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# A quantitative phase field model for hydride precipitation in zirconium alloys: Part II. Modeling of temperature dependent hydride precipitation

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## ABSTRACT

A quantitative free energy functional developed in Part I (Shi and Xiao, 2014) was applied to model temperature dependent  $\delta$ -hydride precipitation in zirconium in real time and real length scale. At first, the effect of external tensile load on reorientation of  $\delta$ -hydrides was calibrated against experimental observations, which provides a modification factor for the strain energy in free energy formulation. Then, two types of temperature-related problems were investigated. In the first type, the effect of temperature transient was studied by cooling the Zr–H system at different cooling rates from high temperature while an external tensile stress was maintained. At the end of temperature transients, the average hydride size as a function of cooling rate was compared to experimental data. In the second type, the effect of temperature gradients was studied in a one or two dimensional temperature field. Different boundary conditions were applied. The results show that the hydride precipitation concentrated in low temperature regions and that it eventually led to the formation of hydride blisters in zirconium. A brief discussion on how to implement the hysteresis of hydrogen solid solubility on hydride precipitation and dissolution in the developed phase field scheme is also presented.

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## 1. Introduction

The mechanical strength of metal hydrides is low, which causes great concern for structural integrity [2,3]. Zirconium alloys are nuclear materials that can suffer hydride embrittlement. About 5–20% of hydrogen produced by corrosion will migrate into the alloys. Hydrogen in solid solution diffuses to the tensile region at the flaws if there is a hydrostatic tensile stress gradient, or to the cooler part of materials if there is a temperature gradient. Complicated patterns of hydride precipitates can develop inside the alloys, which may lead to fracture even without an increase of mechanical load. A well-documented example of serious failure occurred in August 1983 in Unit 2 of Pickering A Nuclear Generating Station. In this case, a Zircaloy-2 pressure tube developed an approximately 2 m long axial crack due to the growth of hydride blisters at points of contact between the pressure tube and the cooler calandria tube surrounding it [4,5]. Failure due to hydride blisters has been a serious safety concern in other zirconium alloys such as Zircaloy-4

cladding in pressurized light water reactors (PLWR) in countries such as the USA, France and Japan [6–9], and Zr–2.5Nb pressure tubes in pressurized heavy water reactors (PHWR) in Canada, Argentina, India and South Korea [10–14]. Hydride embrittlement has not only occurred in zirconium alloys, but also in other important metals such as niobium [15], titanium [16], vanadium [17], and more recently in magnesium–aluminum alloy [18,19], the latter being a candidate in advanced automobile technology.

It is believed that the critical conditions for fracture initiation in hydrides are controlled by the morphology of hydride precipitates in the alloys [20–22]. For example, under nominally the same peak stress state, fracture may or may not initiate, depending on the distribution, density and orientation of the hydride cluster. The morphology of hydride precipitates is controlled by, among other factors, the crystal structure and mechanical properties of hydrides, the orientation of host metal crystals, the temperature distribution, and the orientation and magnitude of the applied stress. One of the stable hydride phases is  $\delta$ -hydride [23]. It has a face-centered cubic structure as compared to the hexagonal-close-packed structure of zirconium. It is known that the formation of  $\delta$  hydrides involves a large volume expansion of about 17% as compared to the original zirconium matrix, resulting in elastic

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and plastic deformations around the hydrides [23]. Such large elastic and plastic deformations during the formation and dissolution of hydrides are believed to be the reason for a significant hysteresis in hydrogen solid solubility limits found in zirconium [24,25]. It is also well-known that externally applied stress can cause re-orientation of hydride precipitates (see for example, [26]).

There are a large number of experimental observations and modeling studies in the literature on hydride precipitation and embrittlement. Nevertheless, a comprehensive, quantitative and multi-dimensional theory that handles all key processes such as hydrogen diffusion, hydride precipitation and fracture under temperature, stress and concentration gradients has not yet been accomplished. Early theoretical investigations considered the process of blister formation using only the concept of the thermal diffusion of hydrogen [27–29], neglecting the effects of volume changes during hydride formation/dissolution (i.e. elastic and plastic deformation). Recent efforts have included the effects of hysteresis on hydrogen solubility [13,30,31], and in some cases, researchers have also considered the effects of volume change due to hydrides by evaluating the percentage of hydrides in a given volume [12,32]. However, these models only estimate hydride volume fraction and cannot predict hydride morphology. Recent progress in phase-field modeling of hydride morphology provided great promise in developing a feasible theoretical and computation scheme that can handle all key processes in multi-dimensional space [33–39], while these studies are at best, still semi-quantitative: quantitative in stress-strain analysis and not quantitative in real time and length scales, or not quantitative in dealing with temperature transient and temperature gradient. Toward developing a comprehensive, quantitative and multi-dimensional phase field model, the Part I of this work [1] presented the development of a quantitative description of chemical free energy density and interfacial gradient coefficients that are a function of temperature and other materials parameters. This is a necessary step for a fully quantitative modeling of hydride precipitation. The developed free energy functional was applied to study  $\gamma$ -hydride precipitation in single crystal zirconium after high speed quenching, and the results were compared to TEM observations [40] with reasonable agreement in terms of average hydride size and density [1].

In this work, we will apply the developed free energy functional from Part I [1] to study the effect of temperature on  $\delta$ -hydride precipitation in zirconium. To the best knowledge of the authors, there is no phase field modeling of  $\delta$ -hydride in zirconium in the literature, even though the most prevalent hydrides in reactor conditions are  $\delta$ -hydride. In fact, the phase field framework developed in Part I should be applicable to both  $\gamma$  or  $\delta$ -hydride. We will first calibrate the developed model against limited experimental observations. Then, we will study two types of problems related to temperature: the effect of temperature transient and the effect of temperature gradient on  $\delta$ -hydride precipitation in zirconium.

## 2. Kinetic equations and quantitative free energy functional for hydride-Zr system

The kinetic equations in our phase field model are given as follows.

The Cahn–Hilliard diffusion equation with thermal diffusion,

$$\frac{\partial C}{\partial t} = \nabla \cdot \left( M \nabla \frac{\delta F}{\delta C} + \frac{DCQ^*}{RT^2} \nabla T \right) + \xi \quad (1)$$

The Allen–Cahn phase field equation

$$\frac{\partial \eta_p}{\partial t} = -L \frac{\delta F}{\delta \eta_p} + \zeta_p \quad (p = 1, 2, 3 \dots) \quad (2)$$

and the kinetic equation for plastic deformation – an Allen–Cahn type

$$\frac{\partial e_{ij}^{pl}}{\partial t} = -N_{ijkl} \frac{\delta E^{dis}}{\delta e_{kl}^{pl}} \quad (3)$$

In the above equations,  $C$  is hydrogen concentration in the unit of atomic percent,  $M$  is the mobility of hydrogen atoms in zirconium,  $F$  is the total free energy of the system,  $D$  is the chemical diffusion coefficient,  $Q^*$  is the heat of transport,  $R$  is the gas constant,  $T$  is the temperature,  $\eta_p$  ( $p = 1, 2, 3, \dots$ ) are long range order parameters representing the crystalline variants of the hydride phase,  $L$  is a relaxation coefficient,  $\xi$  and  $\zeta_p$  are noise terms satisfying the fluctuation–dissipation theorem,  $e_{ij}^{pl}$  are the plastic strains,  $E^{dis}$  is the distortion strain energy,  $N_{ijkl}$  are kinetic coefficients characterizing the plastic deformation rate, and  $t$  is the time.

The total free energy of the Zr–H system is given by

$$F = \int \left[ f(C, \eta_p) + \sum_p \frac{\kappa_p}{2} (\nabla \eta_p)^2 + \frac{\lambda}{2} (\nabla C)^2 + Modif \times E \right] dV \quad (4)$$

where  $f(C, \eta_p)$  is the local chemical free energy density,  $\kappa_p$  and  $\lambda$  are interface gradient coefficients,  $Modif$  is a modification factor that properly accounts for the weighting of the strain energy density  $E$  compared to other energy terms in the free energy functional. Ideally, this factor should be unity if all energy terms are completely accurate and inclusive, while in reality, this is not possible because the theoretical model may never be able to take all possible factors into account. For examples, some elastic properties of Zr and hydrides were obtained under ideal conditions, such as in stress-free neutron scattering tests for powder materials, while in engineering applications, Zr alloys often contain chemical inhomogeneity, crystalline defects and residual stresses. Therefore, it may be simpler to introduce the modification factor when calibrating the model against experimental observations. In the above equations,  $E$  and  $E^{dis}$  are given by [34,36]

$$E = \frac{1}{2} \int_V C_{ijkl} e_{ij}^0(\mathbf{r}) e_{kl}^0(\mathbf{r}) d^3 r - \frac{1}{2V} C_{ijkl} \int_V e_{kl}^0(\mathbf{r}) d^3 r \int_V e_{ij}^0(\mathbf{r}') d^3 r' \\ - \frac{1}{2} \int_V \frac{d^3 k}{(2\pi)^3} n_i \tilde{\sigma}_{ij}^0(\mathbf{k}) \Omega_{jk}(\mathbf{n}) \tilde{\sigma}_{kl}^0(\mathbf{k})^* n_l - \sigma_{ij}^a \int_V e_{ij}^0(\mathbf{r}) d^3 r \\ - \frac{V}{2} S_{ijkl} \sigma_{ij}^a \sigma_{kl}^a \quad (5)$$

$$E^{dis} = \frac{1}{2} \int_V C_{ijkl} e_{ij}^0(\mathbf{r}) e_{kl}^0(\mathbf{r}) d^3 r - \frac{1}{2V} C_{ijkl} \int_V e_{kl}^0(\mathbf{r}) d^3 r \int_V e_{ij}^0(\mathbf{r}') d^3 r' \\ - \frac{1}{2} \int_V \frac{d^3 k}{(2\pi)^3} n_i \tilde{s}_{ij}^0(\mathbf{k}) \Omega_{jk}(\mathbf{n}) \tilde{s}_{kl}^0(\mathbf{k})^* n_l - s_{ij}^a \int_V e_{kl}^0(\mathbf{r}) d^3 r \\ - \frac{V}{2} S_{ijkl} s_{ij}^a s_{kl}^a \quad (6)$$

where  $V$  is the system volume; the integral  $\int$  in infinite reciprocal space is evaluated as a principal value excluding the point  $\mathbf{k} = 0$ ;  $\Omega_{jk}(\mathbf{n})$  is the Green function tensor, which is the inverse of the tensor  $\Omega_{jk}^{-1}(\mathbf{n}) = n_i C_{ijkl} n_l$ ;  $S_{ijkl}$  is the elastic compliance tensor, which is the inverse to the elastic modulus tensor,  $C_{ijkl}$ ;  $\mathbf{n} = \mathbf{k}/k$  is a unit directional vector in reciprocal space;  $\tilde{\sigma}_{ij}^0(\mathbf{k}) = C_{ijkl} \tilde{e}_{kl}^0(\mathbf{k})$ ;  $\tilde{e}_{kl}^0(\mathbf{k})$  are the Fourier transforms of  $e_{kl}^0(\mathbf{r})$  (i.e.,  $\tilde{e}_{kl}^0(\mathbf{k}) = \int e_{kl}^0(\mathbf{r}) \cdot \exp(-i\mathbf{k} \cdot \mathbf{r}) d^3 r$ ); the superscript asterisk (\*) indicates the complex conjugate;  $\sigma_{ij}^a$  is the applied external stress;  $e_{ij}^0(\mathbf{r}) = e_{ij}^0(\mathbf{r}) - 1/3 e_{kk}^0(\mathbf{r}) \delta_{ij}$  are the deviatoric strains, and the deviatoric stress in Fourier space is  $\tilde{s}_{ij}^0(\mathbf{k}) = \int_V C_{ijkl} e_{kl}^0(\mathbf{r}) \exp(-i\mathbf{k} \cdot \mathbf{r}) d^3 r$ . In estimating plastic strains, the von Mises yield criterion is used.

The chemical free energy density was defined in Part I [1] as

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