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Modelling Accident Tolerant Fuel Concepts

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Abstract

The catastrophic events that occurred at the Fukushima-Daiichi nuclear power plant in 2011 have led to widespread interest in research of alternative fuels and claddings that are proposed to be accident tolerant. The United States Department of Energy (DOE) through its Nuclear Energy Advanced Modeling and Simulation (NEAMS) program has funded an Accident Tolerant Fuel (ATF) High Impact Problem (HIP). The ATF HIP is a three-year project to perform research on two accident tolerant concepts. The final outcome of the ATF HIP will be an in-depth report to the DOE Advanced Fuels Campaign (AFC) giving a recommendation on whether either of the two concepts should be included in their lead test assembly scheduled for placement into a commercial reactor in 2022.

The two ATF concepts under investigation in the HIP are uranium silicide fuel and iron-chromium-aluminum (FeCrAl) alloy cladding. Utilizing the expertise of three national laboratory participants (Idaho National Laboratory, Los Alamos National Laboratory, and Argonne National Laboratory), a comprehensive multiscale approach to modelling is being used that includes atomistic modelling, molecular dynamics, rate theory, phase-field, and fuel performance simulations. Model development and fuel performance analysis are critical since a full suite of experimental studies will not be complete before AFC must prioritize concepts for focused development.

In this paper, we present simulations of the two proposed accident tolerant fuel systems: U_3Si_2 fuel with Zircaloy-4 cladding, and UO_2 fuel with FeCrAl cladding. Sensitivity analyses are completed using Sandia National Laboratories' Dakota software to determine which input parameters (e.g., fuel specific heat) have the greatest influence on the output metrics of interest (e.g., fuel centreline temperature). We also outline the multiscale modelling approach being employed.

Considerable additional work is required prior to preparing the recommendation report for the Advanced Fuels Campaign.

1. Introduction

The nuclear energy community seeks accident tolerant fuel (ATF) technology in response to the sequence of events at the Fukushima-Daiichi nuclear power plant in March of 2011. Accident tolerant fuels are defined as those that provide significantly increased response time in the event of an accident while providing performance at least as good as the standard UO_2 – Zircaloy fuel system during normal operation [1].

The United States Department of Energy (DOE) coordinates ATF research at national laboratories, universities, and industrial partners through the Fuel Cycle Research and Development Advanced Fuels Campaign (AFC). The AFC faces an aggressive development schedule. From a broad set of initial ATF concepts, the AFC must give recommendations for a prioritization, where promising concepts will receive focused research attention, in the 2016-2017 timeframe. Despite having studied ATF concepts since shortly after March 2011, a complete set of experimental data on these new materials will not be available in time for the prioritization. Even in 2022, when the first ATF rod or assembly is to be placed in a commercial reactor, the amount of experimental data on the selected ATF will be small.

Computational science research, particularly nuclear fuel performance software development and application, is underway to augment experimental data.

The remainder of this paper is organized as follows. We outline the relationship between our ATF model development work and the AFC. We review model development and application for two ATF concepts, U_3Si_2 fuel and FeCrAl cladding. We then include sensitivity results that help identify important model parameters and potential high-value experiments. The paper ends with a brief set of conclusions.

2. NEAMS ATF HIP

The Nuclear Energy Advanced Modeling and Simulation (NEAMS) program in DOE develops next-generation nuclear engineering software. Among the packages developed by NEAMS is BISON, a fuel performance code, and Marmot, a mesoscale, phase field code tailored to nuclear material behaviour. Other tools (neutronics, thermal hydraulics) are also under development.

A relatively new emphasis in NEAMS is support for High Impact Problems (HIPs). A HIP is defined as a problem seen as high-value by an external entity that can be resolved in a short period of time (3 years or less). NEAMS selected development and application of ATF models as its first HIP. Investigating every ATF concept being too large a task, the HIP focuses on U_3Si_2 fuel and FeCrAl cladding. The ultimate deliverable of the ATF HIP is a report to AFC giving recommendations of whether either of the two studied materials should receive further focused research and development.

Just as a lack of experimental data makes it difficult to select promising ATF concepts from among all proposed, a lack of experimental data also makes it difficult to develop material models. This being the case, a multiscale modelling approach is being undertaken in the ATF HIP.

Material models in fuel performance codes are often empirical fits to experimental data. In the multiscale approach, an empirical model is replaced or improved through lower length scale studies. For example, Marmot simulations at the grain level, in combination with atomistic simulations, can lead to improved engineering-scale models of thermal conductivity and fission gas behaviour [2-3].

Lower length scale material model development of course requires time. In order to begin to provide insight into overall ATF behaviour, we have added several empirical models to BISON based on the best experimental data available. This allows preliminary numerical studies of the ATF concepts to take place while the multiscale models mature. The initial ATF models may be used to suggest experimental work or particular lower length scale modelling needs.

3. U_3Si_2

Uranium silicide fuels have the advantage of a considerably higher thermal conductivity compared to UO_2 (approximately five times higher). This will result in lower fuel temperatures and temperature gradients. Lower thermal gradients should result in less fuel cracking. These effects combine to suggest U_3Si_2 may have much lower fission gas release than UO_2. In addition, silicide fuel has a higher uranium density than oxide fuel.

However, the data available for U_3Si_2 is almost exclusively for dispersion fuel, not monolithic fuel. It is unclear if the dispersion fuel data can be used reliably in modelling fuel pellets. Also, silicide fuel has a lower melting temperature compared to oxide fuel, which could be of concern during rapid high power transients such as during a Reactivity Insertion Accident (RIA).

In BISON, empirical correlations have been added based upon available experimental data for the material behaviour of U_3Si_2. Materials are available for thermal conductivity and specific heat [4], gaseous and solid swelling [5], and fission gas release [6]. Since there is no available creep data for U_3Si_2, the fuel is treated as an elastic material with a Young's modulus of 140 GPa and a Poisson's ratio of 0.17 as per Metzger et al. [7]. The fission gas release model used is the same as for UO_2 in the absence of data suggesting differences in fission gas release behaviour. Since the fuel conductivity of U_3Si_2 is higher than UO_2, it is expected that the fuel temperatures will remain lower during operation and fission gas release will be negligible. However, the rates of diffusion of the fission gas species (such as krypton and xenon) within the U_3Si_2 matrix are not well known. Therefore, bounding studies and sensitivity analyses that vary the intergranular and grain boundary diffusion coefficients can provide insight into fission gas release in U_3Si_2 fuel rods and its effect on temperature. The results of preliminary work in this area can be found in Gamble, et al. [8]. To provide additional insight into the material behaviour during irradiation, the swelling model (a quadratic function of burnup) was broken into two linear functions representing the solid and gaseous components as separate mechanisms. The onset of gaseous swelling is occurs at a burnup of 5% FIMA based upon data by Finlay et al. [5]. Additional details and the equations behind these models can be found in the BISON theory manual [9].

4. FeCrAl

Iron-chromium-aluminum alloys are widely used where low oxidation rate and high temperature performance are important (e.g., coatings on gas turbine blades). The AFC has studied a variety of FeCrAl alloys as possible cladding materials. Clear advantages compared to Zircaloy are the high strength and oxidation resistance of the alloy.

Disadvantages of FeCrAl are its low melting point relative to Zircaloy and its neutronic penalty. Thinner cladding walls, slightly larger pellets, and perhaps higher enrichment will be necessary to compensate for the neutronic penalty.

Similarly to U_3Si_2, empirical models have been added to BISON for the mechanical and thermal behaviour of FeCrAl alloys used for cladding. Models are available for three commercial and one laboratory optimized alloy. Piecewise linear data exists for the thermophysical properties (e.g., specific heat, thermal expansion, thermal conductivity) for the current and previously available commercial alloys Kanthal APMT, PM2000 (no longer available commercially), and MA956. The data is obtained from the respective datasheets provided by the manufacturers. A high temperature thermal creep developed by Seiler et al. [10] is available for MA956, which is temperature and stress dependent. In addition to the commercial alloys, mechanical properties exist (i.e., Young's Modulus and Poisson's Ratio) for the optimized alloy being developed at Oak Ridge National Laboratory (ORNL) called C35M [11]. C35M is the alloy of choice for the studies completed in this work. For thermal properties, those of Kanthal APMT are used, and the high temperature thermal creep model for MA956 is also applied. Representative models have been added for isotropic swelling and irradiation creep of FeCrAl claddings based upon engineering judgment. Recently, an oxidation model has been added to BISON that calculates the oxide thickness during irradiation. The parabolic rate constant is taken from Pint, et al. [12], and the conversion from mass gain to oxide thickness is adopted from Jönsson, et al. [13]. Once again, detailed explanations of the models can be found in the BISON theory manual.

5. Sensitivity Analyses

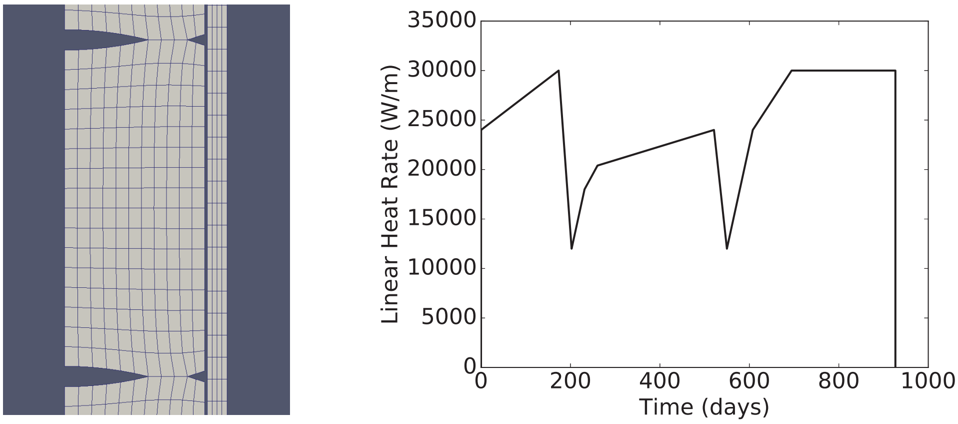
Due to the limited availability of experimental data available for the materials being considered for accident tolerant fuel concepts, investigative studies can be used to gain insight into which parameters have the greatest influence on outputs of interest. These studies can guide experiment design for more in-depth investigations. A variety of sensitivity and bounding study techniques are available including sampling methods, main effects studies, and surrogate models. This paper focuses on main effects studies for two ATF systems: U_3Si_2 fuel and Zircaloy-4 cladding, and UO_2 fuel with FeCrAl cladding. Sandia National Laboratories' Dakota software [14] is coupled to BISON to perform the statistical analyses.

To ensure all of the material and behavioural models are working together as expected, baseline simulations were performed for three systems: U_3Si_2 – Zircaloy-4, UO_2 – FeCrAl, and the traditional UO_2 – Zircaloy-4 system. The model used in this analysis is a modification of the BISON discrete 10-pellet rodlet example problem. A discrete pellet simulation is one that explicitly models the geometric features of fuel dishes and chamfers. To enable comparisons between the different systems, the initial rod diameter (cladding outer diameter) and fuel-to-cladding gap are the same in all cases. The cladding thickness is varied depending on whether the material is Zircaloy-4 or FeCrAl to simulate the thinner cladding required with FeCrAl to overcome the neutronic penalty. To compensate for the thinner cladding, the pellet diameter was increased for the UO_2 – FeCrAl system. Table 1 summarizes the geometric dimensions of the rodlet for the three different systems.

Table 1. Specifications of the fuel rodlets.

	UO_2 − Zircaloy-4 System	**U_3Si_2 − Zircaloy-4 System**	**UO_2 – FeCrAl System**
Number of pellets	10	10	10
Pellet length (mm)	9.83	9.83	9.83
Pellet outer diameter (mm)	8.1915	8.1915	8.6345
Dish depth (mm)	0.3	0.3	0.3
Chamfer width (mm)	0.5	0.5	0.5
Chamfer height (mm)	0.16	0.16	0.16
Radial gap width (μm)	82.55	82.55	82.55
Clad thickness (mm)	0.5715	0.5715	0.35
Rodlet length (mm)	107.203	107.203	107.203
Rodlet diameter (mm)	9.4996	9.4996	9.4996
Initial Fill Pressure (MPa)	2.0	2.0	2.0
Initial Fill Gas	Helium	Helium	Helium
Initial Grain Size (μm)	20	20	20

The mesh density used for all three cases was the same including 2448 QUAD8 finite elements producing 8242 nodes. Figure 1(a) shows a section of the rodlet to illustrate the mesh. The baseline simulations consisted of a representative normal operating base irradiation followed by a Loss of Coolant Accident (LOCA), evaluated for 60 seconds. A flat axial profile is assumed for this short rodlet. To simulate the LOCA, the 1D coolant channel model's inlet mass flux was dropped to 1 kg/m^2-s, and the coolant pressure reduced to atmospheric, thereby significantly reducing the cladding-to-coolant heat transfer coefficient. Meanwhile the power to the fuel is dropped to zero and decay heat is turned on as a source term during the duration of the transient. Figure 1(b) illustrates the base irradiation assumed.



(a) **(b)**

Figure 1. (a) Illustration of the mesh density for the baseline cases and (b) base irradiation power history.

The outputs of interest in this study are the fuel centreline and cladding temperature, cladding strain at the midplane of the rodlet, rod pressure, fission gas released, and oxide thickness. Figure 2 compares results between the three cases during the LOCA transient.

Figure 2(a) shows centreline temperature for the three cases. Initially, temperatures drop due to decreased power (decay heat). As the coolant mass flux drops, the temperatures begin to increase, and the increase continues from that point forward. The silicide fuel case results in the lowest temperatures. At the end of one minute, the fuel centreline temperature is lower for both ATF models, although the difference is slight.

Figure 2(b) gives cladding temperature. Again, the silicide fuel case gives, as expected, the lowest temperatures, with both ATF models showing lower temperatures than the UO2 – Zircaloy case.

Cladding hoop strain, given in Figure 2(c), shows very little change for the FeCrAl case, expected due to its higher strength at high temperature. The UO_2 – Zircaloy-4 model shows an abrupt changing in hoop strain at 10 s. This appears to be due to the elimination of the coolant pressure along with increasing cladding temperatures.

Changes in fission gas release during the 60 seconds modelled here are negligible (Figure 2(d)). Figure 2(e) shows rod internal pressure, which depends on changing internal temperatures and volumes. The UO_2 – Zircaloy-4 case shows a downward trend that is attributed to increased volume (see Figure 2(c)). The FeCrAl model shows, after an initial decrease, increasing pressure. This matches the temperature change well considering the small volume change. The U3Si2 case is relatively flat, with increasing temperature and increasing volume somewhat neutralizing one another.

Very little oxide thickness change can be seen in Figure 2(f). It is worthwhile to note that the amount of oxide for the FeCrAl case is extremely low.

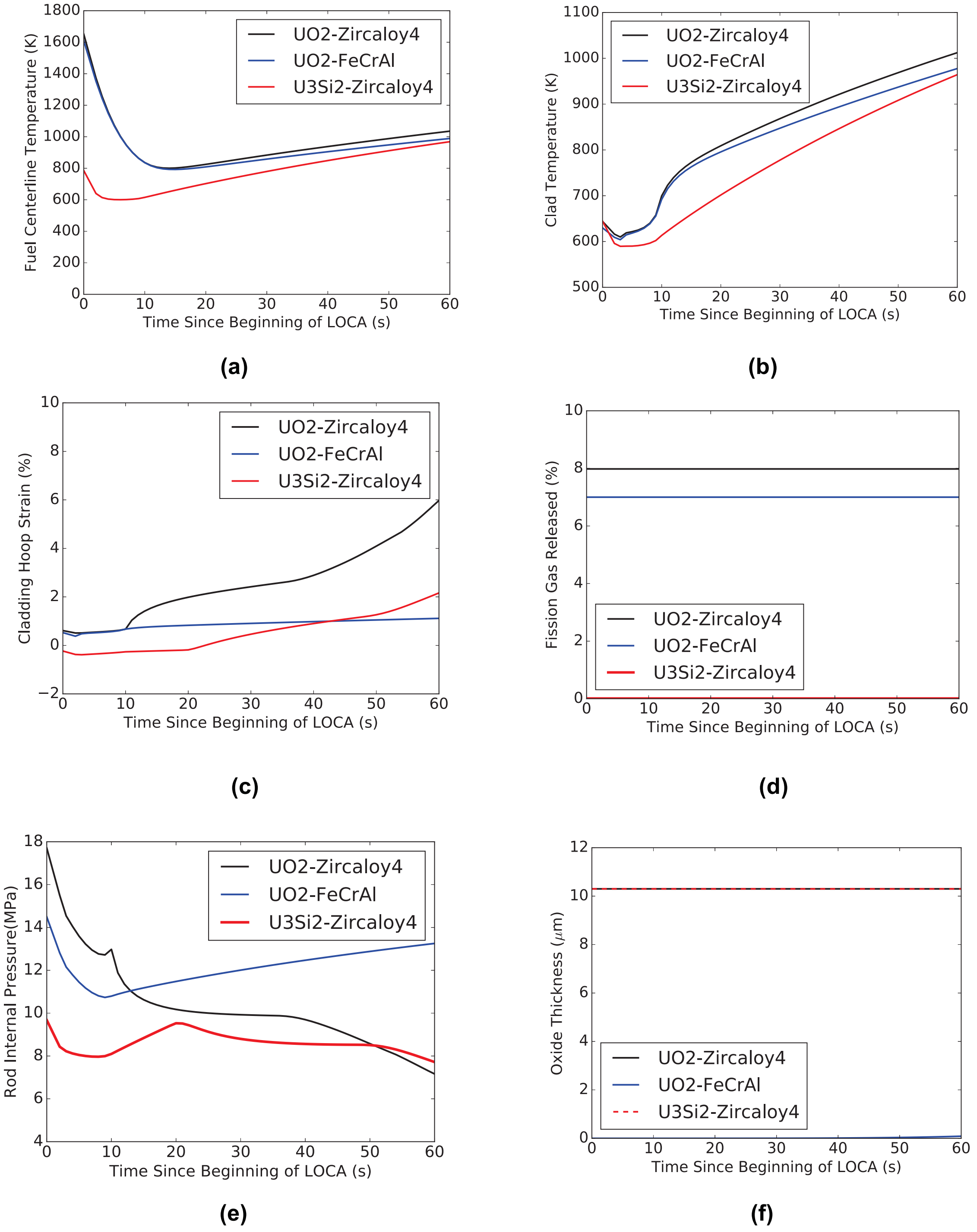


Figure 2. Comparison results of the baseline simulations during the LOCA transient including (a) fuel centreline temperature, (b) cladding temperature, (c) cladding hoop strain, (d) fission gas released, (e) rod internal pressure, and (f) oxide thickness on the outside of the cladding.

In addition to a comparison analysis between the different systems, a main effects study was performed on the U_3Si_2 – Zircaloy-4 and UO_2 – FeCrAl models to investigate the effect uncertain input parameters have on the outputs of interest (i.e., centreline temperature, cladding hoop stress and strain, internal rod pressure, fission gas released, and oxide thickness). For each system, certain input material properties were varied. The chosen material properties were determined based upon the limited experimental data available to develop these models. Thus, the main effects study will highlight which of these parameters have the largest impact on the output metrics. In this main effects analysis each input parameter had three values including a maximum, minimum and mean. Having four input parameters each with three distinct values yields 81 simulations per system. Table 2 summarizes the input parameters of interest and their maximum, minimum and mean values. The first four parameters (intergranular diffusion coefficient, grain boundary diffusion coefficient, thermal conductivity, and gaseous swelling scaling factors) apply to the main effects analysis of the U_3Si_2 – Zircaloy-4 system. The last four parameters apply to the analysis of the UO_2 – FeCrAl system.

Table 2. Parameters varied in the main effects analysis including their minimum, mean and maximum values.

Parameter	Minimum	Mean	Maximum
Inter. Diff. Coeff. Scale Factor	1	5000.5	10000
GB Diffusion Coeff. Scale Factor	1	5000.5	10000
Thermal Conductivity Scale Factor	0.9	1.0	1.1
Gaseous Swelling Scale Factor	0.9	1.0	1.1
Thermal Creep Scale Factor	0.8	1.0	1.2
Irradiation Creep Scale Factor	0.8	1.0	1.2
Swelling Scale Factor	0.8	1.0	1.2
Young's Modulus Scale Factor	0.8	1.0	1.2

Main effects plots provide insight into the influence certain input parameters have on the output metrics. In this study we have four input parameters per system and six output metrics. Therefore, for each case six independent main effects plots are generated. Figure 3 shows the six main effects plots for the UO_2 – FeCrAl system and Figure 4 shows the results for the U_3Si_2 – Zircaloy-4 system.

To explain how to read a main effects plot, consider the point at 0.8 for the swelling scaling factor. This point represents the average centreline temperature for all of the simulations for which the swelling scale factor was 0.8. An increasing slope in the plot represents a positive correlation between the output parameter and corresponding input. A decreasing slope represents a negative correlation. A line close to horizontal indicates that there is no significant correlation between the variation of the input parameter and the output. Note that in the analyses completed here the point represents the average value at the end of the LOCA calculation (after 60 seconds).

Figure 3(a)-(f) highlights the results of the main effects study for the UO_2 – FeCrAl ATF system. The results indicate that the swelling scale factor and Young's modulus scale factors have strong correlations with the outputs of interest whereas the thermal and irradiation creep scaling factors have minimal effect on the results. As expected the Young's modulus has a strong correlation with cladding hoop stress and strain results. The reader is drawn to the scales on the output parameters. Even though there are strong correlations between the swelling scaling factor and the Young's modulus scaling factor, the average increase or decrease in the output parameters is quite small (e.g., 1.8 K for centreline temperature). Therefore, it can be concluded from this study that for

FeCrAl alloys, varying the swelling rate, Young's Modulus, and thermal and irradiation creep strain rates by ± 20% has minimal influence on the centreline temperature 60 seconds into a LOCA transient following a typical base irradiation.

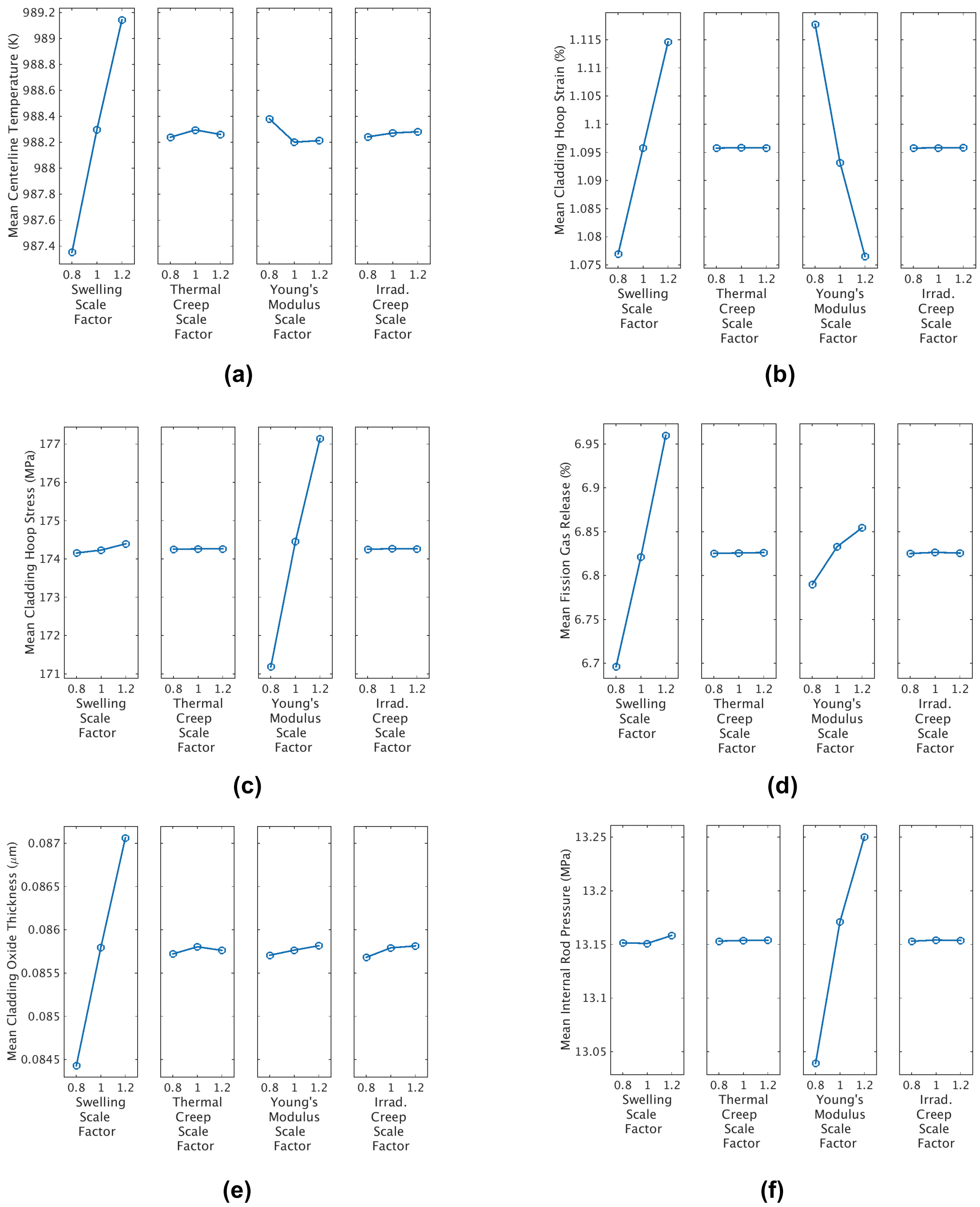


Figure 3. Main effects plots for the UO_2 – FeCrAl system for (a) fuel centreline temperature, (b) cladding hoop strain, (c) cladding hoop stress, (d) fission gas release, (e) cladding oxide thickness, and (f) rod internal pressure.

Figure 4(a)-(f) highlights the results of the main effects study for the U_3Si_2 – Zircaloy-4 ATF system. The results indicate that further investigation into the mechanisms of fission gas release and gaseous and solid swelling are required to improve our understanding of the behaviour of U_3Si_2 in accident conditions. A quick overview of Figures (a)-(f) indicates that the uncertainty in the intergranular diffusion coefficient has a significant positive correlation with all the outputs. Therefore, research in the area of fission gas diffusion and release mechanisms in U_3Si_2 will be valuable. As expected, the thermal conductivity has a negative correlation with the fuel centreline temperature in Figure 4(a) and a positive correlation with the cladding oxide thickness Figure 4(e). The gas swelling factor had minimal effect on all the output metrics. It is known that after a burnup of approximately 5% FIMA there is significant gaseous swelling. However, the power history shown in Figure 1(b) did not produce a high enough burnup in the U_3Si_2 fuel to induce significant swelling. The higher uranium density of U_3Si_2 also reduces the burnup reached compared to UO_2 for the same power history.

6. Conclusion

In this paper we have outlined the format of the NEAMS ATF HIP and described the multiscale multiphysics approach being used to provide an informed recommendation on which ATF concepts to prioritize for further investigation. In addition, fuel performance simulations were presented for two accident tolerant fuel systems (U_3Si_2 – Zircaloy-4 and UO_2 – FeCrAl) during the first 60 seconds of a LOCA following a representative base irradiation. It was observed that fuel and cladding temperatures during the LOCA approach similar values for both the uranium silicide and FeCrAl cases. Rod internal pressure is higher in the FeCrAl case due to less cladding creep, whereas the uranium silicide rod experienced a decreasing rod internal pressure as the Zircaloy-4 cladding began to balloon.

Sensitivity analyses were also performed to investigate the effect uncertainty in the material properties and behaviour models of the proposed accident tolerant materials has on the output parameters of interest during accident scenarios. It was observed that uncertainty in the FeCrAl material properties had minimal effect on the outputs of interest. The intergranular diffusion coefficient used in fission gas release modelling of U_3Si_2 has a relatively large influence on all the outputs studied, indicating that research into fission gas release mechanisms will be needed to improve the quality of predictions.

As the lower length scale research develops, mechanistic models of FeCrAl and U_3Si_2 will be available for input into BISON for more advanced simulation and analysis.

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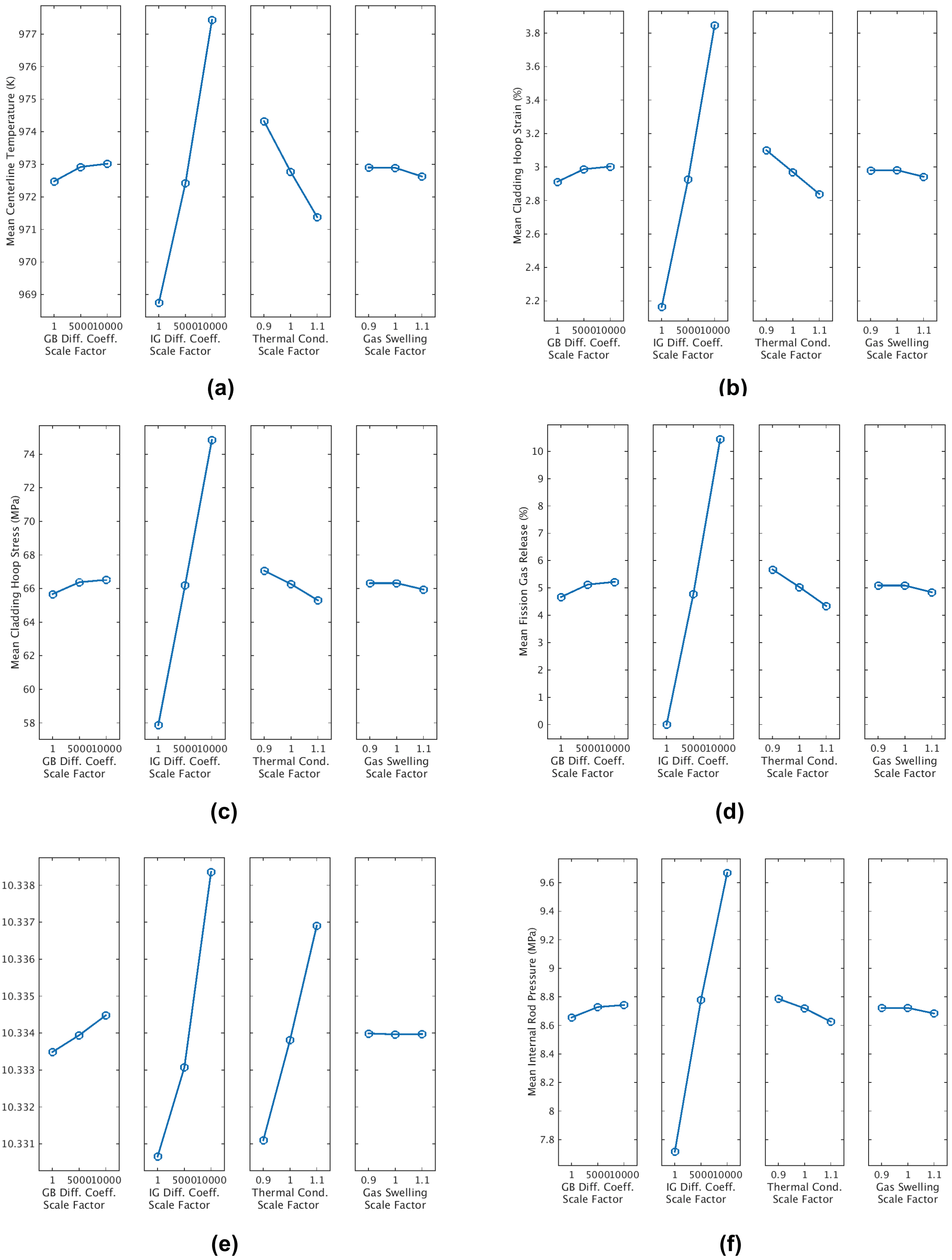

Figure 4. Main effects plots for the U_3Si_2 –Zircaloy-4 system for (a) fuel centreline temperature, (b) cladding hoop strain, (c) cladding hoop stress, (d) fission gas release, (e) cladding oxide thickness, and (f) rod internal pressure. The x-axis label for the grain boundary and intergranular diffusion coefficients have values at 1, 5000, and 10000.

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