX fuel has been developed into a mature commercial technology for use in some light water reactor designs[1]. In 2015Russia commissioned a new commercial MOX fuel production facility in order to meet the anticipated growth in MOX usage[2]and is considering an international fuel leasing and recycling program based on a Regenerated Mixture (REMIX) fuel cycle[3]. In the UK, commercial MOX production occurred at the Sellafield site, which is now discontinued, although there is still interest in MOX as a means of addressing the accumulated stockpile of Pu[4]. A MOX fuel plant is under construction at the Savannah River site in the UnitedStatesfor disposing of surplus plutonium, although its future is currently uncertain [5]. There is also MOX interest in China[6] and India[7] as a means of improving uranium utilization and energy self-sufficiency while meeting the growing demand for energy.

The Canadian Deuterium Uranium (CANDU) type Pressurized Heavy Water Reactors (PHWR) are well suited for using alternative/advanced fuel cycles because of the high neutron efficiency and refueling flexibility afforded by on power refueling and modular fuel bundles. PHWR reactors can use MOX fuel manufactured from separated plutonium and natural/recycled uranium, or through advanced fuel schemes such as Direct Use of PWR fuel In CANDU reactors (DUPIC)[8]. Atomic

Energy of Canada's (AECL) development work included manufacturing processes in the Recycle Fuel Fabrication Laboratory (RFFL),irradiation testing in the National Research Experimental reactor (NRX), NRU and Whiteshell Reactor No. 1 (WR-1) reactors, and post-irradiation analysis of the fuels[9].As a continuation of this work, Canadian Nuclear Laboratories (CNL) is developing computer models of these MOX irradiations in order to assess code predictions and identify gaps in knowledge and capability.

1. Background

In this work, computer models were developed representing the Parallex experiment in which ~50 MOX elements,originating from three bundles, were irradiated in the NRU reactor. Two of the three bundles have already undergone Post-Irradiation Examination (PIE) after reaching the target burnup while the thirdbundle requires additional irradiation. The geometry of the bundles is nominally the 37 element design, with the central element removed to accommodate the fuel handling system of the NRU. The outer ring of elements was composed of natural UO_2 doped with dysprosium (Dy) in order to achieve the target linear power in the inner and intermediate rings which contained the MOX fuel.

Bundle AKL was comprised of MOX pellets from USA and Russia and two slightly-enriched uranium 'witness' elements. Bundle AMD included Russian MOX along with MOX from the RFFL. The USA and Russian fuel pellets were produced at Los Alamos National Laboratory and Bochvar Institute from surplus weapons grade plutonium (WPu) in depleted uranium (dU) respectively, in order to demonstrate technical feasibility of WPu dispositioning in PHWRs. The pellets produced in the RFFL utilized reactor grade, civilian plutonium(CivPu)also in dU, and were therefore prepared with higherPu concentrations in order to achieve similar concentrations of fissile isotopes. Overall, the Pu concentrations ranged from 3.1%-5.3%.

Different methods of mixing U and Pu during fuel fabrication produce different degrees of compositional homogeneity within the fuel, which may impact fuel performance. All three techniques began with PuO_2 powder. The Bochvar process directly combines the PuO_2 powder with UO_2 powder to the desired proportions and mills them using electromechanically-oscillated steel needles. The LANL fabrication technique involved forming a mastermix of 10 wt% WPu in UO_2which was ball milled together, and then combined with pure UO_2 to the desired U-Pu proportions before ball milling again. The technique used in the RFFL was to simulate the Micronized Mastermix (MIMAS) technique used commercially by Belgonucleaire by creating a mastermix of 30% Pu in UO_2, which is ball milled, and then combined with UO_2 to the desired proportions and mixed mechanically. The Bochvar and LANL techniques resulted in a comparable degree of homogeneity of the Pu in the U matrix, whereas the RFFL techniques produced islands of Pu enrichment. A more detailed overview of the experimental aspects of the Parallax experiment and a summary of the post-irradiation results obtained thus far were published at previous CANDU fuel conferences by Dimayuga et al.[10]–[12].

The linear power of the fuel elements in each ring of a bundle decreases towards the center due to thermal neutron flux depression. As a result, the elements in the same ring and with the same composition are assumed to experience the same power history (neglecting flux tilt across the bundle). This introduces a 1% or less error in linear power according to results of the reactor physics code, BURFEL-PC, which calculates the power history in individual elementsbasedon neutron transport and burnup calculations normalized toneutron flux and coolant temperature measurements[13]. This results in nine combinations of power history and compositions for the two irradiated bundles:two bundles, with two fuel compositions each and tworings containing MOX per bundle, and a single set of UO_2witness fuel elements. Data from two of the MOX cases and the witness elements has not been released, leaving six cases for comparison. A representative power history from bundle AMD is shown in Figure 1 for the top, middle, and lower axial locations of the fuel bundle. Midway through the irradiation, the position of the fuel is changed and fuel experiences a significant gradient in linear power in the axial direction due to the axial flux profile of the reactor. This causes the power in the three locations to diverge.

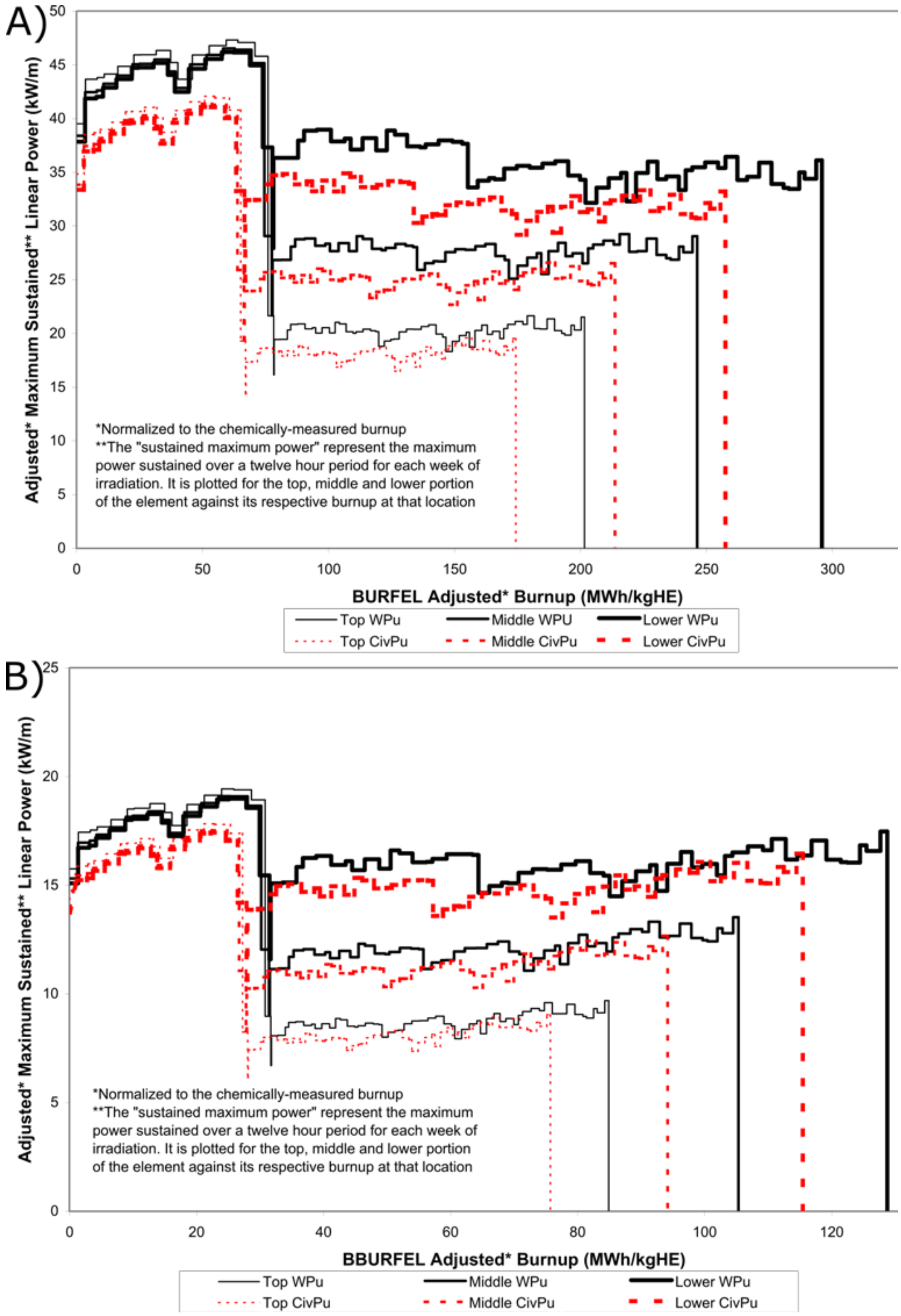

Figure 1 Power histories from bundle AMD, containing RFFL elements composed of civilian plutonium (CivPu), and Bochvar elements containing weapons grade plutonium (WPu) placed in a) the intermediate ring and b) the inner ring divided into top, middle and lower segments[12]**.**

2. Model Description

The elements were modelled using the BISON fuel performance code developed at the Idaho National Laboratory[14]. It utilizes the Multiphysics Object-Oriented Simulation Environment (MOOSE) framework [15]to obtain finite-element approximations to coupled, non-linear ordinary and partial differential equations representing phenomena occurring within nuclear fuel. The MOOSE framework is capable of modelling 1D, 2D, and 3D geometries on small laptops/workstations and large high performance computers with distributed memory and thousands of cores. BISON specifically incorporates coupled heat-transfer and solid-mechanics from MOOSE along withnuclear-specific phenomena such as burnup, fission gas release, fission product swelling and densification.At the time of this paper was being written,MOOSE/BISON was being developed as an advanced R&D tool. Although it was being verified numerically[16] and validated/benchmarked against experiments[17]–[19], it was not licensed at the time by nuclear regulators for safety analysis. BISON has primarily been developed for UO_2 fuel but it also includes some models for other fuel types such as MOX, TRISO, and plate fuel[20].

A finite-element mesh, as shown in Figure 2, representing a full length fuel element (>34 pellets) was produced with each pellet having appropriate dishing and chamfers. The pellets were connected together at adjacent lands where adjacent pellets come into contact to form a solid fuel pellet stack in order to avoid the computational cost of numerous pellet-to-pellet contact pairs and the complexities associated with free body motion. Similarly, the landat the bottom of the pellet stack was fixed in the axial direction in order to approximate gravity and prevent free movement of the stack while still allowing it to expand vertically such that it may contact the top end-cap (depending on linear power, initial axial gap, and burnup). Mechanical contact between pellets and cladding was enforced by the 'penalty method', with a penalty factor coefficient of 10^8. Heattransfer from the pellet stack to the sheath and endcaps was modelled using the thermal contact (1D gap heat-transport) feature in BISON, accounting for the dynamic fuel-to-sheath gap size, internal gas composition, and contact pressure. The volumetric heat generation was defined using the TUBRNP [21]modelin BISON accounting for isotopes ^{235}U, ^{238}U and ^{239}Pu-^{242}Pu.

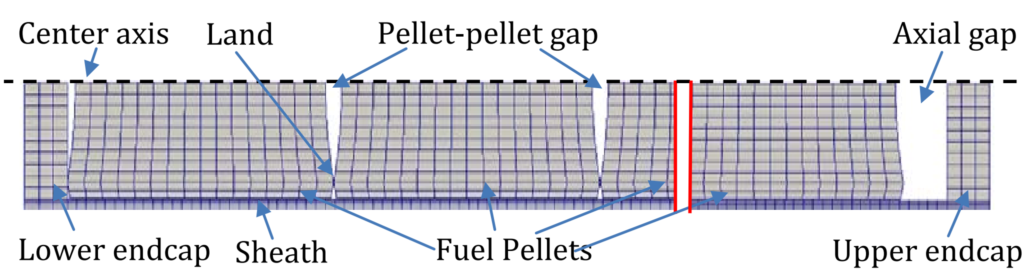

Figure 2Sample finite element mesh used for simulation with labels indicating included geometric features.

The pellets were modelled elastically (no creep/plastic deformation) while the cladding (sheath) included elastic and plastic deformation and creep. Simulations were performed with and without the 'fuel relocation'module in order to assessits impact, since fuel relocation can behave differently with collapsible cladding compared to freestanding light water reactor cladding. The existing BISON material models were used for the pellets and cladding in order to assess their applicability for modelling PHWR MOX. The thermal conductivity of the MOX (including Pu) was modelled with the Duriez-Lucuta model recommended by Carbajo et al.[22]. The remaining material properties such as Young's modulus, Poisson's ratio, heat capacity, etc. utilized UO_2 correlations available in BISON without modification as a simplifying approximation.

Fission gas behavior was modelled with the Simplified Integrated Fission Gas Release and Swelling (SIFGRS) model, which attempts to predict the size and area density of fission gas bubbles on the grain boundary in order to predict fuel swelling and gas release in a unified model[23]. The SIFGRS model was used without modifications to account for the presence of additional Pu or variation in microstructure other than average grain size.

3. Simulation Results and Comparison

3.1 Profilometry

During post irradiation examination (PIE),the bundles were disassembled into individual fuel elements to be examined. The element diameter along the length of each element was measured at three equally spaced angles, which determined the effective diameter accounting for ovality and azimuthal (hoop) strain. A comparison of the end-of-life diameter profiles is presented in Figures 3, 4, and 5 with thin colored lines representing the measured values and the thick black and grey lines the simulation results. The BISON results with the relocation model active are labeled"BISON-relocation" while the results without relocation are labeled "BISON-elastic". As previously mentioned, each bundle contained multiple elements with the same nominal design parameters (geometry, fuel composition) and irradiation history. The profilometry of each element in these sets is plotted to provide an indication of the variability of fuel performance. Note that the measurements in the vicinity of the element ends are believed to be influenced by the clamps holding the elements in the profilometryapparatus. Similarly, element diameter measurements at the fuel element spacer locations lie outside the calibration region producing a sharp spike in the vacinity of 240 mmwhich should also be ignored.

A number of interesting features of these diameter/strain profiles can be compared: the average deformation, the pellet-to-pellet ridge heights, and the trend along the axial length of the element. The average cladding deformation is in reasonable agreement with the measurements for most of the six observed cases, with some cases slightly under- or overpredicting, but not strongly biased in either direction. A large variation in the ridge height is predicted by the model, with the inner elements having small ridges compared to the intermediate elements. The direction of this trend is consistent with the measured profilometry, however, qualitatively the magnitude of the ridges appears to be underpredicted for all of the inner ring cases as well as the Russian MOX in the intermediate ring of bundle AMD.

The predicted trends in the axial direction are a result of the axial linear power profile calculated from the BURFEL code (all other boundary conditions are axially independent). Qualitatively, the trend for bundle AKL Russian and USA (Figures 3 and 4) is in reasonable agreement (decreasing clad diameter away from the reference end).However, for bundle AMD (Figure 5), BURFEL predicts a decreasing linear power along the element length, which is consistent with the trend of decreasing diameter observed in the inner ring of elements, but the intermediate ring shows a slightly increasing trend. The reason for this observed difference is being investigated, but it is thought that it may stem from the temperature and burnup dependence of densification and swelling, or the axial dependence of the coolant temperature along the element length.

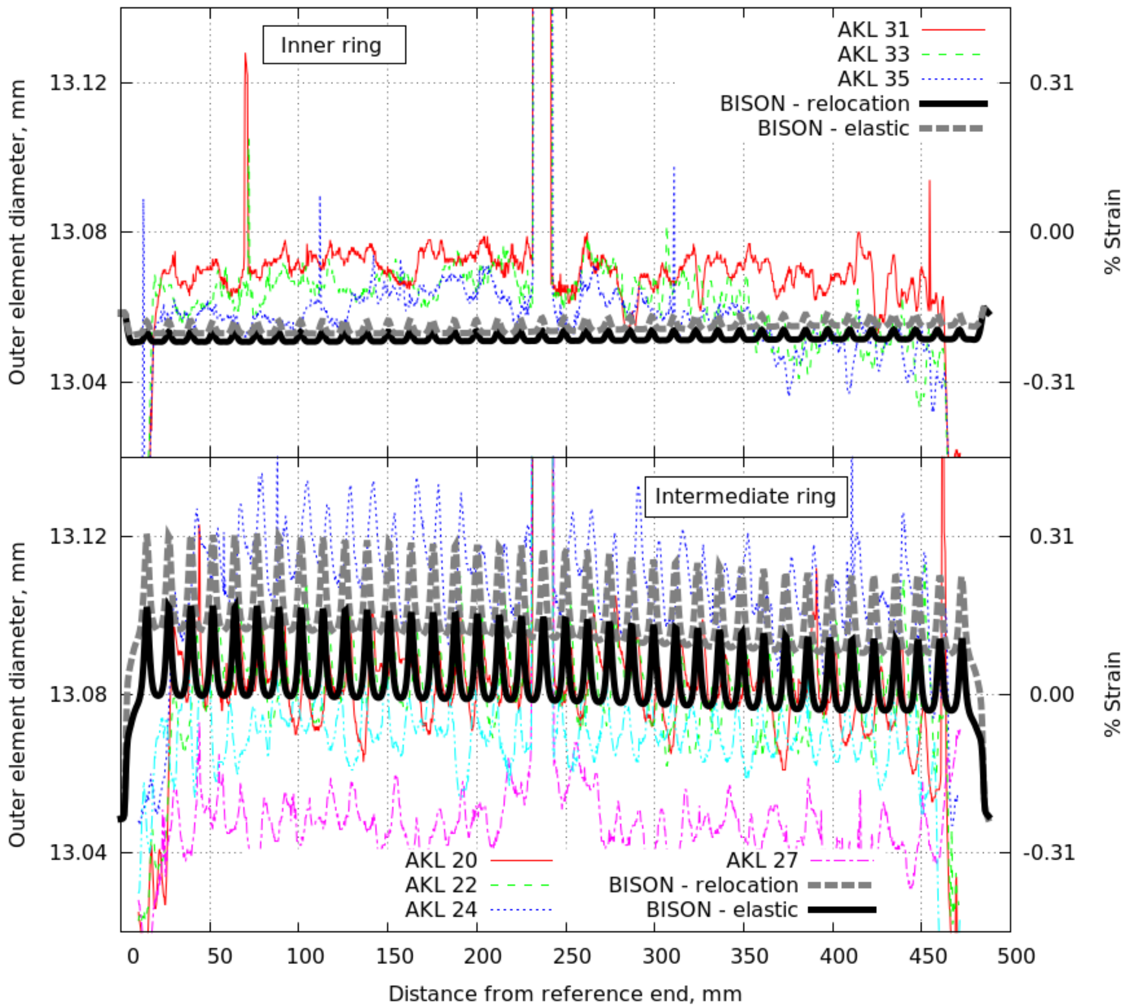

Figure 3Predicted end-of-life cladding diameter (left axis) and strain (right axis), for inner and intermediate ring elements, compared to PIE measurements for Russian manufactured MOX elements of bundle AKL.

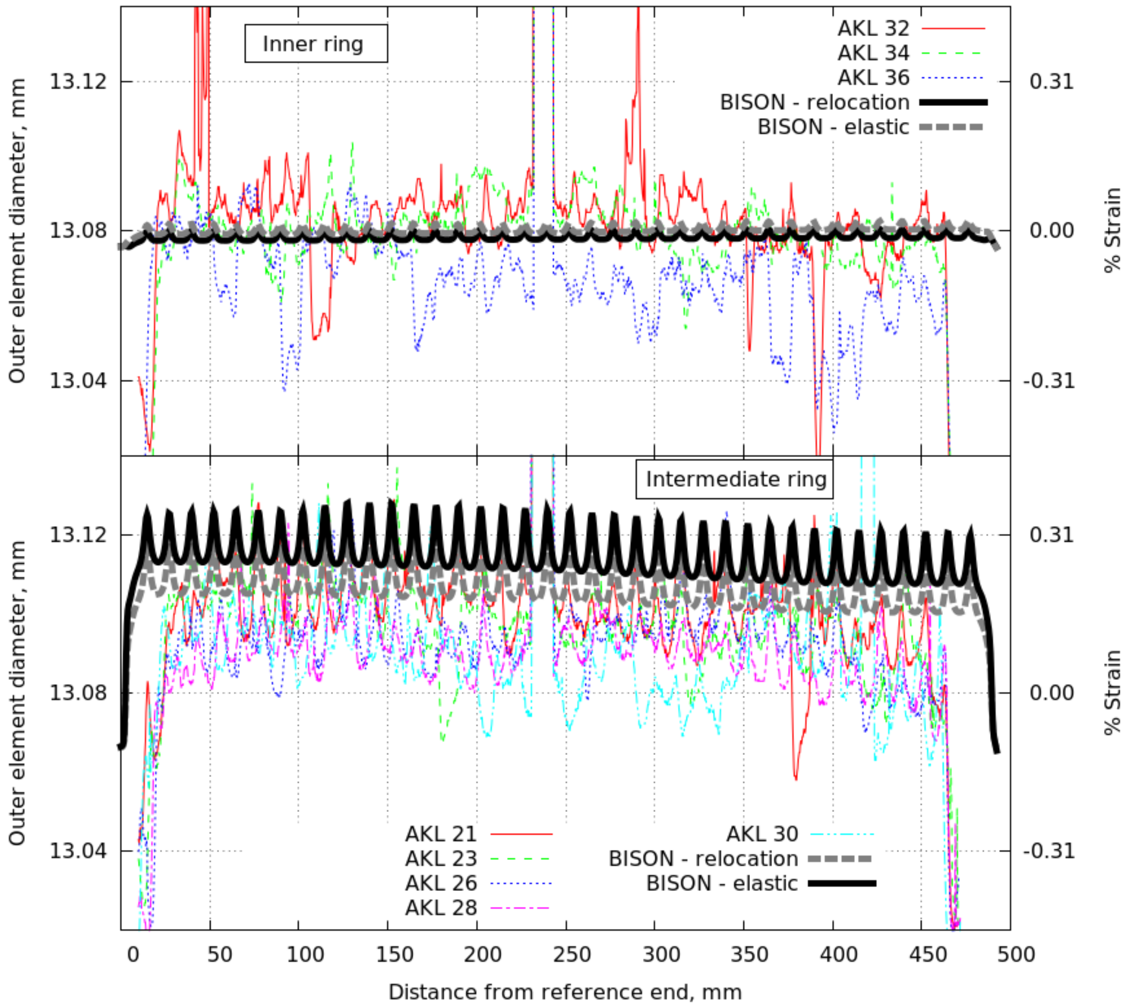

Figure 4Predicted end-of-life cladding diameter (left axis) and strain (right axis), for inner and intermediate ring elements, compared to PIE measurements for USA manufactured MOX elements of bundle AKL.

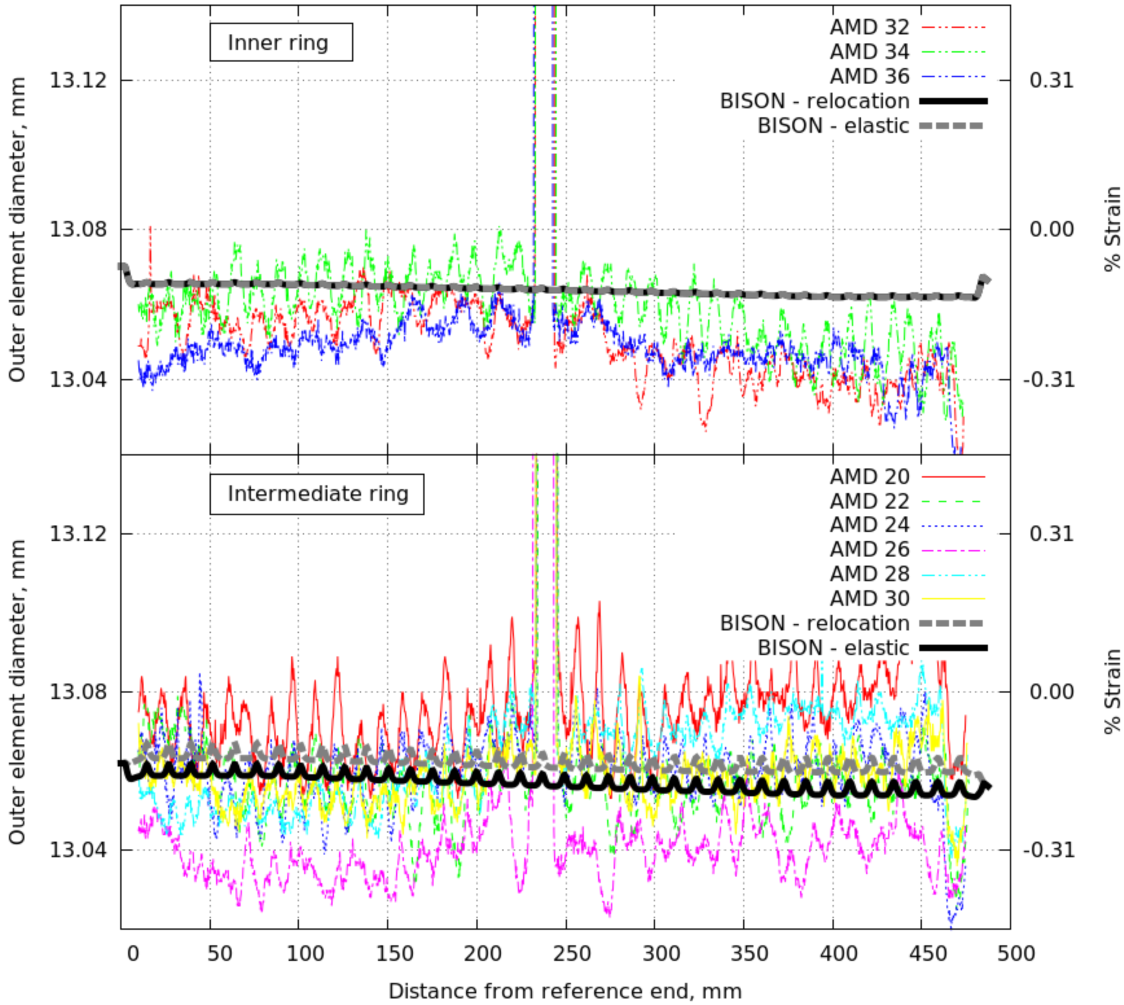

Figure 5Predicted end-of-life cladding diameter (left axis) and strain (right axis), for inner and intermediate ring elements, compared to PIE measurements for Russian manufactured MOX elements of bundle AMD.

3.2 Fission Gas Release

The comparison of predicted and measured fission gas release is presented in Figure 6in terms of gas volume (a) and fraction of fission gas released (b). The BISON models with the elastic pellets (no relocation) predict slighly higher fission gas release (approximately 0.5 to 5%) compared to the models with relocation. This is consistent with the behavior of the relocation models for LWR fuel in which relocation reduces the pellet-to-cladding gap, thereby improving gap heat transfer and reducing pellet temperatures.

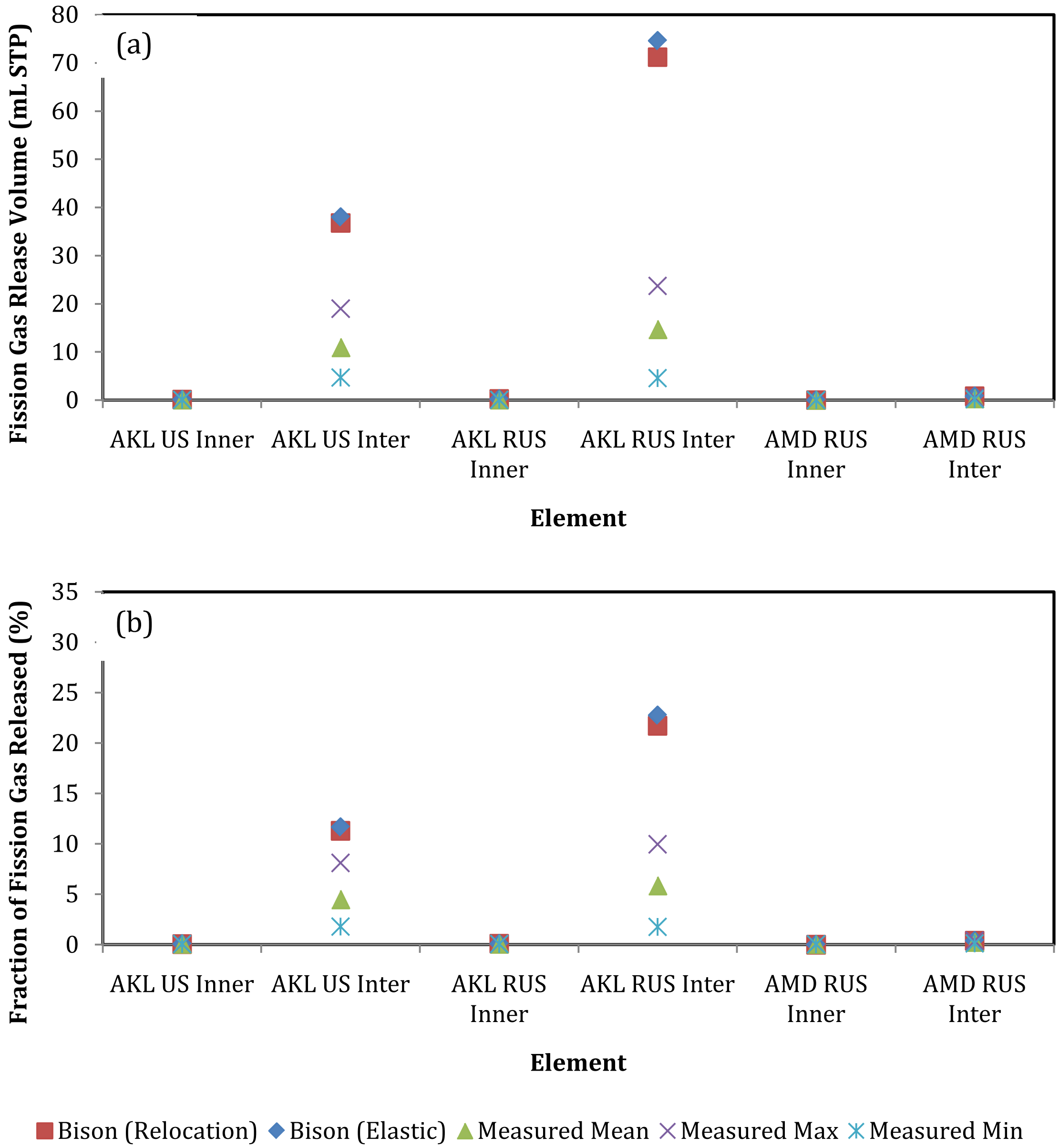

Figure 6Comparisons of predicted (BISON) vs measured fission gas released in terms of gas volume (a) and fraction of fission gas released (b).

As expected, the intermediate elements are predicted to release more fission gas than the inner elements as a result of the higher linear power and burnup. For bundle AMD, the linear power and burnup are sufficiently low that very little gas was released from the Russian fuel. The only two cases with significant fission gas release are from the intermediate ring of the AKL bundle. In both of these cases, it was found that the BISON results overpredicted the measured results by a factor of ~3-5 times. However, discrepancies of this magnitude are not unexpected for fission gas release predictions due to the uncertainty in the microstructural data, such as distribution of grain sizes, the positive feedback effects, as well as the scatter in the measured data (which is roughly a factor of 3 between lowest and highest release, despite being nominally the same irradiation case).

The USA and Russian MOX pellets show similar fission gas retention despite the different manufacturing techniques. Differences and discrepancies between the predictions and the experimental values can be explained by differences in the initial microstructure and the presence of regions of high Pu content. During irradiation, the Pu rich areas have a much higher local fission rate than the bulk material. This leads to the formation of High Burnup Areas (HBA) where the local burnup and accumulation of fission products is greater than the element average. Fission gas diffusing from the grains causes the same intergranular bubble formation, swelling and gas release as observed in UO_2 but is locally accelerated due to the elevated fission rate. This implies that the RFFL fuel may show significantly different behaviour due to the heterogeneity of the fuel, however the data for this fuel was unavailable as of this document.

Any inhomogeneous distribution of fissile isotopes is not accounted for in the SIFGRS model which was developed for UO_2, which usually has a high degree of homogeneity. In addition, fission gas released to the gap usually degrades the thermal conductivity of the gap, causing higher temperatures and more gas release in a positive feedback fashion. It may be possible to modify the SIFGRS model to account for microstructural heterogeneity either empirically, by modifying the effective yields and saturation conditions to match observed releases for heterogeneous fuel, or by more sophisticated means such as solving two variants of the SIFGRS model, one for the bulk material and one for the Pu rich fraction.

4. Conclusions

BISON models of the Parallex experiment have been developed and run. Comparisons of measured and predicted fuel cladding deformation, viz the end-of-life diameter and hoop-strain, and fission gas release have been presented. The cladding deformation was found to be in reasonable agreement with the measured values, both in terms of the average diametral strain as well as the pellet-to-pellet ridge heights. The cases modelled without the relocation model predicted smaller end-of-life diameters and less pellet-to-sheath contact. The fission gas release predictions followed similar trends as the measurements, although in the all three of the cases, with significant gas release, the magnitude of the release was overestimated. Modification of the SIFGRS model in BISON to account for the additional plutonium in MOX as well as the degree of homogeneity may further improve fission gas release predictions.

5. Awknowledgements

Funding for this work was provided by CNL's project on High-Fidelity Modeling. The authors would also like to acknowledge our coleagues at INL for allowing us to use the BISON code and providing technical support. This research made use of the resources of the High Performance Computing Center atIdaho National Laboratory, which is supported by the Office of Nuclear Energy of the U.S.Department of Energy under Contract No. DEAC0705ID14517

6. References

[1] AREVA, “Fabrication of recycled fuel,” *The fabrication of MOX fuel: recycled nuclear power*. [Online]. Available: http://www.areva.com/EN/operations-1173/the-fabrication-of-mox-fuel-recycled-nuclear-power.html. [Accessed: 26-May-2016].

[2] World Nuclear News, “Russia charts progress with new MOX fuel development,” *World Nuclear News Website*, *World Nuclear Association*, London, UK, 11-Sep-2015.

[3] Mikhail Baryshnikov, “REMIX Nuclear Fuel Cycle,” presented at the World Nuclear Fuel Cycle Conference, Abu Dhabi, UAE, 2016.

[4] World Nuclear Association, “Mixed Oxide (MOX) Fuel,” 21-Apr-2016. .

[5] S. Mufson, “Obama plan to de-fund Savannah River plutonium conversion plant draws fire,” *The Washington Post*, Washtington, D.C., USA, 25-Feb-2016.

[6] Z. Zhang, M. Chen, P. Zhou, Q. Li, Z. Meng, and G. Zhang, “CANDU Position and Prospect in Chinese Nuclear Fuel Cycle,” in *2013 21st International Conference on Nuclear Engineering*, Chengdu, China, 2013, pp. V001T02A046–V001T02A046.

[7] P. K. Dey and N. K. Bansal, “Spent fuel reprocessing: A vital link in Indian nuclear power program,” *Indias React. Past Present Future*, vol. 236, no. 7–8, pp. 723–729, Apr. 2006.

[8] H. Keil, P. Boczar, and H. S. Park, “Options for the direct use of spent PWR fuel in CANDU (DUPIC),” presented at the Third international conference on CANDU fuel, 1992, p. 597.

[9] F. C. Dimayuga, M. R. Floyd, and D. S. Cox, “Fabricatoin and performance testing of CANDU mixed-oxide fuel,” Atomic Energy of Canada Limited, Chalk River Nuclear Laboratory, International symposium on MOX fuel cycle technologies for medium and long term deployment IAEA-SM-358/22, May 1999.

[10] F. C. Dimayuga, “The Parallex Project: irradiation testing and pie of the first bundle,” presented at the The 8th international conference on CANDU fuel, Toronto, Ontario, 2003, pp. 126–136.

[11] F. C. Dimayuga, J. Montin, and P. Valliant, “PIE of the first parallex bundle,” presented at the The 9th international conference on CANDU fuel, 2005, pp. 1–9.

[12] F. C. Dimayuga, M. Karam, and J. Montin, “Post-irradiation examination of CANDU MOX fuel bundle containing weapons grade plutonium,” presented at the The 10th international conference on CANDU fuel, Ottawa, Ontario, 2008.

[13] R. Davis, “Simplification of coding of NRU loop experiment software with dimensional generator,” in *PHYSOR 2006: Advances in Nuclear Analysis and Simulation*, Vancouver, BC, Canada, 2006.

[14] Fuel Modeling and Simulation Department, *BISON: A Finite Element-Based Nuclear Fuel Performance Code*. Idaho Falls, Idaho: Idaho National Lab.

[15] D. Gaston, C. Newman, G. Hansen, and D. Lebrun-Grandié, “MOOSE: A parallel computational framework for coupled systems of nonlinear equations,” *Nucl. Eng. Des.*, vol. 239, no. 10, pp. 1768–1778, Oct. 2009.

[16] J. D. Hales, S. R. Novascone, B. W. Spencer, R. L. Williamson, G. Pastore, and D. M. Perez, “Verification of the BISON fuel performance code,” *Ann. Nucl. Energy*, vol. 71, no. 0, pp. 81–90, Sep. 2014.

[17] R. L. Williamson and S. R. Novascone, “Application of the BISON Fuel Performance Code to the FUMEX-III Coordinated Research Project,” Idaho National Laboratory, Idaho Falls, Idaho, United States, INL/EXT-12-25530, Apr. 2012.

[18] D. M. Perez, R. L. Williamson, S. R. Novascone, T. K. Larson, J. D. Hales, B. W. Spencer, and G. Pastore, “An evaluation of the nuclear fuel performance code BISON,” presented at the Mathematics & Computation 2013, Sun Valley, ID, USA, 2013.

[19] G. Pastore, J. D. Hales, S. R. Novascone, D. M. Perez, B. W. Spencer, and R. L. Williamson, “Analysis of fission gas release in LWR fuel using the BISON code,” presented at the LWR Fuel Performance Meeting/ Top Fuel 2013, Charlotte, NC, USA, 2013.

[20] J. D. Hales, S. R. Novascone, G. Pastore, D. M. Perez, B. W. Spencer, and R. L. Williamson, “BISON theory manual.” Fuels Modeling and Simulation Department Idaho National Laboratory, Oct-2013.

[21] K. Lassmann, C. T. Walker, and J. van de Laar, “Extension of the TRANSURANUS burnup model to heavy water reactor conditions,” *J. Nucl. Mater.*, vol. 255, no. 2–3, pp. 222–233, Jun. 1998.

[22] J. J. Carbajo, G. L. Yoder, S. G. Popov, and V. K. Ivanov, “A review of the thermophysical properties of MOX and UO2 fuels,” *J. Nucl. Mater.*, vol. 299, no. 3, pp. 181–198, Dec. 2001.

[23] G. Pastore, L. Luzzi, V. Di Marcello, and P. Van Uffelen, “Physics-based modelling of fission gas swelling and release in UO2 applied to integral fuel rod analysis,” *Nucl. Eng. Des.*, vol. 256, pp. 75–86, Mar. 2013.