[I] OVERVIEW

Pressurized water reactors (PWRs) are the most common reactor design in the world. PWRs use water as both coolant and moderator. Zirconium (Zr) alloys are used as the fuel cladding material due to their corrosion resistance and low thermal neutron capture cross section. However, under the harsh reactor environment and high temperatures, the alloy does corrode. The corrosion reaction produces Hydrogen as both coolant and moderator. Zirconium (Zr) is a low solubility limit (60 ppm) in Zr. It also tends to embrittle the zircaloy and can cause delayed hydride cracking (DHC), which is a result of a H assisted phase transformation within the metal. Fuel cladding failures impact on both the safety and economics of the fuel cycle of nuclear reactors. This project is focused on modelling DHC with input from first principles calculations.

[II] RESEARCH SUMMARY

• The purpose of this work is to develop a model for stress driven diffusion of H to crack tips. This is significant to the nuclear industry particularly with regard to improving the safety and lifetime of the fuel pin.
• A first principles approach has been used to calculate the relaxation of atomic H in tetrahedral interstitial in Zr.
• Using these first principles data, we have calculated the elastic interaction energy between atomic H and edge dislocations in a way that respects the symmetry of the local atomic environment. This is in contrast to a misfitting sphere model of the elastic interaction.
• In this work we have shown that the two methods for calculating interaction energies are equivalent, only when the relaxed atomic environment has spherical symmetry. This approach is significant in its multiscale nature, combining atomic-scale data with linear elasticity theory.
• A stress driven diffusion equation has been derived, which incorporates the new expression for the elastic interaction energy. The diffusion equation has been implemented to determine the time evolution of H in the presence of a single dislocation.

[IV] RESULTS

A tetrahedral interstitial in Zr: the force on the three basal sites are identical, the force exerted along the c-axis is different because the point is not related to the others by any symmetry operation.

The elastic interaction driving diffusion is captured in the first two terms of the diffusion equation:

\[ \frac{\partial c_H}{\partial t} = \frac{D}{k_B T} \left( c_H(r, t) \left( \nabla E_{int}(r) + c_H(r, t) \nabla^2 E_{int}(r) \right) + k_B T \nabla^2 c_H(r, t) \right) \]

The steady state solution to the diffusion equation is:

\[ c_H(r, t) = \frac{c_H^0}{1 + k_B T \nabla^2 c_H(r, t)} \]

\[ \frac{c_H(r, t) \nabla^2 c_H(r, t)}{k_B T} \]

[VI] CONCLUSIONS

• The defect forces of H on Zr atoms in a tetrahedral site have been determined through DFT calculations.
• When the defect forces are fully symmetric, the resulting elastic interaction profile agrees with that calculated using the misfitting sphere model.
• Using our novel approach to defect force calculations, we have shown the elastic interaction energy profile for atomic H in its most stable tetrahedral site in Zr. Our approach captures the full symmetry of the local atomic environment of Zr, which has a non-ideal c/a ratio.
• We have developed a stress driven diffusion model into which the elastic interaction energy is incorporated, and have shown that the steady-state solution to the diffusion equation also captures the symmetry of the atomic environment of Zr.
• The significance of this work is that we have used a multi-scale approach to determine the evolution of H within Zr in the presence of stress concentrations, with the possibility of extending the work to modelling diffusion to crack tips and the first stage of DHC.

REFERENCES


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Prerequisites to obtaining a model that replicates the primary stages of DHC include understanding the relationship between H and Zr on the atomic scale, incorporating H interactions with line defects and determining H-H interactions.

The behaviour of H in Zr is studied using DFT and results for the volume dilatation, and forces exerted on neighbouring atoms by hydrogen are presented. The hydrogen-dislocation interaction can thus be determined analytically. The effect of dislocation screening by hydrogen is determined and the resulting reduction in dislocation spacing is shown.

The scope for future work using microscale modelling methods such as Discrete Dislocation Dynamics are presented.

**FIGURE 3**

Zirconium has a hexagonal close-packed structure, with a c/a ratio of 1.593. There are five interstitial sites in which a solute atom might sit. For hydrogen, the most energetically favourable site is the Tetrahedral.
REFERENCES


FUTURE WORK

• Incorporate a stress driven diffusion equation for hydrogen into the DDD model.
• Use DDD simulation technique to model the effects of hydrogen on plasticity and vice versa.
• Assess the role of dislocations as possible solute traps and sites for hydride nucleation

RESULTS

The interaction energy is calculated by combining the Kanzaki forces with the displacement field of the prismatic edge dislocation. The concentration profile is then determined using Fermi-Dirac distribution function. The results are shown in Figures 5 and 6.

CONCLUSIONS

We have found that there is an elastic interaction between hydrogen and an edge dislocation, and that hydrogen has a tendency to accumulate in the tensile region of a dislocation. As more edge dislocations are placed in close proximity, their elastic fields results in a greater accumulation of hydrogen and so the screening between dislocations can be significant. Low temperatures and high concentrations of hydrogen are required to provide sufficient dislocation screening, and since concentrations in the elastic field of a dislocation can reach the solvus we find that dislocations can act as nucleation sites for hydride phases.

FUTURE WORK

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REFERENCES


