BISON for Metallic Fuels Modelling

C.Matthews[1], J.Galloway[1], C.Unal[1], S.Novascone[2], R.Williamson[2]

[1]Los Alamos National Laboratory, Los Alamos, NM, USA

[2]Idaho National Laboratory, Idaho Falls, ID, USA

E-mail contact of main author: cmatthews@lanl.gov

Abstract. The fuel performance code BISON has recently been extended to simulate U-Zr and U-Pu-Zr metallic fuel rods irradiated in the US sodium cooled fast reactor EBR-II. By introducing fuel and clad specific material models, the backbone provided by the MOOSE/BISON software architecture has allowed rapid development of metallic fuel capabilities. Zirconium based fuels present unique challenges due to the different phases that exist at irradiation temperatures. Each phase possess differing thermo-mechanical properties, necessitating explicit tracking of the relative concentration of phases throughout the fuel rod in order to capture the integral behavior. In addition, the transition locations between phases change over the course of irradiation due to zirconium diffusion in the fuel, necessitating coupling of the thermo-mechanical simulation to the Fickian and Soret diffusion of Zr. Along with a robust U-Zr and U-Pu-Zr zirconium redistribution model, coupling of zirconium concentration to local power deposition allows for several coupled thermo-diffusion simulations to be compared based on different fabrication and operating conditions. Through the inclusion of flexible models into the BISON framework, core designers can utilize fuel performance simulations as an additional tool to support irradiation experiments and push for novel fuel designs.

Key Words: U-Pu-Zr, BISON, fuel performance modeling.

1. Introduction

Nuclear engineering is reaching a domain space in which computational power and methods are starting to surpass the models that are used to simulate nuclear fuel rods. As these fuel models are updated, simulations utilizing different compositions, operating conditions, fabrication techniques, and geometries can provide core designers with an important first step towards fuel qualification. Due to the cost associated with fuel irradiations, it can be expected that time and money spent building better fuel models may provide an attractive return during initial studies of advanced fuels, especially with regards to fuels beyond the extensively studied uranium oxide system.

In general, the primary U.S. experience with irradiated fuel rods beyond traditional oxide fuels is with binary (U-Zr) and ternary (U-Pu-Zr) metal fuels. The bulk of the experimental data for metal fuels is obtained from rods irradiated in the EBR-II sodium fast reactor (SFR), thus many of the correlations developed for metal fuels comes from the post-irradiation (PIE) examination of EBR-II rods.

One of the unique characteristics that set U-Pu-Zr fuels apart from traditional oxide fuels is the presence of different phases that develop in the fuel during irradiation. Each of these different phases has unique thermal, mechanical, and diffusive behavior. The transitions between these phases are not static as the initially homogenous zirconium diffuses towards the center and edge of the fuel rod, resulting in a low-Zr, high-U ring (FIG. 1). Along with the differing material properties that occur in each phase, the increase in uranium corresponds to an increase in localized fission and power production. The direct coupling between the thermal and diffusion solves highlights one of the many examples of tightly coupled phenomenon that are present in nuclear fuel, and provide motivation for the development of codes that simultaneously solve each piece of physics in a fully-coupled manner.

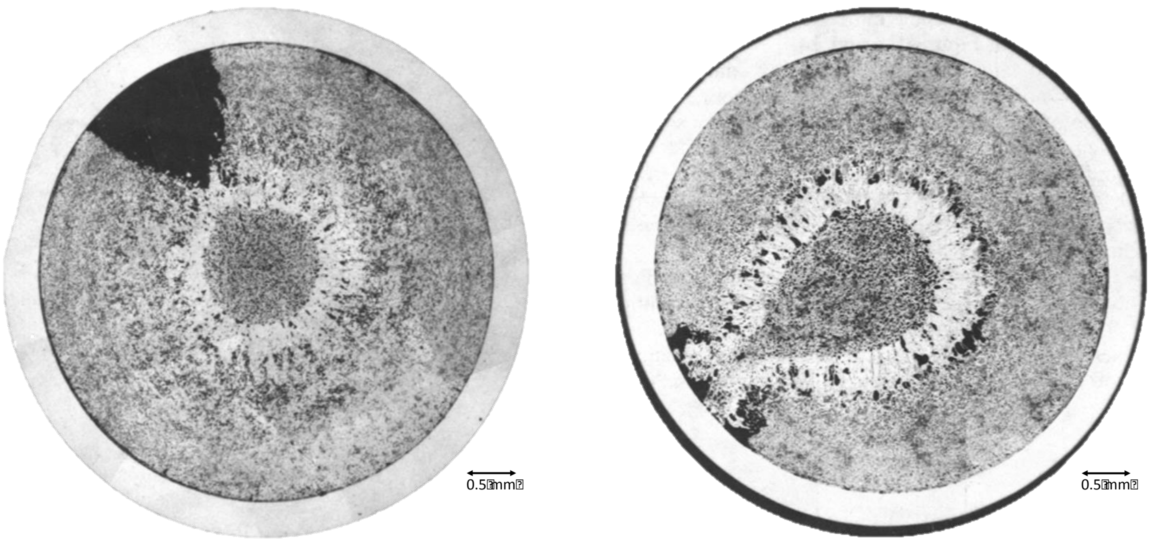

FIG. 1. Typical micrographs of irradiated ternary fuel rods showing the different phases, identified here by the local density of the material. Also visible is the fresh (left) and healed (right) cracks that occurs during irradiation [1].

Along with the complications created by the constantly evolving phase topography in the U-Pu-Zr fuel, recent studies on zirconium diffusion in ternary fuels have shown that the phase diagram may be different under irradiation when compared to fresh fuel equilibrium measurements. Specifically, the temperature transition lines that are measured in out-of-pile experimental setups and used to develop the equilibrium phase-diagrams that are essential towards understanding the fuel are likely 50 – 100 K higher than reported. While the actual temperatures have been estimated through the zirconium redistribution modelling, the consequences are not confined only to diffusion calculations. In general, thermo-mechanical properties such as thermal conductivity and elastic moduli are measured out-of-pile at different temperatures such that a property vs. temperature correlation can be developed. In many cases, these properties are measured across temperature transition regions and are able to roughly indicate phase-transition temperatures. Unfortunately, the region of those phase-transitions may be at much higher temperatures in-reactor, rendering temperature based property calculations inaccurate.

In an effort to separate material properties from the in-reaction temperature transitions, advanced U-Pu-Zr simulations need to use phase-dependent material properties in order to capture the fundamental state variables of the fuel such as temperature and stress. These formulations may benefit from thorough re-examination of past experimental data, but in general must be separated by phase, as well as temperature.

Coupled with zirconium redistribution and local phase determination, these new properties will provide researchers the freedom to explore beyond the narrow parametric window of measured data. By reformulating properties as phase-dependent, novel fuel designs can be explored with a greater degree of confidence.

In an effort to show some of the strengths of the modular structure of BISON, a typical U-19Pu-10Zr fuel rod has been simulated according to the best available operating data.

Comparing against this baseline, changes in the composition and the operating conditions may provide insight into future design considerations.

2. Models

New models to simulate U-Pu-Zr fuel have been implemented in BISON [2]. Much of the thermo-mechanical physics is provided by the MOOSE/BISON backbone, including heat conduction, volumetric swelling, and contact, and has been described elsewhere [2,3]. Some of the unique material models utilized to describe the characteristics of U-Pu-Zr fuel will be briefly included here. In addition, unique physics such as zirconium redistribution and phase-dependent properties have been added to BISON, and will be discussed.

Several groups have studied zirconium redistribution in the past [4-8], but only recently have new values been calculated with the coupled thermal-diffusion physics solved simultaneously [4]. In general, the zirconium diffusion process can be described through the combination of both thermal and temperature gradient driven diffusion, resulting in a transport equation of the form,

$$\frac{\partial X_{Zr}}{\partial t} = \nabla \cdot D\left(X_{Zr}, T\right) \nabla X_{Zr} + \nabla \cdot S\left(X_{Zr}, T\right) \nabla T, \qquad (1)$$

where the Fickian (D) and Soret (S) diffusion coefficients are phase dependent. The typical bathtub shape of the zirconium concentration, exemplified in FIG. 2 results from a change in sign of the Soret diffusion coefficient, pushing the zirconium to the center of the rod in the gamma region, and towards the edge of the rod in the alpha/beta/delta regions.

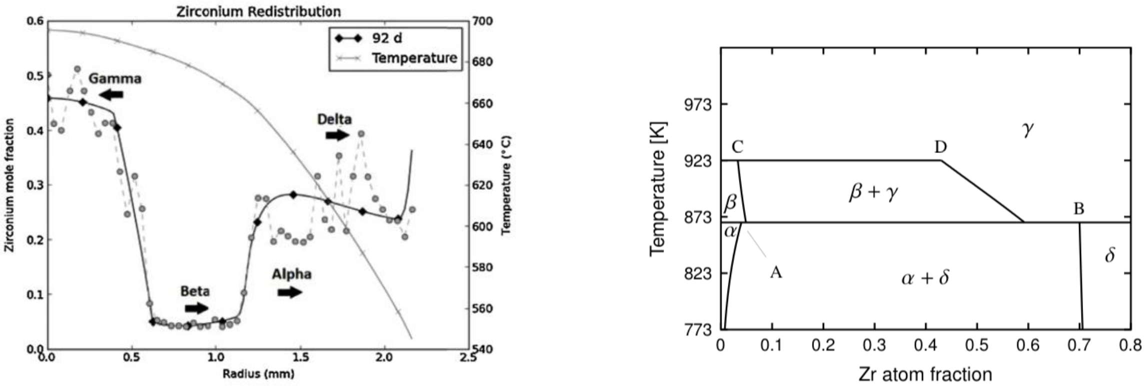

FIG. 2. Typical zirconium redistribution compared to experimental data (left) [4], and a typical simplified phase diagram used in determining phase distribution for zirconium redistribution simulations (right) [8].

In general, plutonium has been observed to remain relatively stationary during irradiation. Consequently, the moving zirconium is replaced by uranium, resulting in a high uranium ring in the fuel. Consequently, the temperature profile deviates from a smooth arc, as displayed in FIG 2. Using representative MCNP runs, a linear fit between power (P) vs. zirconium atom fraction has been implemented to modify the local heat deposition rate during irradiation [4], calculated by,

$$\dot{q}\left(r, z, X_{Zr}\right) = \dot{q}_l \cdot L \cdot a(z) \cdot P\left(X_{Zr}\right), \qquad (2)$$

where q_l is the linear heat generation rate given by operational data, L is the height of the rod, $a(z)$ is the axial offset given as a function of height.

Due to the importance of phase dependent properties, accurate determination of the phase is essential. This requires providing each property model the fraction of each of the four phases at all locations, with appropriate mixing strategies in the two-phase regions. In addition, simulations become difficult to solve numerically when sharp transitions occur. As a result, a smoothing method in both the temperature and X_{Zr} direction has been implemented to artificially prevent sharp transitions during simulations. The details of the smoothing process can be found in previous publications [8]. In reality, any phase diagram represents the completely equilibrated system. In other words, the fuel will not present sharp phase boundaries, thus providing some physical justification to the smoothing schemes utilized.

The temperature boundary condition on the exterior of the cladding is calculated using a traditional convective heat transfer model utilizing operation sodium flow data [3], and the heat flow between the fuel/cladding gap is calculated using a typical gap model [9]. The remaining material models utilized in the simulations that follow are of the traditional property vs. temperature formulation for simplicity. The details of the properties can be found elsewhere [3].

3. Simulations

The fuel rod T179 is a typical U-19Pu-10Zr fuel rod irradiated in EBR-II in 1985. The rod is of interest due to the PIE results that provided micrographs at several locations, as well as microprobe examinations near the top of the fuel. As a result, it has been utilized in previous studies for calibration of zirconium diffusion coefficients [3-8]. The rod was simulated using the best available operating conditions, and provides the baseline case for which variations in composition or initial conditions can be modified. Although the assembly containing the T179 rod experienced slightly different flow rates and power ratings during the course of irradiation, the changes were slight enough that a daily weighted linear heat generation and flow rate were utilized in the simulations.

Two cases were run to compare with the baseline T179 simulation. The “Axial shift” case utilizes an axial fission rate distribution that mimics the behavior that may be expected in advanced fast reactor concepts being proposed for power generation, in which the power profile shifts from the bottom of the core to the top [10]. For this case, the peak fission rate location will shift from the lower section of the rod to the upper, as displayed in FIG 3. In addition, the integral rod power will also shift from high to low from beginning of life (BOL) to end of life (EOL). At the middle of life (MOL), the power profile will exactly match the baseline T179 case.

The second case represents changing the fabrication of the fuel rod in an effort to address operational concerns. One of the primary apprehensions with ternary metal fuel is the existence of a low temperature Pu-Fe eutectic that may form between the fuel and cladding. In addition, fuel/cladding chemical interaction (FCCI) has been observed to occur at the locations of highest fuel temperatures. This is due to enhanced presence of lanthanides near the top third of the rod due to higher diffusivity, as well as more prevalent cracking that provides pathways for the lanthanides to diffuse to the fuel. In general, plutonium fuels experience greater cracking, thus limiting the plutonium content in the top third of the fuel may reduce the consequences of FCCI, both through preventing the Pu-Fe eutectic, which has been shown to accelerate FCCI [11], and by reducing the source term of lanthanide on the fuel. By linearly varying the concentration of plutonium as a function of axial height, the impact on fuel performance can be explored. For the second comparative case, the plutonium atom fraction was varied from 20 a/o at the bottom of the rod to 0 a/o at z/L = 0.67 (FIG. 3).

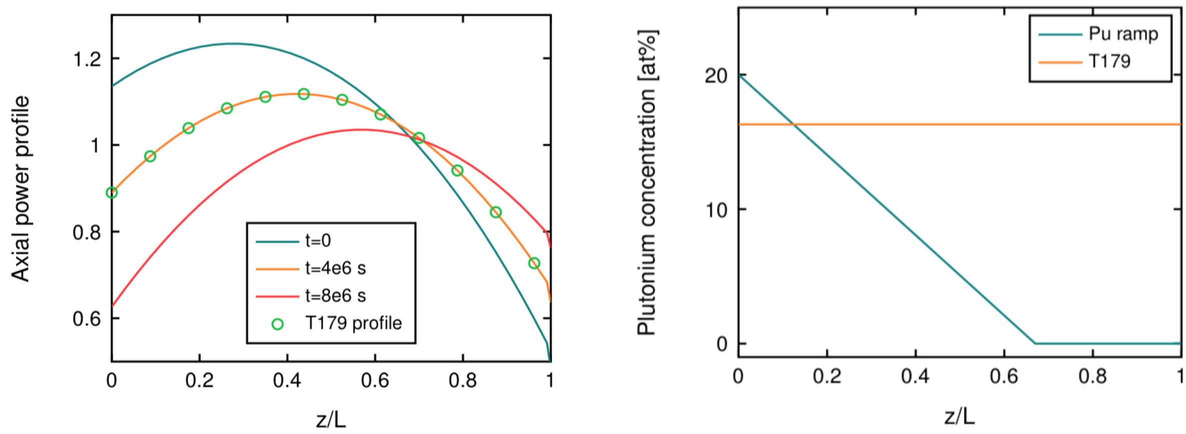

FIG. 3. Variations from the base case in a) power and b) plutonium concentration, as a function of axial location.

The temperature and zirconium distribution for each of the three cases were compared at several time steps, roughly representing 1/3, 2/3 and the final state during irradiation, as presented in FIGS. 3-5. The baseline "T179" case shows similar behavior as was modelled previously [2, 6].

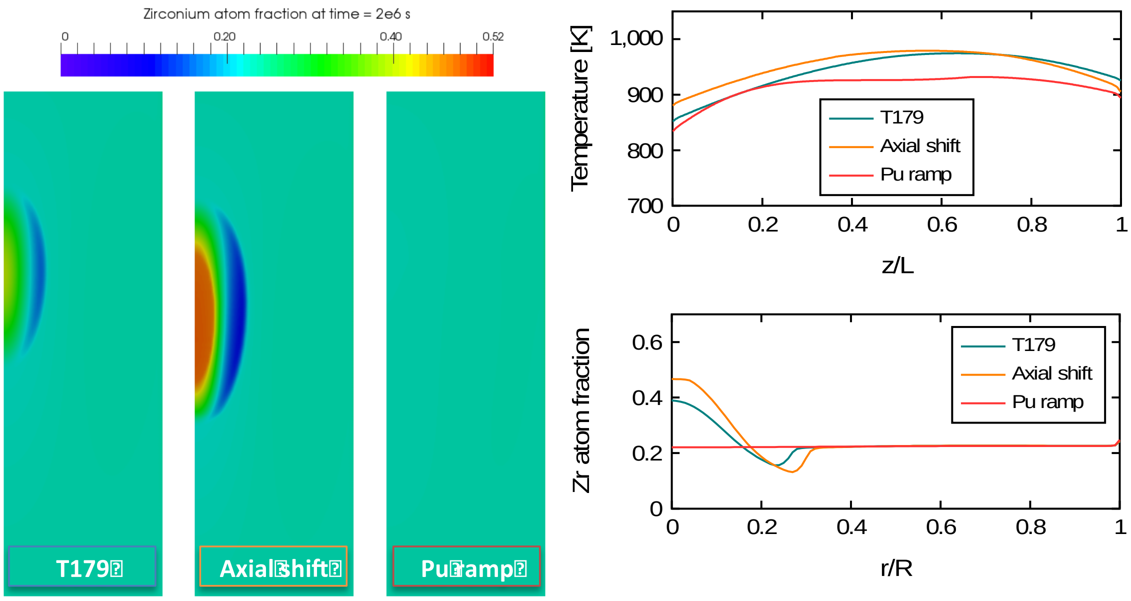

FIG. 4. Simulation results at 2e6 seconds showing zirconium redistribution in a 2D-RZ slice of the fuel (left), centerline temperature (right top), and centerline zirconium atom fraction (right bottom).

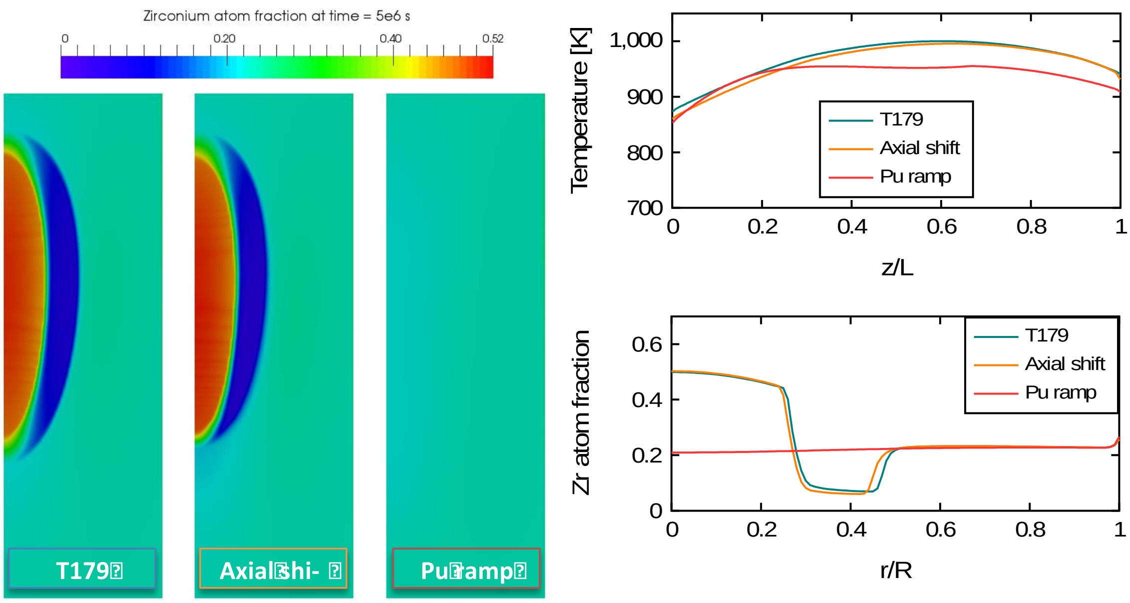

FIG. 5. Simulation results at 5e6 seconds showing zirconium redistribution in a 2D-RZ slice of the fuel (left), centerline temperature (right top), and centerline zirconium atom fraction (right bottom).

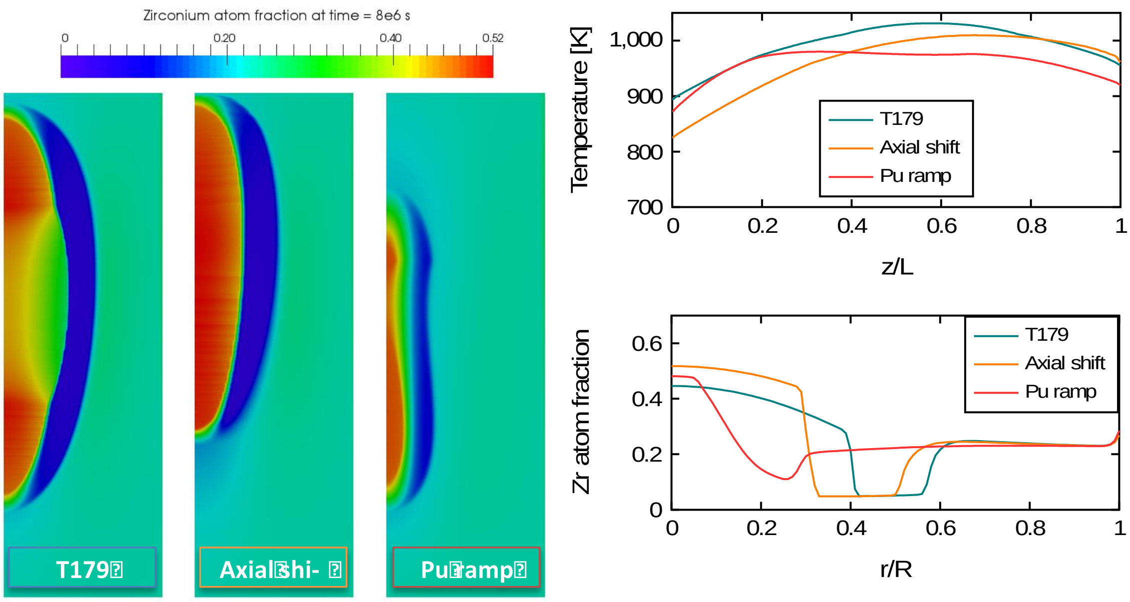

FIG. 6. Simulation results at 8e6 seconds (EOL) showing zirconium redistribution in a 2D-RZ slice of the fuel (left), centerline temperature (right top), and centerline zirconium atom fraction (right bottom).

The "Axial shift" simulation, which experienced initially higher integral power peaking lower in the rod, and ending with a lower final power peaked at the top of the rod, showed that the maximum redistribution effectively followed the movement of the power peak. In FIG. 4, the location of maximum deviation from the initial 22.5 at% zirconium concentration occurred lower than the baseline T179 case, and presented a larger gamma phase region. As the power shifted towards the upper portions of the rod, the gamma phase tended to grow axially, rather than creating a radially large gamma zone, as observed in the T179 rod at the EOL.

Temperatures for the axial shift case were similar to the T179 case, albeit the peak centerline fuel temperature tended to follow the peak axial power shift.

The “Pu ramp” case showed the most deviation from the baseline T179 case, presenting a much more subdued zirconium redistribution. In general, increasing plutonium content results in a lower thermal conductivity of U-Pu-Zr fuels [9]. As a result of axially decreasing the concentration of plutonium in the rod, the temperature profile maintains a flattened arc, and avoids the highly peaked temperature profile. This in turn slows zirconium redistribution such that evidence of movement is not apparent until near the end of irradiation. In addition, the zirconium redistribution radially extends only half as much as the T179 case.

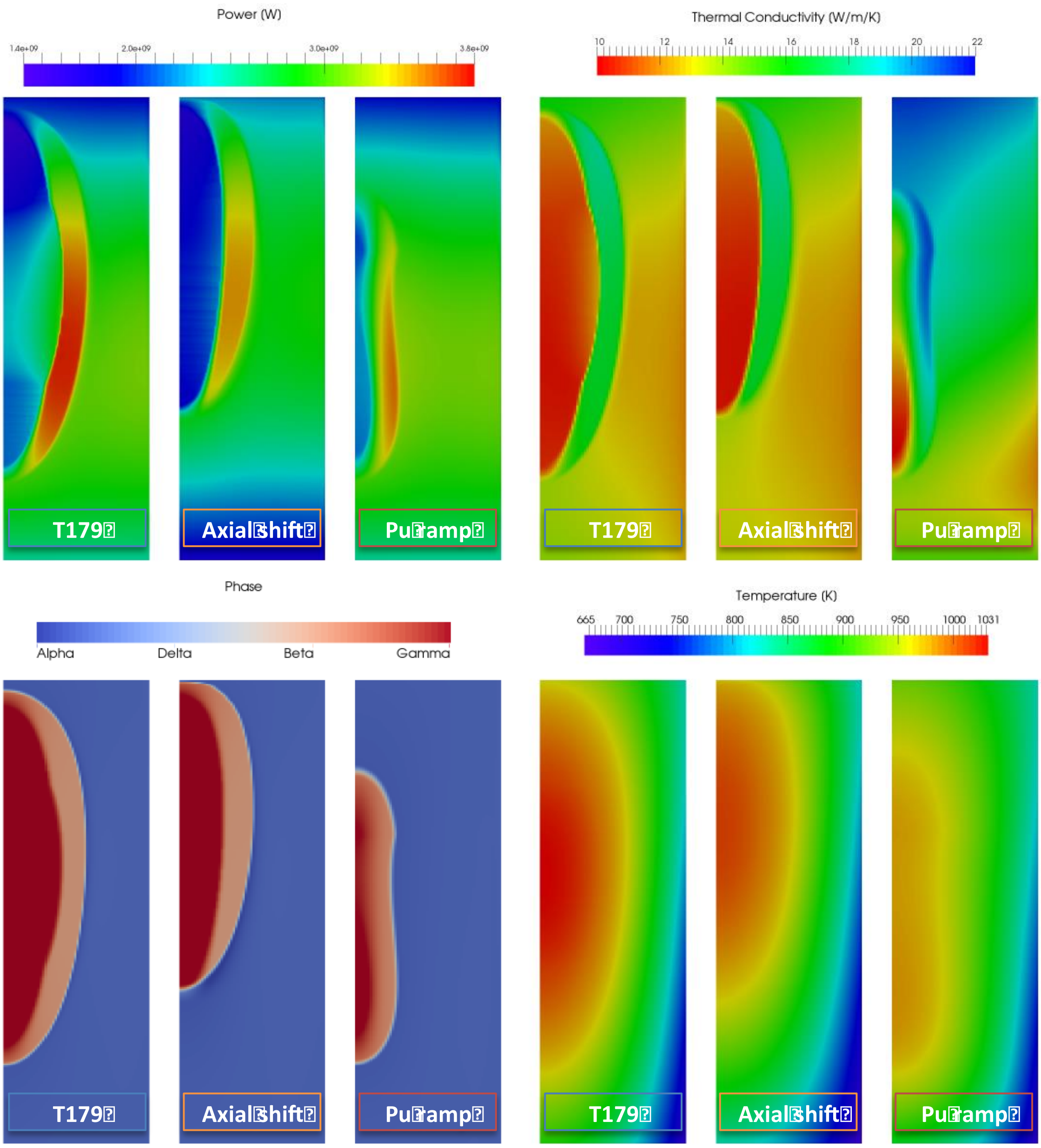

FIG. 7. Simulation results at 8e6 seconds (EOL) showing thermal conductivity, temperature, local power, and phase distribution in a 2D-RZ slice of the fuel.

The variation in temperature distributions becomes most evident when comparing the thermal conductivity, power, temperature, and phase distribution of the different cases, as exemplified in FIG. 7, which displays various conditions at the EOL. Following Eq. 2, the power distribution mimics the zirconium in FIG. 6, resulting in localized power region in the center of the rod that is half as much as in the corresponding surface power. The variation in thermal conductivity follows the phase distribution, as evident when comparing the T179 and Axial shift simulations. In addition, the axial decrease in plutonium content vastly increases the thermal conductivity at the top of the rod. As a result of the deviations in thermal conductivity and power, it is clear that the temperature distribution closely follows the zirconium redistribution, and results in a much cooler rod if the plutonium concentration is decreased axially.

4. Discussion & Conclusions

These simulations only attempted to address the thermal-diffusion problem utilizing the models in BISON. The fuel is assumed to swell due to both solid and gaseous contributions, of which the later affect the thermal conductivity through the introduction of gas bubbles. This contribution is accounted for in the simulations here, but is included using only a simple model, thus further discussion should be limited to future simulations when advanced models become available. In the meantime, it should be recognized that fission gas modifications to the temperature distribution will likely affect all rods in a similar fashion the simulation comparisons here are for the exact same irradiation conditions (e.g. rod power, flow rate, irradiation time).

In addition, the diffusion coefficients and phase diagram utilized to model the zirconium redistribution are only formulated assuming 16.3 at% plutonium concentrations, and are not necessarily directly applicable to binary fuels. However, the transition temperatures for binary fuels are higher and decrease as a function of plutonium [9], thus the zirconium redistribution will only be further reduced if U-Zr coefficients are utilized.

Due to the adaptability of BISON and the recent inclusion of flexible models, it is clear that small changes in either the operating or fabrication procedures produce vastly different results. This is especially clear as the traditionally constant plutonium concentration is allowed to vary. With only simple axial changes, the temperature profile is flattened, and the peak temperature is nearly 50 K less than the baseline case. In addition, by avoiding the plutonium concentration at locations of high cladding temperatures that occur in the upper region of the rod, the Pu-Fe eutectic can be avoided.

As previously discussed, U-Pu-Zr fuel presents a complicated system due to the different phases present in the fuel during irradiation. The differing material properties for each phase results in complex interactions as stresses due to thermal cycling combined with fission gas bubble growth may serve to produce the large cracks observed in ternary fuels. By limiting the volume of the gamma region, it may be possible to suppress the consequences of sharp phase transitions. While the shifting axial power peak results in a axially smaller gamma region in the fuel, the decreasing plutonium concentration results in a much smaller radial gamma accumulation. By tailoring the Pu axially, it may be possible to suppress the total volume of the gamma phase, thus reducing the stresses introduced by differing material properties across sharp phase transitions.

The power deviations and corresponding zirconium redistributions displayed in FIG. 7 highlight the importance of coupling the thermal and diffusion simulation to the power distribution. This can be done either through a tight coupling of MCNP/BISON runs, or more simply through a correlation developed independent of the BISON runs, as was done with Eq. 2. It is important to note that Eq. 2 does not account for the changes between U and Pu fission energy properties, and should only be taken as an estimate of the power distribution change.

Nonetheless, the feedback provided by Eq. 2 impacts the phase distribution in the pin, ultimately driving the thermal conductivity and temperature.

Despite some of the simplifications in these simulations, it is clear that the coupled thermo-diffusion problem can be solved for different model parameters by leveraging the adaptability of BISON to new parameters, and the flexibility of the material models to handle different simulation conditions. Following this baseline, advances in other models such as phase-dependent properties and gaseous swelling rates will further enhance the ability of fuel designers to utilize fuel performance codes as a predictive code to study advanced fuel types and reactor conditions.

5. Acknowledgment

We acknowledge support from the U.S. Department of Energy, Office of Nuclear Energy through the Advanced Fuels Campaign of Fuel Cycle Research and Development Program.

6. References

[1] HOFMAN, G. L., et al., "Swelling behavior of U-Pu-Zr fuel", Metall Trans A **21** (1990) 517–528.

[2] WILLIAMSON, R. L., et al., "Multidimensional multiphysics simulation of nuclear fuel behavior", J. Nucl. Mater. **423** (2012) 149–163.

[3] HALES, J. D., et al., Bison Theory Manual: The Equations Behind Nuclear Fuel Analysis, Idaho National Laboratory, (2016).

[4] GALLOWAY, J., et al., "Modeling constituent redistribution in U–Pu–Zr metallic fuel using the advanced fuel performance code BISON", Nucl. Eng. and Des. **286** (2015) 1–17.

[5] HOFMAN, G. L., et al., "Temperature gradient driven constituent redistribution in U-Zr alloys", J. Nucl. Mater. **227** (1996) 277–286.

[6] KIM, Y. S., et al., "Constituent redistribution in U–Pu–Zr fuel during irradiation", J. Nucl. Mater. **327** (2004) 27–36.

[7] KIM, Y. S., et al., "Modeling of constituent redistribution in U–Pu–Zr metallic fuel", J. Nucl. Mater. **359** (2006) 17–28.

[8] MATTHEWS, C., et al., "Updated Phase Calculation Method for Metallic Nuclear Fuel in BISON", Trans Am Nucl Soc **114** (2016) 1152–1155.

[9] JANNEY, D. E., & PAPESCH, C. A., FCRD Transmutation Fuels Handbook 2015, INL/EXT-15-36520, Idaho National Laboratory, (2015).

[10] YACOUT, A. M., Long-Life Metallic Fuel for the Super Safe, Small and Simple (4S) Reactor, AFT-2008-000056, Argonne National Laboratory, (2008).

[11] COHEN, A. B., et al., "Fuel/cladding compatibility in U-19Pu-10Zr/HT9-clad fuel at elevated temperatures", J. Nucl. Mater. **204** (1993) 244–251.