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INTRODUCTION

Nuclear microreactors promise to open new markets for nuclear industry. This expectation is due to their potential competitive cost in non-traditional market segments, e.g., mines, forward military basis, extraterrestrial surfaces and remote areas, and inherently safe characteristics that make them deployable where other power sources are not available or difficult to exploit. Transition metal dihydrides have been considered among the most promising candidates for moderating nuclear microreactors. In particular, yttrium hydride (YH_x) has been selected for high temperature applications due to its high thermal stability and relatively high hydrogen retention at temperatures exceeding $870°C$. One of the main issues associated with the use of hydrides is that, when exposed to temperature, stress, or concentration gradients, the hydrogen contained in the metallic matrix tends to redistribute and leak from the moderating elements, potentially leading to reactivity losses and power swings.

The purpose of this paper is to gain a better physical understanding of the neutronic feedback associated with the hydrogen redistribution in YH_x, by using Griffin [1] and Bison [2]. This feedback is inherently multiphysics, since the hydrogen distribution is strongly dependent on the moderator temperature spatial distribution. In particular, we want to understand the sign (+/-) of the neutronic feedback, its order of magnitude, and its underlying physical causes. To perform this task, the extruded unit-cell of the "Empire" reactor concept was used [3]. The model selected is one of the simplest computational models containing enough information to capture the primary design characteristics of proposed heat-pipe-cooled monolithic microreactors.

THEORY

Hydrogen Redistribution

Three driving forces determine the redistribution of hydrogen within the YH_x bulk: (1) Fickian diffusion, redistributing hydrogen from regions of high concentration to regions of low concentration, (2) thermal diffusion (the Soret effect), redistributing the hydrogen from higher temperature zones to lower temperature zones, and (3) the stress gradients redistributing the hydrogen from high compression to high tensile regions. The superposition of these three driving forces determine a net hydrogen current in the hydride volume, here denoted as $\boldsymbol{J}_H$ and defined as [4]:

$$\boldsymbol{J}_H = -D\left[\nabla C + \frac{QC}{RT^2}\nabla T - \frac{CV_H}{RT}\nabla\sigma\right], \tag{1}$$

where:

- $D[m^2/s]$ is the hydrogen diffusion coefficient in the metallic matrix.
- $C[mol/m^3]$ is the hydrogen/metal ratio (i.e., H/Zr and H/Y).
- $Q[J/mol]$ is the heat of transport.
- $R[J/molK]$ is the universal gas constant.
- $T[K]$ is the absolute temperature.
- $V_H[m^3/mol]$ the partial molar volume of hydrogen.
- $\sigma[MPa]$ is the applied stress.

For solids, the stress-related term is negligible due to its lower magnitude when compared to the Soret and Fickian contribution [5]. Leveraging the definition of the net hydrogen current, the following partial differential equation can be used to determine the space-time evolution of the hydrogen concentration in the hydride:

$$\frac{\partial C}{\partial t} = -\nabla \cdot \boldsymbol{J}_H. \tag{2}$$

Assuming the stress-caused driving force negligible and assuming null leakage of hydrogen from the surface, Eq. 2 admits the following asymptotic solution (*i.e.,* $t \to \infty$):

$$C_\infty = C_0 \ \exp\left(\frac{Q}{R\,T}\right), \tag{3}$$

where C_0 can be computed from the initial solution and the conservation of the hydrogen mass. It is noticeable that the temperature profile uniquely determines the hydrogen concentration in the hydride for fixed initial conditions, thereby highlighting the tight coupling between heat transfer and hydrogen migration. To determine the temperature in the current model, we are using the standard heat diffusion equation:

$$\rho c_p \frac{\partial T}{\partial t} = \nabla \cdot \lambda \nabla T + S \tag{4}$$

where, ρ and c_p denote the mass density and the specific heat, respectively; λ is the hydride's conductivity, and S is the power density determined by the neutronic solver.

Material properties for hydrogen migration

Two material properties of interest to describe the hydrogen redistribution in the absence of mechanical stresses are: (1) the diffusivity of hydrogen in the YH binary system and (2) the heat of transport determining the Soret contribution. In this work, the following YH_x diffusion coefficient is used:

$$D[m^2/s] = D_0 \exp\left(\frac{-A}{RT}\right), \tag{5}$$

where $D_0 = 10^{-8}[m^2/s]$ and $A = 0.38[eV/mol]$. Eq. 5 is a conservative value for temperature below 1000 K, analogous

to what was used in Ref. [6]. The value was chosen based on the absence of a consistent set of concentration-dependent data for the pre-exponential term, D_0 and the activation energy, A, in Eq. 5.

The heat of transport is unknown for YH_x. In absence of a better experimental result, this paper assumes it to be equal to that of zirconium hydride due to the similarities between the two materials.

$$Q = 5300[J/mol]. \tag{6}$$

This approximation was first made in Ref. [7] and will be utilized throughout the results section.

NUMERICAL RESULTS AND ANALYSIS

Problem Description

The Empire reactor 3D unit-cell is depicted in Fig. 1. It is an extruded 60-cm high triangular cell containing a $YH_{1.8}$ moderator, UN fuel, and sodium heat pipes in a stainless steel (SS316) monolith. The unit-cell is surrounded by a 10-cm axial graphite reflector. Vacuum axial boundary conditions and reflective radial boundary conditions are applied to the geometry. From a thermal perspective, the heat pipes are instead modeled as isothermal sodium at 600 K with heat-transfer coefficient equal to $1e4\ W/K/m^2$. Finally, we assume no leakage of hydrogen from the moderator boundary, therefore resulting into an homogeneous Neumann boundary condition for the hydrogen concentration.

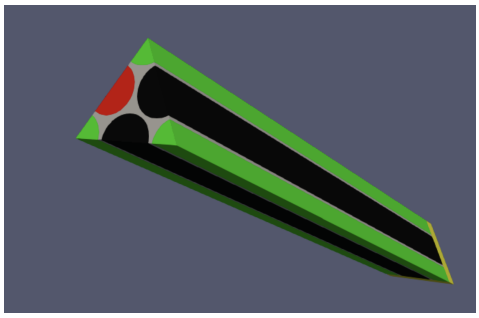

Fig. 1. "Empire" reactor concept unit-cell. The UN fuel corresponds to the red pin, (2) the black pins are the moderator blocks, and (3) the green sectors are the heat pipes

This Empire design concept was originally conceived to be generic enough to avoid the disclosure of proprietary data, while being specific enough to capture the primary design characteristics in proposed heat pipe cooled microreactor. The full description of the geometry and properties can be found in Ref. [3].

Codes and Computational Environment

Three codes were utilized to obtain the results in Sections – .

- Serpent 2 (V 2.1.31) is a Monte Carlo continuous-energy particle transport code developed at the VTT Technical Research Centre of Finland [8]. A Serpent model of the Empire assembly was used to compute the multigroup cross sections using a 6-groups energy structure taken from Ref. [3]. The ENDF-VIII.0 library was utilized to capture the effect of $S(\alpha,\beta)$ libraries in yttrium hydride [9]. The cross sections are parametrized in terms of fuel temperature, $T_f(\boldsymbol{x})$, moderator temperature, $T_m(\boldsymbol{x})$, and stoichiometric ratio, HY. It must be here noticed that the parametrization of the macroscopic cross sections to capture the evolution of the hydrogen content in YH_x is an open topic of research. The technique used in this work does not conserve the mass of hydrogen between state points, therefore constituting a source of inaccuracy. Ongoing studies are being devoted to enhancing the parametrization technique to correctly capture the change in hydrogen distribution within the hydride.
- Bison is a MOOSE-based nuclear fuel performance code able to solve the coupled equations of thermomechanics and species diffusion. In this paper, it was utilized to solve the heat diffusion equation coupled with the hydrogen redistribution equation.
- Griffin is a MOOSE-based neutronics code designed to solve the neutron transport equation and its approximation. In this paper, the CFEM-Diffusion solver is used to compute the eigenvalue and the corresponding fission source spatial distribution.

Multiphysics coupling is performed through Picard iteration. The power density spatial profile computed with the CFEM-Diffusion solver, here denoted as $P(\boldsymbol{x})$, is provided to Bison by internal data transfer, from which the corresponding fuel, moderator temperature profile, and hydrogen distribution are obtained. The cross sections are then updated based on the value of temperature and stoichiometric ratio. The latter are provided to Griffin and an updated power profile is obtained. Iterations are performed until the L-2 relative residual norm between the initial and latest iterations falls below a specified tolerance.

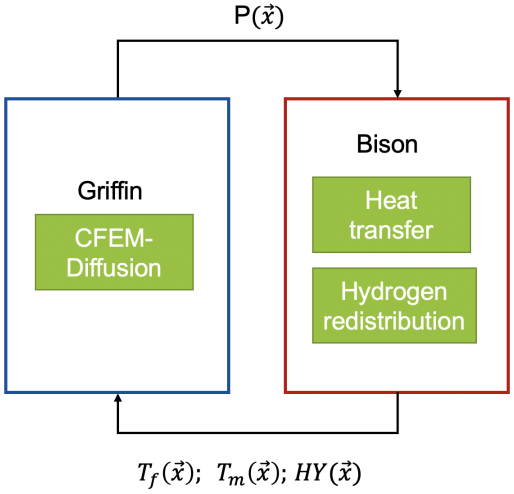

Fig. 2. Picard iteration scheme between Bison and Griffin.

Hydrogen Redistribution Feedback

In this paper, the hydrogen redistribution feedback (HRF) is defined as the difference in multiplication factor, here denoted as k, between the system configuration characterized by flat hydrogen stoichiometric ratio, HY_0, and the one characterized by asymptotic hydrogen spatial distribution, HY_∞. In formula:

$$HRF = k(HY_\infty) - k(HY_0) \tag{7}$$

The HRF of the unit cell is -555 pcm with the multiplication factor changing from 9.206e-01 to 9.1505e-01, as shown in the second column in Tab. I. This is due to the migration of hydrogen toward colder zones of the unit cell, and therefore, spatial zones characterized by lower neutronic importance. The redistribution of hydrogen is evident from Fig. 3, where the moderator temperature and the hydrogen stoichiometric ratio are reported as a function of the axial coordinate for the middle point of one of the moderator rods. It is observed that the redistribution due to the Soret effect leads to an accumulation of hydrogen at the axial periphery of the unit cell. This is substantiated numerically by columns 3–5 of Table I, where the average and extreme values of the stoichiometric ratio are reported. We can observe that the H/Y ratio minimum and maximum pass from 1.8 to 1.71 and 1.97, respectively. Additionally, it is noticeable that the asymptotic hydrogen spatial distribution is inversely proportional to the temperature profile, as predicted by Eq. 3. A final observation is that the redistribution mechanism is a catalyzing mechanism for the leakage of hydrogen from the surface of the moderator (not modeled in the current work). This is because, to a first approximation, the current from the interface to the surrounding environment is proportional to the hydrogen concentration on the surface [6].

TABLE I. Comparison between case without hydrogen redistribution (w/o hyd. red) and the case including hydrogen redistribution (w/ hyd. red).

Case	k	HY^{ave}	HY^{max}	HY^{min}
w/o hyd. red.	9.2060e-01	1.80	1.80	1.80
w/ hyd. red.	9.1505e-01	1.80	1.97	1.71

Moderator Temperature Feedback (MTC)

As is clearly shown in Eqs. 1–2, the hydrogen migration is strongly correlated to the change in moderator temperature distribution and magnitude, and, therefore, to the moderator temperature coefficient (MTC). This section is devoted to finding insight into the mechanisms driving the MTC in YH.

A conservative estimate of the MTC was computed by using the Serpent 2 assembly model. The temperature of the moderator was varied from 300 K to 1200 K and the MTC was computed with the equation:

$$\delta k[pcm/K] = \frac{k(1200K) - k(300K)}{k(300K)\ 900K} \times 10^5 \tag{8}$$

The temperature-related change in volume and density

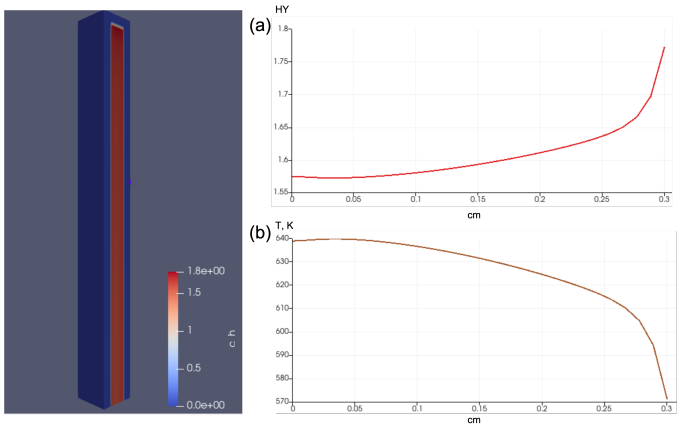

Fig. 3. (a) Hydrogen spatial profile and (b) Temperature spatial profile as a function of the distance from axial midpoint.

due to the temperature change were negligible for the analysis at the assembly and unit-cell level. The computed result was $+5.44 pcm/K$ or, equivalently, that $4890 pcm$ was gained by increasing the temperature of the moderator from $300K$ to $1200K$. To put this result into perspective, for an analogous change in fuel temperature, the Doppler coefficient is $-1.48 pcm/K$. The driving cause of the MTC positivity is the decrease of the absorption in the moderator due to the change in temperature, and the consequent increase in the thermal utilization factor. The decrease in absorption is due to the shift of the neutron flux thermal peak. In fact, the latter moves towards higher values of the energy axis where the yttrium microscopic absorption is lower. This is shown in Figs. **??**.a–c, where the flux spectrum per unit lethargy and the microscopic absorption cross section for yttrium are reported as a function of the neutron energy. The shift of the thermal peak when the moderator temperature increases from 300 K to 1200 K is apparent therein. This, in conjunction with the decrease of the yttrium microscopic absorption cross section, leads to a lower absorption rate in the moderator, thus leading to the increase of the infinite multiplication factor k_∞.

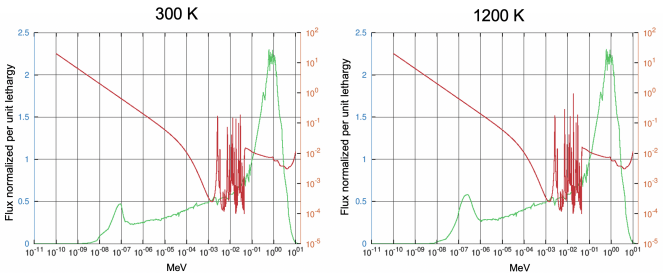

Fig. 4. (a) Absorption cross section of yttrium hydride as a function of the incident neutron energy at 300 K and 1200 K. (b) Absorption cross section of zirconium hydride as a function of the incident neutron energy at 300 K and 1200 K.

The argument is substantiated numerically in Table II, where the neutron balance is reported for the empire reactor unit cell. Using the 6-factor formula, it is clear that the main cause for the change in reactivity is the variation in the thermal

TABLE II. Moderator feedback coefficient

Factor	300 K	1200 K	δ_r, %
Reproduction Factor	1.99E+00	1.97E+00	-9.14E-01
Thermal Utilization Factor	6.02E-01	6.82E-01	**13.2E+01**
Resonance Escape Probability	4.01E-01	3.92E-01	-2.51E+00
Fast Fission Factor	2.42E+00	2.31E+00	-3.98E+00
Non-leakage Probability	8.61E-01	8.60E-01	3.23E-02

utilization factor (+13.2%). This is primarily due to a decrease in absorption in the regions outside the fuel when temperature increases. Among these, the main contributor is the change in absorption in the moderator, where the capture decreases by 26.0%. A large contributor to the change in reactivity is also given by the variation of the temperature in the monolith, -10%.

It must be noted that the MTC computed here is a conservative estimate and that in the context of full-core multiphysics simulations, its value will be lower. This is mainly caused by two effects not currently accounted for: (1) The effect of radial leakage, and (2) The contribution of the density and expansion due to the temperature increase. Further analyses will be performed to understand the full set of factors influencing the MTC for YH_x.

CONCLUSIONS AND FUTURE WORK

This paper presents preliminary results and observations related to the reactivity feedback caused by the hydrogen redistribution in YH_x under temperature and concentration gradients. The extruded unit cell of the Empire reactor concept was used as an assessment problem to mimic typical conditions envisioned in monolithic microreactors. It was found that the redistribution of hydrogen results in negative reactivity feedback on the order of hundreds of pcm. This is caused by the redistribution of the hydrogen towards lower temperature zones that are characterized by lower neutron importance. This paper also discusses the moderator temperature feedback associated to the use of YH_x. The latter was found equal to $5.44 pcm/K$ for the problem considered. The dominant mechanism contributing to the strong positive reactivity feedback was the decrease in moderator absorption rate. This was caused by the shift in the thermal peak spectrum and the simultaneous decrease of the yttrium absorption cross section with the energy of the colliding neutron. The work presented in this paper is a starting point for a more thorough analysis performed at the core level and considering both the effect of thermomechanics and radial neutron leakage. Additionally, ongoing analysis is being performed to accurately capture the hydrogen redistribution effects through alternative cross sections parametrization choices aimed at conserving the mass of hydrogen between state points.Future work will also be devoted to investigate alternative moderators for microreactors applications, such as ZrH, ^{7}LiOH, and NaOH, to compare their performance from a hydrogen redistribution standpoint.

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