EPJ Web of Conferences **302**, 05008 (2024) https://doi.org/10.1051/epjconf/202430205008
SNA + MC 2024

Simulating Hydrogen Diffusion in ZrH Moderator and its Impact on Coupled Neutronic-Thermal Simulation of a Heat Pipe Microreactor

William Reed Kendrick[1,*] and *Benoit* Forget[1,**]

[1]Massachusetts Institute of Technology

Abstract. Metal hydride moderators are a common material choice for moderating compact reactor designs, as their high thermal limits and high density of hydrogen make them favorable for operation. One additional attribute of metal hydrides, zirconium hydride in particular, is the relatively large mobility of hydrogen within the metal lattice, especially at high temperatures and under large thermal gradients. As hydrogen's spatial distribution directly impacts neutron thermalization in the system, and therefore power shape, changes in hydrogen concentration of the moderator may be important to analyze. This work couples OpenMC and MOOSE in order to solve the feedback loop of the coupled power distribution-thermal distribution-hydrogen migration without relying on mesh discretization. Functional expansions are used extensively as a method of transferring spatial information, and continuously varying material tracking is used in the neutronic solver to represent the change in hydrogen concentrations. The resulting hydrogen redistribution is highlighted, as well as the resulting neutronic and thermal impact of this redistribution.

1 Introduction

The current nuclear landscape is evaluating the role for smaller reactors, especially those in the "micro reactor" category [1]. These micro reactors vary widely in their design, however those that are thermal reactors still require moderation. Smaller geometry begets a need for a moderating material with good stopping power and of high density, and zirconium hydride is one such material being strongly pursued.

Zirconium hydride exists in the form of a crystal zirconium lattice with hydrogen dispersed between [2]. That hydrogen is mobile at high temperatures, the likes of which are commonly seen in some reactor designs. Highly mobile hydrogen will migrate, and changes in the hydrogen concentration will impact the neutron spectrum, which in turn impacts power distribution, which causes temperature changes that further affect hydrogen concentration.

Traditionally coupled simulation centers around a neutronic simulation providing power to inform temperatures, and a thermal simulation providing temperature to inform cross-sections [3]. In a large-scale reactor, this process is incredibly computationally costly [4], with the neutronics solve alone requiring roughly 200,000 CPU hours on modern architecture [5]. The uniquely small geometry of micro reactor designs creates an opportunity to expand

*e-mail: rkendric@mit.edu
**e-mail: bforget@mit.edu

© The Authors, published by EDP Sciences. This is an open access article distributed under the terms of the Creative Commons Attribution License 4.0 (https://creativecommons.org/licenses/by/4.0/).

this coupling system to include an additional facet of the feedback loop, specifically the hydrogen migration previously mentioned.

This paper does so by coupling OpenMC and MOOSE as a neutronic-thermal simulation structure to calculate hypothetical hydrogen migration and its resulting effects. A hypothetical heat pipe microreactor geometry is used, and additional work is done to analyze the potential impacts of heat pipe failure.

2 Background

2.1 Diffusion in Zirconium Hydride

Diffusion of hydrogen in the zirconium lattice is governed by two systems, Fick's Law and the Soret effect. Essentially, this is diffusion driven by concentration gradient and thermal gradient. As previously noted, interest in the alpha phase of zirconium hydride for cladding purposes has been a significant source of characterizing hydrogen migration in zirconium. A good example of this is Courtey's work [6] modeling hydrogen behavior in Zircaloy-4, which is the source for the following equation that describes hydrogen migration in zirconium, Eq. 1.

$$J = J_{Fick} + J_{Soret} = -D \cdot \nabla C - \frac{DCQ^*}{RT^2} \cdot \nabla T \tag{1}$$

J_{Fick} is hydrogen flux governed by Fick's law, where C is the hydrogen concentration and the diffusion constant D for hydrogen in the zirconium lattice is determined via the Arrhenius equation. For this work, a diffusion coefficient of $D = 1.53 \times 10^{-7} \cdot \exp(-14000/RT)$, defined in [7] and utilized in [8]. J_{Soret} is hydrogen flux governed by the Soret effect, where Q^* is the heat of transport. This heat of transport is assumed to be a value of 25.1 kJ/mol, in agreement with [6].

It's important to note that zirconium hydride is not a single-phase material, but has multiple phases depending on the temperature and hydrogen concentration [9]. Delta phase zirconium hydride is the phase used for moderating purposes [2], and will be what this paper uses as moderator. In order to avoid crossing into an area where the material may become epsilon phase, attention will be paid to any moderator temperatures that exceed a rough limit of 860 to 870 degrees Kelvin.

2.2 Functional Expansions

One distinct challenge in coupling Monte Carlo neutronics to a finite element-based solver is the difference in physical domain being solved. Monte Carlo neutronics generally use Constructive Solid Geometry (CSG), where surfaces are explicitly defined and volumes are created through boolean operation with those surfaces. Tallies then are accumulated during simulation by volume integration of the tally score over the volume. Finite element solvers calculate node-to-node, or quadrature point-to-quadrature point, and volume averages are a tertiary operation that enacts by parsing the node data.

This implies that traditional coupling of the two simulations would mean passing volume-averaged data from one to inform the behavior of the other. Refinement of the quality of information being passed requires refining the geometry itself, discretizing the CSG model, making the already costly neutronic solve even costlier [10].

Instead, the use of functional expansions to represent spatial heterogeneity has been suggested as a solution to this challenge [11, 12]. By calculating expansion coefficients that fit orthonormal basis sets to an input, a series of easy-to-calculate, continuous functions can

replace the often complicated point-wise data. The choice of function series used for this process strongly impacts the accuracy and efficiency with which the true spatial information can be reproduced. A common choice, and the choice that this work uses, is rebuilding cylindrical geometries using a combination of Zernike polynomials for the radial and azimuthal dimensions, and Legendre polynomials for the axial dimension.

Using the expansion coefficients as an input for MOOSE is easily achievable, but in the case of OpenMC, one is required to introduce another complication as explained in section 2.3.

2.3 Continually Varying Material Tracking

Although the explicit coupling scheme won't be defined until section 3.1, it has already been mentioned that MOOSE will be solving the diffusion of hydrogen in the moderator and reporting this spatial distribution as an output for our neutronics solve to use as an input. What "spatial distribution of hydrogen" means for OpenMC is a set of expansion coefficients, described in section 2.2, that multiply a set of basis functions to represent the spatial distribution of hydrogen's number density in that specific moderator material. This presents a problem, because in all versions of OpenMC publicly available, materials are defined to be constant in their properties within a cell [13].

Part of the Monte Carlo process is calculating the probability of colliding before reaching a surface, which requires an analytical integral along a characteristic line through the material. While the combination of Zernike and Legendre polynomials are simple to evaluate at any point in the cylindrical phase space, they are not analytically solvable along this line.

Instead, it is necessary to utilize the technique of Continually Varying Material Tracking (CVMT), developed by Brown and Martin [14], expounded upon by Griescheimer [12], and first utilized in the context of OpenMC by Ellis [10]. This method is rather complex, but in essence, it is a way of calculating distance to collision via a series of numerical integrals.

This represents a significant increase in computational steps during particle transport. What was once a simple evaluation by integrating a constant cross-section across a distance, is now potentially a large number of numerical integrals. Beyond that, this also means that track-length estimation isn't usable for tallying, as it would require the same numerical integration process. The positives of this method are numerous, however. With the correct code implementation, this allows the work of this paper in performing particle transport on moderator material with spatially varying hydrogen concentration.

CVMT is not the sole method of handling spatially varying material properties in a neutronics solver. Serpent 2 features a method similar to delta tracking where a scheme featuring rejection sampling is implimented [15]. This method was not pursued for this work as calculating the maximum total cross-section, a requirement for the previously mentioned method, can be difficult when using continuous distributions of number densities and CVMT has been previously implemented in the OpenMC framework [10, 16].

2.4 Heat Pipes

Heat pipes are typically metal tubes filled with highly heat-conducting material and feature a central wick to enable capillary action. Heat pipes feature some advantageous characteristics for reactor use, such as the lack of moving components. A downside, is that failure is not uncommon for heat pipes, due to tight manufacturing tolerances and the presence of several operational limits [17]. Heat pipes can also fail as part of a "cascade", where other failures increase the thermal load on a heat pipe and surpass an operational limit, causing secondary failures. This makes their presence an interesting note for the reactor design discussed in this

work. Therefore, this work will include a case study by "failing" four selected heat pipes in the center of the core, aiming to simulate a minor cascade failure, and analyze the impact on hydrogen migration and its effects.

For the purposes of this work, heat pipe behavior is simulated in MOOSE as a custom object. The code within was based on Los Alamos Laboratory's HTPIPE [18], and features a simple 1D iterative pressure and temperature solver. The codes response has been compared against HTPIPE and SOCKEYE results in similar environments, and has been determined to be sufficiently accurate to be used as a hypothetical potassium heat pipe response object.

3 Methods

3.1 Coupling Scheme

Coupling for this process is distinctly separate, with each simulation process occurring standalone, followed by a step for processing results and converting information into a format readable to the next simulation. That information is as previously described; the neutronic OpenMC solve exports volumetric heating rate data, while the thermal-diffusive MOOSE solve exports hydrogen density data. In both cases, this data is in the form of series of expansion coefficients.

The cycle begins with an initial OpenMC step, and for this work the iterations ended after both MOOSE and OpenMC had completed 3 steps. At that point, the results were compared and determined to have converged. Future work will seek to quantify this convergence.

Do note that temperatures are not part of the data exchanged in this work. All neutronics simulations occur with a constant temperature of 800 degrees Kelvin. This was done in order to isolate the effect that hydrogen migration has. If the hydrogen migration causes significant changes in temperature, this would not be acceptable, as the effect of cross-section changes would be important to quantify. As will be seen in the Results section, this is not a concern.

3.2 OpenMC

OpenMC is utilized extensively for all neutronics simulations described in this work. OpenMC is a Monte Carlo particle transport code developed by Massachusetts Institute of Technology and Argonne National Laboratory [13]. Designed with focus on parallelizability and featuring an extremely user-friendly Python API, the OpenMC code is very well suited for a work like described in this paper. Having an open-source code with a varied developer network made implementing an in-depth change like CVMT, described in section 2.3, possible with minimal headache.

3.3 MOOSE

MOOSE is a finite element physics framework developed by Idaho National Laboratories [19]. It features extremely parallelizable code for high efficiency on clusters, as well as well documented and user-friendly methods of implementing one's own physics kernels. MOOSE is used for all thermal and hydrogen simulation in this work, relying on modules that are readily available as an open-source download [20, 21].

Of note for the diffusion solve for hydrogen, leakage is not taken into account. Hydrogen is an element well known for its ability to penetrate and escape its containment, so zirconium hydride moderator is expected to have a cladding of some kind [22]. Both OpenMC and MOOSE models do not feature any cladding on the moderator, however its assumed that no leakage will occur for the sake of simplicity.

4 Results

The reactor geometry simulated in this work is one provided by the Idaho National Laboratory Virtual Test Bed Reactor repository on GitHub [23], specifically the "MRaD" geometry [24]. A visual of the core geometry can be seen in figure 1. Fuel material is UCO TRISO fuel at 14% and 40% packing fraction, moderator before diffusion is $ZrH_{1.67}$, heat pipes are homogenized potassium-stainless steel. The monolith is graphite, reflector is boron steel, and the absorbers are boron carbide.

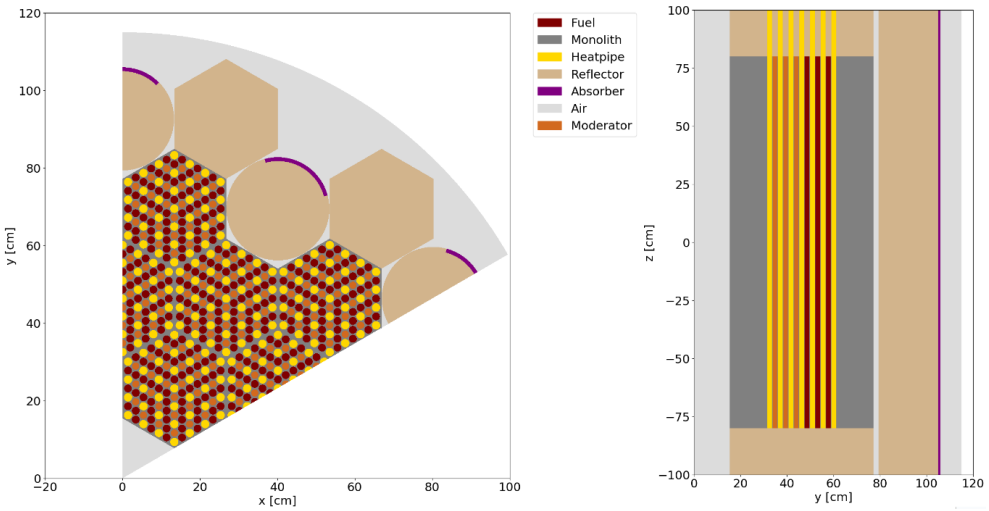


Figure 1: Core geometry and material assignments.

The reactor is simulated to produce 345.6 kW of thermal power. Steady state temperature results show moderator temperatures do not exceed the previously mentioned 860-870 Kelvin range, important in establishing that the zirconium hydride remains in its delta phase. All results in the following sections are the results from the third iteration cycle. By the third cycle, variations were low enough to suggest that the process is converged.

4.1 Hydrogen Migration

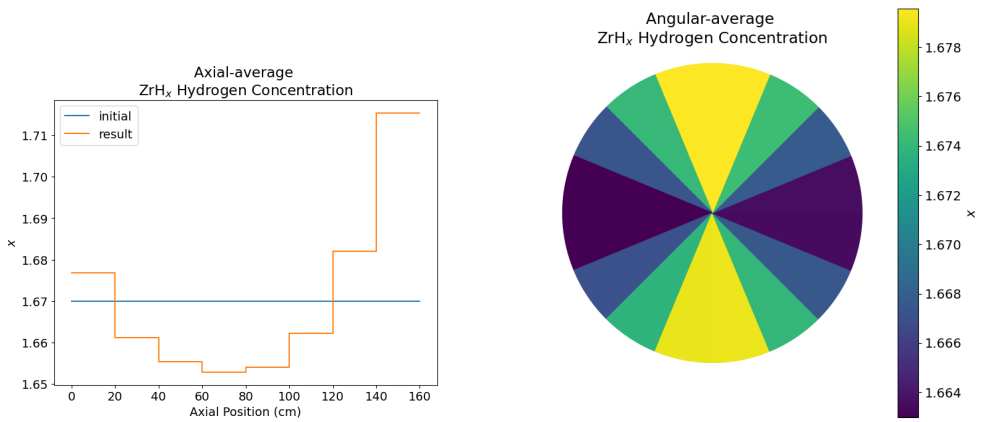
The first category of results analyzed is the hydrogen migration itself. Hydrogen migration will vary between moderator rods in the core depending on the orientation of heat pipes and fuel rods surrounding the rod, therefore this section will solely focus on a single rod. That rod is the central-most moderator rod of the central-most assembly, and feature the highest average temperature in the MOOSE outputs. That way these results may be viewed as a "worst-case" for the hydrogen response of the moderator rods in this core.

Because polynomial expansion coefficients are returned for every moderator rod at the end of the MOOSE solve, it is also possible to numerically integrate over the cylindrical volume to return volume-average quantities. For this work these integrals were performed while subdividing one dimension of the geometry at a time. For example, the first results seen here in figure 2a are averaging over the axial segments of the moderator rod in order to return a plot of axial-averaged ZrH_x hydrogen content. Similarly, figure 2b displays the results of angularly-averaged hydrogen content in the moderator rod. Not displayed is the radial-averaged case, which was also calculated. All three analyses show clear movement

Table 1: Global neutronic results and differences from initial flat-hydrogen case. "AR" is Absorption Rate, "FF" is fission factor. Absorption rates are in units of "per source particle".

Quantity	Result Value	Difference From Initial	Pct. Dif. (%)
Eigenvalue	1.02689 ± 0.00021	-0.00032 ± 0.00025	-
Thermal FF	0.79989 ± 5.0e-5	-0.00028 ± 6.0e-5	-0.034 ± 0.008
Fuel AR	0.65513 ± 1.2e-4	0.00003 ± 1.4e-4	0.004 ± 0.021
Heat Pipe AR	0.10636 ± 2.9e-5	-0.00012 ± 3.5e-5	-0.112 ± 0.033
Moderator AR	0.04373 ± 9.0e-6	-0.00016 ± 1.1e-5	-0.371 ± 0.024
Reflector AR	0.13731 ± 7.0e-5	0.00022 ± 8.0e-5	0.160 ± 0.060

of hydrogen, though the axial distribution stands out as the largest magnitude of change displayed. It should be noted that the angular-average values are particularly minor, due to the angular shape being highly varied depending on the axial position. This makes the integrating and averaging process "wash out" the actual angular disparities. For example, an axial midplane slice of the moderator rod has a range of ZrH_x values from $x = 1.633$ to $x = 1.682$.



(a) Axial average.

(b) Angular-average

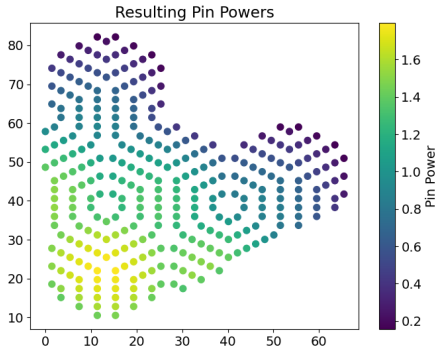
Figure 2: Discritized volume-averages for hydrogen content of moderator rod before and after coupled solves.

4.2 Neutronic & Thermal Response

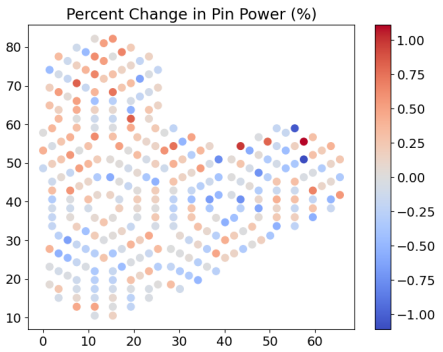
While seeing how hydrogen responds to the thermal profiles in the core is interesting, what is of most importance for this work is how that new hydrogen spatial distribution affects the neutronic and thermal solution of the core. To begin with, some global neutronic effects are listed in table 1. The general trend is a very minor impact to global neutronics.

The next category to analyze is fuel pin powers. Pin power is a measure of the power generated by a fuel pin, where units are individual power normalized by average fuel pin

power. This core features uniform, fresh fuel, so pin power are expected to be quite non-uniform. This is what we see in figure 3a, a plot of fuel pins and their pin powers. Also included in figure 3b is a plot of the percent change in pin power from the initial flat hydrogen case to the resultant iterated hydrogen shape case.



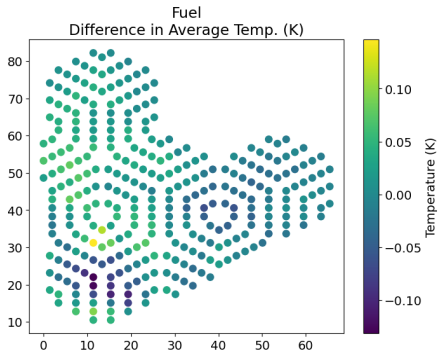
(a) Coupled simulation resultant pin power distribution.



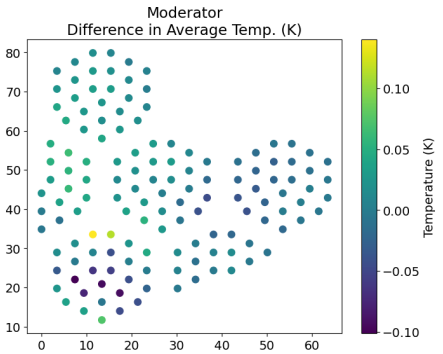
(b) Percent change in pin power, comparing final result with initial flat-hydrogen case.

Figure 3: Pin power impact analysis from hydrogen migration inclusion.

These pin powers represent volumetric heating rates, which in MOOSE can produce temperatures that can be compared to gauge one dimension of the effect that implementing hydrogen migration has. On average, however, both fuel pins and moderator rods changed at most 0.1 degrees Kelvin. In general, looking at figure 4a and 4b, the hottest-temperature area of the core saw a very slight decrease in temperature of both moderator and fuel due to hydrogen migration's impact.



(a) Average fuel temperature change from initial flat-hydrogen case to final result.



(b) Average moderator temperature change from initial flat-hydrogen case to final result.

Figure 4: Fuel and moderator temperature impact analysis from hydrogen migration inclusion.

Because the volumetric heating rates for moderator rods are expressed via functional expansions, the same type of analysis can be performed as was shown in section 4.1. These values, however, show relatively little change in spatial power recorded for both moderator and fuel. For both materials, there is a slight shift in axial power from the center to the top and bottom of their lengths. The moderator rod also sees a slight shift radially outwards in power. Beyond this, the changes are small enough to be hard to distinguish from the stochastic errors' influences.

4.3 Heat Pipe Failure Case

In order to simulate a minor heat pipe failure cascade in the reactor, four heat pipes were "failed" and set to not remove heat during simulation. These heat pipes can be seen in figure 5.

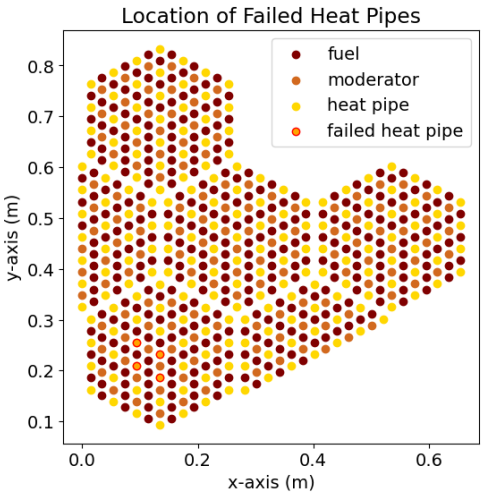
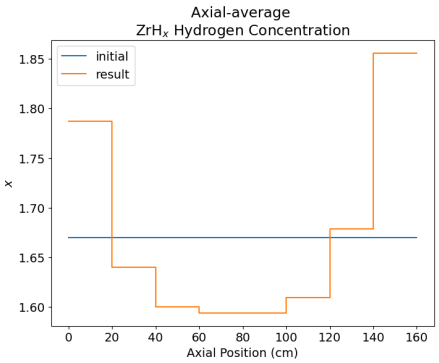


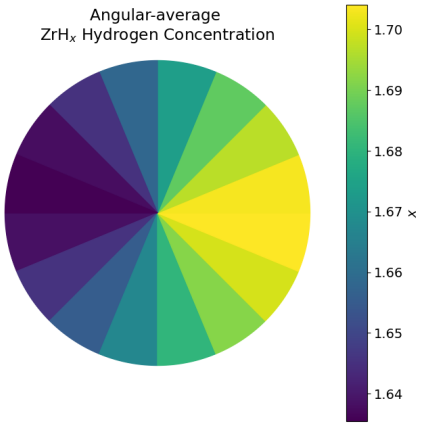
Figure 5: Reactor cylindrical geometries and selected heat pipes for failure simulation.

The immediate effect of these heat pipe failures is an increase in temperature of the central assembly of the core. The average temperature of the hottest fuel pin increases by 66 degrees K, and the average temperature of the hottest moderator rod increases by 54 degrees. The outer assemblies see less impact, below 10 degrees kelvin, as the functioning heat pipes pick up the load in heat removal. This centralized temperature increase then has an impact on the hydrogen shape of the moderator rods in that area. The selected moderator's axial hydrogen shape, seen in figure 6a, has an even stronger peak at each end. The angular shape, seen in figure 6b becomes peaked towards the nearest functioning heat pipe.

The feedback loop continues as the hydrogen shape alters the average temperatures of the fuel and moderator, seen in figure 7a and 7b. This impact remains small in the overall scheme of things, however the previous trend seen in section 4.2 and figures 4a and 4b continues; In high temperature regions, the increased hydrogen mobility in the moderator causes a minor decrease in power and therefore temperature. This effect is limited to the area in which the heat pipes failed. Globally, the pin power peaking factor decreased by 0.64 percent, but on average, power remained where it was and at its previous levels from before the hydrogen migration.

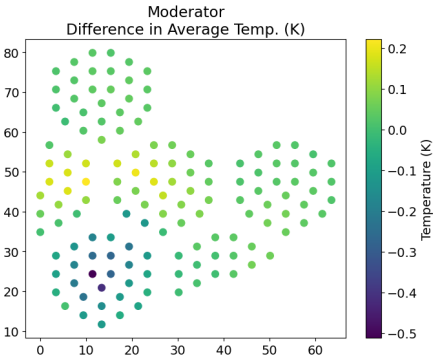


(a) Axial-averaged moderator hydrogen concentration post-heat pipe failure.

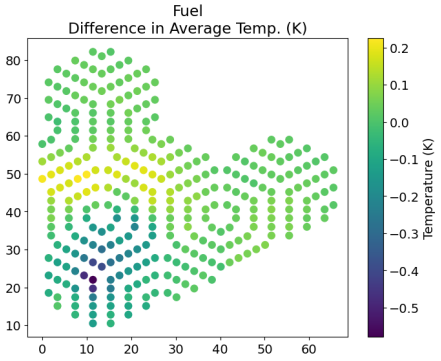


(b) Angular-averaged moderator hydrogen concentration post-heat pipe failure.

Figure 6: Discretized volume-average hydrogen concentration analysis of moderator rod after heat pipe failure.



(a) Moderator temperature differences compared to flat-hydrogen case post-heat pipe failure.



(b) Fuel temperature differences compared to flat-hydrogen case post-heat pipe failure.

Figure 7: Temperature differences between flat-hydrogen case and hydrogen migration case post-heat pipe failure.

It is also very important to note that in this case of heat pipe failure temperatures have exceeded the 860-870 Kelvin range for $ZrH_{1.67}$. This limit was mentioned because beyond this limit zirconium hydride begins to form into its epsilon phase, and the diffusion parameters used are not necessarily applicable. This failure case study must be seen as an approximation, and not a true representation of real life zirconium hydride behavior.

5 Conclusion

In terms of the global neutronic-thermal operation of the reactor, the effect of including hydrogen migration into coupled simulation can be accurately described as "minimal". Eigenvalue change is on the order of tens of pcm at most, and the distribution of power in the core remains unchanged. This should be the expected result, given two points:

- The core is designed to be roughly homogeneous, with an even distribution of fuel, moderator, and heat pipes. If the core design was a single row of fuel, a single row of moderator, and a single row of heat pipes, the radial/azimuthal shift might be important, because every moderator would have the same shift.
- A maximum increase or decrease in hydrogen content of 4% is, at the end of the day, just 4%. Because no hydrogen is lost, just redistributed, a swing of ±4% shouldn't be expected to drastically alter the neutronic response of this small core.

This doesn't mean that there was no impact, however. It's clear that axial hydrogen migration takes place in every moderator rod in the core, and this has an impact when temperatures are at their highest, such as the failure case in section 4.3. The increased axial migration appears to dampen the local power. Overall, the inclusion of hydrogen diffusion may not be a concern for reactor designers, given the results seen here. A minor positive damping effect due to axial hydrogen migration is recorded, but few other trends can be observed. However, this work does not include the leakage of hydrogen from the moderator, which may change these results.

References

[1] *Fact sheet: Biden-Harris Administration Announces New Steps to Bolster Domestic Nuclear Industry and Advance America's Clean Energy Future* (2024), [Online; posted 29-May-2024], https://www.whitehouse.gov/briefing-room

[2] A.P. Shivprasad, J.R. Torres, T.E. Cutler, J.K. Jewell, V.K. Mehta, S. Widgeon Paisner, C.A. Taylor, C.N. Taylor, H.R. Trellue, D.W. Wootan et al. (2022)

[3] C. Matthews, V. Laboure, M. DeHart, J. Hansel, D. Andrs, Y. Wang, J. Ortensi, R.C. Martineau, Nuclear Technology **207**, 1142 (2021), https://doi.org/10.1080/00295450.2021.1906474

[4] J. Yu, H. Lee, H. Kim, P. Zhang, D. Lee, Nuclear Engineering and Technology **52**, 661 (2020)

[5] G. Gunow, B. Forget, K. Smith, Annals of Nuclear Energy **134**, 299 (2019)

[6] O. Courty, A.T. Motta, J.D. Hales, Journal of Nuclear Materials **452**, 311 (2014)

[7] G. Majer, W. Renz, R.G. Barnes, Journal of Physics: Condensed Matter **6**, 2935 (1994)

[8] J. Huangs, B. Tsuchiya, K. Konashi, M. Yamawaki, Journal of Nuclear Science and Technology **37**, 887 (2000), https://doi.org/10.1080/18811248.2000.9714969

[9] K. Barraclough, C. Beevers, Journal of Nuclear Materials **34**, 125 (1970)

[10] M.S. Ellis, Ph.D. thesis, Massachusetts Institute of Technology, Cambridge, MA, USA (2017), http://hdl.handle.net/1721.1/112381

[11] D.P. Griesheimer, W.R. Martin, J.P. Holloway, Journal of Computational Physics **211**, 129 (2006)

[12] D.P. Griesheimer, Ph.D. thesis, University of Michigan (2005)

[13] P.K. Romano, N.E. Horelik, B.R. Herman, A.G. Nelson, B. Forget, K. Smith, Annals of Nuclear Energy **82**, 90 (2015), joint International Conference on Supercomputing in Nuclear Applications and Monte Carlo 2013, SNA + MC 2013.

[14] F. Brown, W. Martin, ANS Mathematics and Computation Topical Meeting (2003)
[15] J. Leppänen, Nuclear Science and Engineering **174**, 318–325 (2013)
[16] T. Viitanen, J. Leppänen, Nuclear Science and Engineering **171**, 165–173 (2012)
[17] J.D. Galloway, V.J. Lawdensky, D.I. Poston, H.R. Trellue, M.E. Blood, Tech. rep., Los Alamos National Lab.(LANL), Los Alamos, NM (United States) (2020)
[18] K.A. Woloshun, M.A. Merrigan, E.D. Best (1988)
[19] G. Giudicelli, A. Lindsay, L. Harbour, C. Icenhour, M. Li, J.E. Hansel, P. German, P. Behne, O. Marin, R.H. Stogner et al., SoftwareX **26**, 101690 (2024)
[20] E. Shemon, Y. Miao, S. Kumar, K. Mo, Y.S. Jung, A. Oaks, S. Richards, G. Giudicelli, L. Harbour, R. Stogner, Nuclear Science and Engineering **197**, 1656 (2023), https://doi.org/10.1080/00295639.2022.2149231
[21] D.P. Adhikary, C. Jayasundara, R.K. Podgorney, A.H. Wilkins, International Journal for Numerical Methods in Engineering **109**, 218 (2016)
[22] M.N. Cinbiz, C.N. Taylor, E. Luther, H. Trellue, J. Jackson, Nuclear Technology **209**, S136 (2023), https://doi.org/10.1080/00295450.2022.2121583
[23] G.L. Giudicelli, A. Abou-Jaoude, A.J. Novak, A. Abdelhameed, P. Balestra, L. Charlot, J. Fang, B. Feng, T. Folk, R. Freile et al., Nuclear Science and Engineering **197**, 1 (2023)
[24] N.E. Stauff, K. Mo, Y. Cao, J.W. Thomas, Y. Miao, C. Lee, C. Matthews, B. Feng, Proceedings of the American Nuclear Society Annual 2021 Meeting (2021)