An Elastoplastic Phase Field Model
for Morphological Evolution of Hydrides in Zirconium

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Summary

The linear elastic phase field model on γ-hydride morphological evolution in zirconium is extended to the case of perfectly elastoplastic matrix around a hydride. The temporal evolution of the spatially dependent field variables is determined by numerically solving the time-dependent Ginzburg-Landau (TDGL) equations for the structure variables and the Cahn-Hilliard diffusion equation for the composition variable. In addition, a new set of time-dependent equations for plastic deformation variables is introduced to describe plastic zones around hydrides without pre-assumption on microstructure morphology and shape of plastic zone along the evolution path. This new development extends our capability of simulating much more types of phase transformations, as well as opens up opportunities for simulating cracks and notches that involve plastic deformation.

Introduction

Zirconium and its alloys are primary structural materials in the nuclear power industry owing to its combination of good mechanical properties, excellent corrosion resistance and low neutron absorption cross-section. However, zirconium and its alloys suffer a strength degradation due to hydride formation under service condition in reactors. It is found that the critical crack initiation and propagation at hydrides in zirconium are controlled by the morphology, orientation and distribution of hydride precipitates.

Recently, Shi and co-worker [1-4] have published a phase-field model based on the linear elasticity theory of Khachaturyan [5] to investigate the morphological evolution of γ-hydride precipitation under an applied load in single crystalline, bi-crystalline, or polycrystalline zirconium. These investigations are the first quantitative description of complex hydride formation patterns in metals. However, the theory is not a complete one because of the lack of plastic deformation during hydride precipitation. On the other hand, it is well known that hydride formation in zirconium involves about 15% volume expension, therefore, significant plastic deformation. The effect of plastic

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deformation has to be studied, particularly when stress concentrators, such as cracks and blunt flaws are present. The major difficulty in developing such a theoretical phase-field model is to account for the presence of arbitrary-shaped plastic zone around hydrides that produces image and screen forces. This report describes an elastoplastic phase field theory which models hydrogen diffusion and γ -hydride precipitation coupled with the elastoplastic deformation of zirconium.

Methodology and Results

In the phase field model, an arbitrary multiphase microstructure is described by a set of spatially dependent field variables. A coherent precipitate with a point symmetry reduction usually produces a number of variants which are oriented in different, but equivalent crystallographic directions. For example, γ - hydride precipitate in a hexagonal zirconium or zirconium-rich alloy has a face-centered tetragonal structure as a result of high rates of cooling. One conservative concentration variable $c(r,t)$ and three non-conservative long-range structural order variables $\eta_1(r,t)$, $\eta_2(r,t)$, $\eta_3(r,t)$ are adopted to describe hydride phase transformation. The former distinguish the hydrogen difference between the precipitate and the matrix, while the latter distinguish the structural difference between the precipitate and the matrix. Across the interface boundaries, these field variables change gradually from one phase to another within finite thickness.

According to thermodynamic principle, the equilibrium state of a multiphase microstructure corresponds to the minimum free energy. The driving force for the temporal evolution of a coherent microstructure consists of the following: (a) reduction in the bulk chemical free energy; (b) the decrease in the total interfacial energy of the boundaries between different phases or between differently oriented orientations; (c) the relaxation of strain energy caused by lattice mismatch between the precipitate and the matrix; (d) the interaction energy between hydride formation/dissolution and external loads.

For a coherent precipitate, an important contribution to the system's total free energy is the elastic strain energy caused by the lattice mismatch between hydride and matrix. In general, the degree of lattice mismatch between precipitate and matrix is characterized by the stress-free transformation strain which is nonzero only within the precipitates and can be represented as

$$\varepsilon_{ij}^0 = \frac{a_p - a_m}{a_m} \tag{1}$$

where a_p and a_m are the lattice parameters of a precipitate variant p and matrix phase respectively. In the diffuse-interface description, we can defined transformation strain field related to long-range order parameter and hydrogen concentration through

$$\varepsilon_{ij}^0(r) = \sum_{p=1}^{3} \varepsilon_{ij}^\eta(p)\eta_p^2(r) + \varepsilon_{ij}^c \delta c(r) \tag{2}$$

where $\eta_p(r)$ is the normalized structural order parameter describing the p th variant and $\varepsilon_{ij}^\eta(p)$ is the corresponding stress-free strain for the p th variant when $\eta_p(r) = 1$. ε_{ij}^c is the strain caused by the expansion of hydrogen atom. $\delta c(r)$ is hydrogen concentration difference related on average hydrogen concentration in zirconium matrix.

In classical metal plasticity theory, the yield stress is independent of hydrostatic stress. The von-Mises equivalent stress

$$\sigma_s^2 = 3J_2' = \frac{3}{2}\sigma_{ij}'\sigma_{ij}'$$

$$= \frac{1}{2}\left[(\sigma_{xx} - \sigma_{yy})^2 + (\sigma_{yy} - \sigma_{zz})^2 + (\sigma_{zz} - \sigma_{xx})^2\right] + 3\sigma_{xy}^2 + 3\sigma_{yz}^2 + 3\sigma_{zx}^2$$

For plain strain, $\sigma_{zz} = \nu(\sigma_{xx} + \sigma_{yy})$. Yield criterion is $\sigma_s = \sigma_Y$ (yield stress measured in tensile tests). The yield potential is then defined

$$F = \frac{1}{2}\left[(\sigma_{xx} - \sigma_{yy})^2 + (\sigma_{yy} - \sigma_{zz})^2 + (\sigma_{zz} - \sigma_{xx})^2\right] + 3\sigma_{xy}^2 + 3\sigma_{yz}^2 + 3\sigma_{zx}^2 - \sigma_Y^2$$

Based on strain energy relations, yield criteria in isotropic media can also be written as

$$F = E_{el}' - \frac{\sigma_Y^2}{(\lambda + 2\mu)} \tag{3}$$

where E_{el}' is the distortion strain energy. In this work, the elastic, perfectly plastic constitutive relation is assumed.

The distortion strain energy for the stress-controlled condition

$$E_{el}' = \int_v \frac{1}{2} C_{ijkl}\left[e_{ij}(r) - e_{ij}^0(r)\right]\left[e_{kij}(r) - e_{ij}^0(r)\right]d^3r$$

$$= \frac{1}{2}\int_v C_{ijkl} e_{ij}^0(r) e_{kl}^0(r) d^3r - \frac{1}{2V}C_{ijkl}\int_v e_{ij}^0(r)d^3r\int_v e_{kl}^0(r')d^3r'$$

$$- \frac{1}{2}\int \frac{d^3k}{(2\pi)^3} n_i \tilde{s}_{ij}^0(k)\Omega_{jk}(n)\tilde{s}_{kl}^0(k)^* n_l - s_{ij}^{appl}\int_v e_{kl}^0(r)d^3r - \frac{1}{2}S_{ijkl}s_{ij}^{appl}s_{kl}^{appl}$$

For isotropic media

$$E'_{el} = \int_v \frac{1}{2} \times 2\mu \left[e_{ij}(r) - e^0_{ij}(r) \right] \left[e_{ij}(r) - e^0_{ij}(r) \right] d^3r$$

$$= \int_v \mu e^0_{ij}(r) e^0_{ij}(r) d^3r - \frac{1}{V}\mu \int_v e^0_{ij}(r) d^3r \int_v e^0_{ij}(r') d^3r'$$

$$- \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} n_i \tilde{s}^0_{ij}(k) \Omega_{jk}(n) \tilde{s}^0_{kl}(k)^* n_l - s^{appl}_{ij} \int_v e^0_{kl}(r) d^3r - \frac{1}{4\mu} s^{appl}_{ij} s^{appl}_{kl}$$

where $e_{ij} = \varepsilon_{ij} - \frac{1}{3}\varepsilon_{kk}\delta_{ij}$.

The plastic deformation can be described by introducing a plastic strain ($\varepsilon^p_{ij}(r)$) as a new field variable [6]. Then the whole stress-free strain is $\varepsilon^0_{ij}(r) = \varepsilon^\eta_{ij}(r) + \varepsilon^c_{ij}(r) + \varepsilon^p_{ij}(r)$.

$\varepsilon^p_{ij}(r)$ can be obtained as a solution of the time-dependent Ginzburg-Landau Equation,

$$\frac{\partial \varepsilon^p_{ij}(r_{plas},t)}{\partial t} = -K_{ijkl} \frac{\delta E'_{el}}{\delta \varepsilon^p_{kl}(r_{plas},t)} \qquad (4)$$

$$\frac{\delta E'_{el}}{\delta \varepsilon^p_{kl}(r_{plas},t)} = -2\mu \left[\int \frac{d^3k}{(2\pi)^3} n_k \Omega_{lm}(n) \tilde{s}^0_{mn}(k) n_n e^{ik\cdot r} - e^0_{kl}(r) + \bar{e}^0_{kl} \right] - s^{appl}_{ij}$$

The above equation is only applied at plastic zone. A convenient choice for K_{ijkl} is $K_{ijkl} = KC^{-1}_{ijkl}$. Let

$$s_{ij}(r) = 2\mu \left[\int \frac{d^3k}{(2\pi)^3} n_k \Omega_{lm}(n) \tilde{s}^0_{mn}(k) n_n e^{ik\cdot r} - e^0_{kl}(r) + \bar{e}^0_{kl} \right] + s^{appl}_{ij}$$

Then, $\dfrac{\partial \varepsilon^p_{ij}(r_{plas},t)}{\partial t} = K_{ijkl} s_{kl}$, which is similar to classical Prandle-Reuss yield surface theory. The temporal evolution of hydrogen diffusion and hydride growth are determined by solving the above equation together with the time-dependent Cahn-Hilliard diffusion equation for $c(r)$ and Ginzburg-Landau equations for $\eta_p(r,t)$. Since the stress equilibrium is established much faster than the microstructure evolution, K is chosen as a number larger than L, M used in Cahn-Hilliard diffusion equation and in Ginzburg-Landau equations.
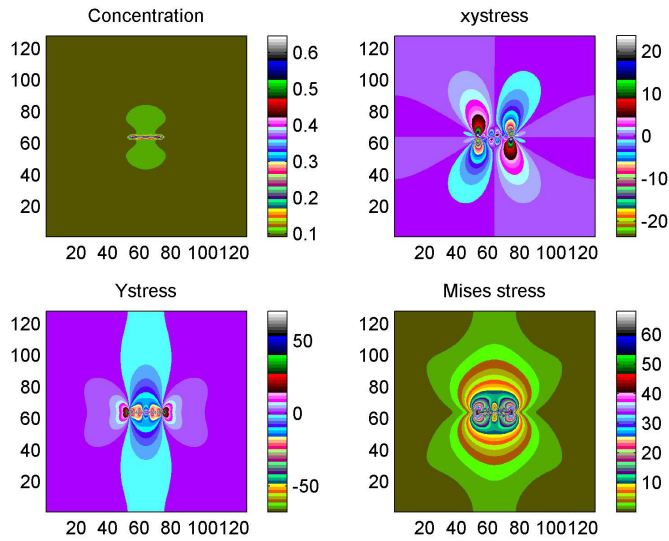
Figures in the end show typical simulation results on hydrogen concentration distribution and stress distributions around a single hydride precipitate, based on the elastoplastic phase field theory (upper 4 figures), which is significantly different as compared to linear elastic simulations (lower 4 figures). The methodology will be used to study the morphological evolution of hydride precipitation/dissolution when multiple hydrides and external loads are present. The same methodology can also be easily adopted to study crack initiation and propagation phenomena in engineering materials when crack-tip plastic zone is an important controlling factor.

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Elastoplastic phase field simulation at 10000 time steps

Linear elastic phase field simulation at 10000 time steps