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Preliminary Modeling of Hydrogen Diffusion, Desorption, and Adsorption in YH For Micro-Reactors Applications

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Preliminary Modeling of Hydrogen Diffusion, Desorption, and Adsorption in YH For Micro-Reactors Applications

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Micro-reactor moderator: Metal hydrides (e.g., YH) are good candidate for moderation of nuclear microreactor due to their high hydrogen concentration at high temperature and good thermal properties.

Problem: Models to describe hydrogen diffusion, desorption, and adsorption are still under investigation.

Goal: Preliminary modeling of hydrogen diffusion, desorption, and adsorption for YH based on experiments. The model developed in Bison will then be used in coupled multiphysics simulation to get preliminary results for a prototypical microreactor.

❖Experimental set-up [1]: Pure yttrium samples (0.142 cm x 1.27 cm x 1.3 cm) are inserted into a reaction tube with fix temperature (no gradient of temperature) and pressure. Adsorption of hydrogen into the yttrium is measured.

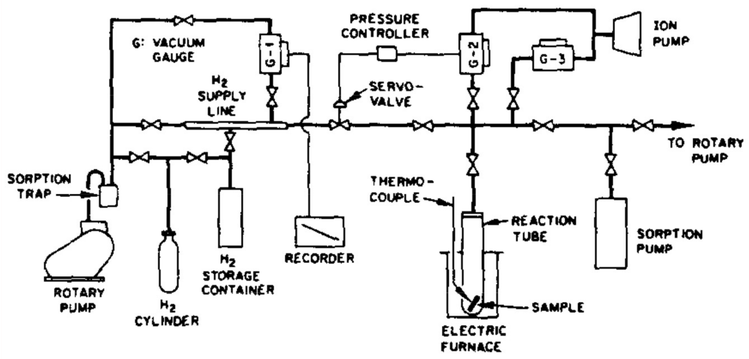

Figure 1. Experimental set-up [1].

❖Physics model [1, 2]:

$$\frac{\partial C_H}{\partial t} = \nabla \cdot \left[-D\left(\nabla C_H + \frac{Q C_H}{RT^2}\nabla T\right)\right].$$

$$D = D_0 E^{-\frac{E_{ad}}{RT}},$$

At the surface: $\vec{j}.\vec{n} = -k \cdot \left(C_H - C_{eq}\right)$,

C_H concentration of hydrogen in the hydride, D_0 diffusion coefficient, E_{ad} activation energy, k reaction rate, $\vec{j}$ flux of hydrogen, $\vec{n}$ normal to the surface, C_{eq} equilibrium concentration (function of the gas pressure and temperature, fixed here).

➢Reaction rate (k) simple model developed:

$$k(T,\ C_H) = k_0 \cdot E^{-\frac{E_a}{RT}},$$

with E_a activation energy, k_0 positive constant.

➢A second more complex model was also investigated:

$$k(T,\ C_H) = E^{-\frac{E_a}{RT}} \cdot \left(k_0 + k_1 \cdot e^{-k_2 \frac{C_H}{C_{eq}}}\right),$$

with E_a activation energy, k_i positive constant.
The exponential term should provide some correction on the reaction rate as more adsorption happens.

❖ Model fitting process:

➢ Using Bison, the values for the reaction rate were determined to reproduce the experimental results at five different temperatures (963 K, 1001 K, 1061 K, 1111K, 1161K).

➢ Then, a fitting of both models using these values were realized.

➢ The final models were then implemented in Bison and the results obtained were compared to the experimental values.

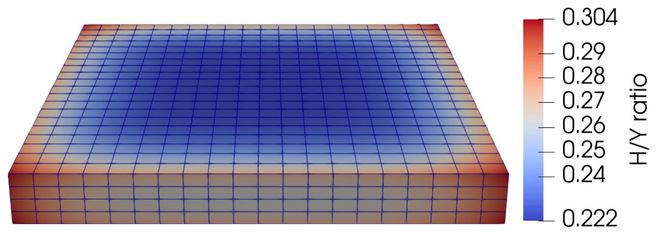

Figure 1. H/Y ratio at a temperature of 963 K and t = 2800 s.

❖ Results:

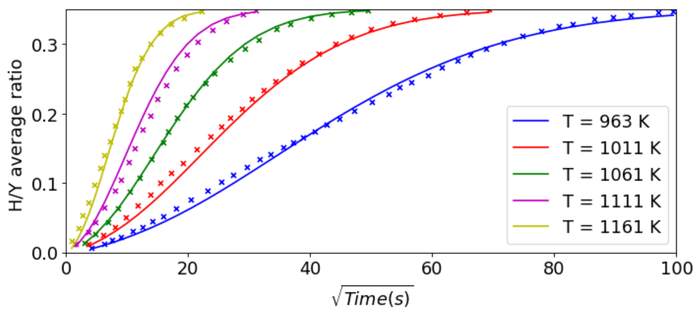

Figure 2. Temporal evolution of the average stochiometric ratio at different temperatures. The solid line are BISON results using the simple model, and the crosses are experimental values.

➢ Simple model:
k_0 = 45.2363 m/s, E_a = 152154 j/mol.

Table 1. L2 norm comparison between experimental value and BISON results obtained with the simple model.

Temperature (K)	963	1011	1061	1111	1161
e (%)	3.08	4.04	1.23	5.97	5.43

➢ The more complex model leads to a slight increases of accuracy of the results. Therefore, the simple model is preferred.

Table 2. L2 norm comparison between experimental value and BISON results obtained with the more complex model.

Temperature (K)	963	1011	1061	1111	1161
e (%)	2.38	3.03	1.22	5.14	6.04

❖ Conclusion:

Good agreement between 963 K and 1161 K. At higher temperature, the model accuracy due to the onset of temperature-driven dependencies not captured by an Arrhenius-type law.

❖ A similar process of model fitting was realized to determine the relationship between C_{eq}, temperature, and H2 partial pressure using experimental data of [3].

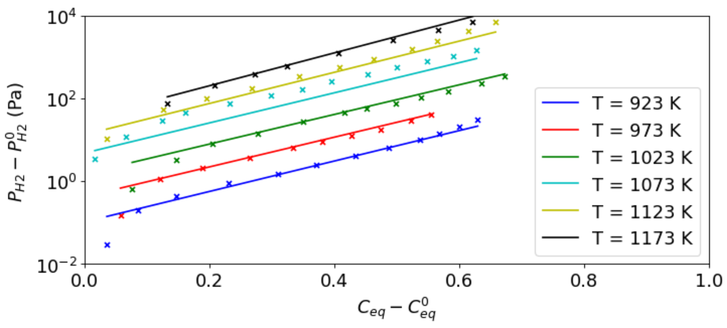

Figure 3. Fitting of the differential gas pressure as function of the the differential stochiometric ratio and temperature using a model implemented in Bison. Solid line: BISON results, crosses: experimental values

❖ Moderator pin modeling: a Bison model was created using all the models developed. It was then used to model a fuel pin in 2D – RZ symmetry.

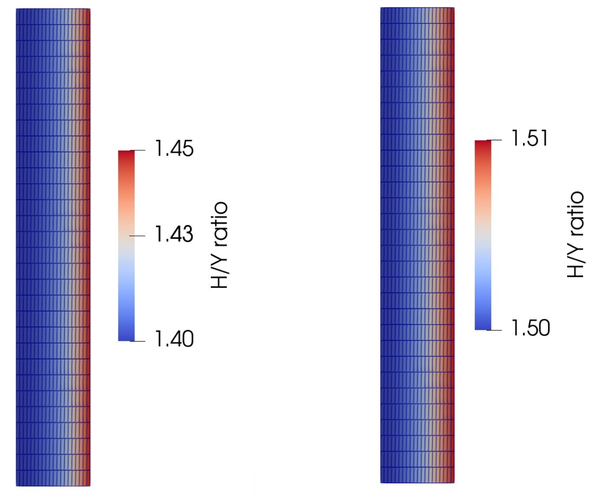

Figure 4: Moderator pin H/Y ratio at t = 100 s (left) and t = 1000 s (right) for a simulation at fixed gap pressure. The radial dimensions have been scaled up by a factor of 5.

References:
[1] P. W. FISHER and M. TANASE, "Diffusivities Of Hydrogen In Yttrium And Yttrium Alloys", Journal of Nuclear Materials 122 -123, (1984), 1536-1540.
[2] J. HUANGS , B. TSUCHIYA , K. KONASHI, M. YAMAWAKI, "Estimation of Hydrogen Redistribution in Zirconium Hydride under Temperature Gradient", Journal of Nuclear Science and Technology, 37, 10 (2000).
[3] F. KAI, et al., “Experimental Investigation and Thermodynamic Assessment of the Yttrium-Hydrogen Binary System”, Progress in Natural Science: Materials International, 28 (2018), 332-336.