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Multi-field modelling of hydride forming metals. Part I: Model formulation and validation

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ABSTRACT

A computational model for hydrogen transport, hydrogen induced deformation and fracture in metals that form binary hydrides, such as Zr and Ti alloys, is presented. The model uses a continuum description of the two-phase (metal + hydride) material, and solves the multi-field partial differential equations for temperature and stress-directed hydrogen diffusion together with mechanical equilibrium in a three-dimensional finite element setting. Point-kinetics models are used for metal-hydride phase transformation and stress-directed orientation of hydride precipitates, while a cohesive zone fracture model caters for initiation and propagation of cracks. The local fracture properties of the hydrided material are correlated to the calculated local concentration and orientation of the hydride precipitates, which have a strong embrittling effect on the material.

In Part I of this two-part paper, we present sub-models applied for the aforementioned phenomena together with a detailed description of their numerical implementation. The applicability of the model is then demonstrated by simulating five independent experiments on hydrogen transport, metal-hydride phase transformation and stress-directed hydride orientation in zirconium alloys. Based on the results, we conclude that the model captures these phenomena over a wide range of thermo-mechanical loading conditions, including thermal cycling. Part II of the paper is focussed on fracture, and includes details on the fracture model and its validation against tests and experiments on initiation and propagation of hydride induced cracks.

1. Introduction

Hydrogen absorption by some transition metals may lead to precipitation of a binary metal hydride phase, MH_x, wherein x ranges from about 1.5 to 2.0 [1]. Since the metal-to-hydride phase transformation is associated with a volumetric expansion of 15–20%, the hydride formation may cause problems by swelling and geometrical distortion of structural components. However, the far most important effect of hydride precipitates is that they embrittle the material. Since the hydride phase is much more brittle than the metal, hydride precipitation may significantly degrade the material's tensile strength and ductility [2]. The metals of primary practical importance for hydride induced embrittlement and fracture are zirconium and titanium alloys, which are in widespread use in the nuclear energy industry [3] and the aerospace and marine [4] industries, respectively. Zirconium and titanium are group 4 metals, and have similar metal-hydrogen phase

systems [5]. This is illustrated by the binary phase diagrams shown in Fig. 1. In both Zr and Ti, there are two metal phases; a hexagonal closed-packed (hcp) α-phase and a body-centered cubic (bcc) β-phase. Metal hydrides exist in two stable phases: face-centered cubic (fcc) δ-hydride with $x \approx 1.6$ and face-centered tetragonal (fct) ϵ-hydride at higher hydrogen concentrations. There is also a metastable hydride phase, γ-hydride with $x \approx 1.0$ and fct-structure, which can exist at lower temperatures. For both Zr and Ti, there is a low-temperature two-phase region, in which precipitates of δ-hydride exist in a matrix of α-metal over a wide range of hydrogen concentrations. This two-phase region is crucial for hydride induced embrittlement and fracture, and it will therefore be our main focus in this paper.

A comprehensive review of the effects of hydrogen and hydrides on the mechanical properties and fracture behaviour of zirconium and titanium can be found elsewhere [2], and details on the subject will be given in Part II of our work [6]. Here, we will just summarize some characteristics of hydride embrittlement and fracture in the mixed α-metal + δ-hydride region. It is well known that the degree of embrittlement in this region depends on both the volume fraction and the orientation of the platelet-shaped precipitates of δ-hydride. At high volume fractions of hydrides, the

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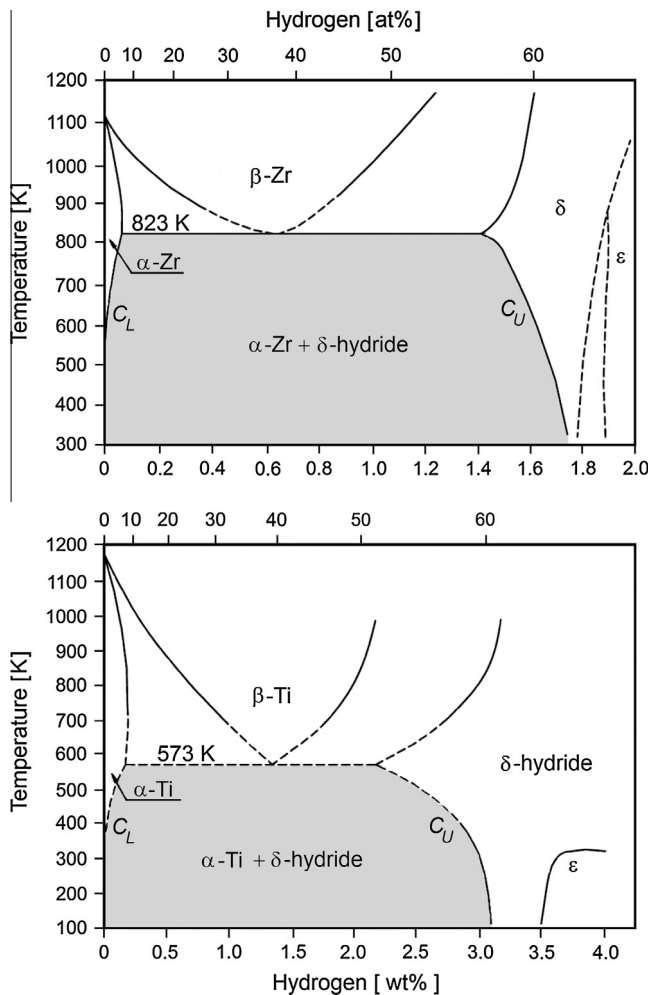
Fig. 1. Phase diagrams showing stable phases of the Zr–H and Ti–H binary systems [5]. The α-metal + δ-hydride region (shaded) is considered in the present work.

Moreover, current models are restricted to simple loading and environment conditions, such as constant and uniaxial loading of a material with constant and uniform temperature. To our knowledge, models for hydride induced embrittlement and fracture under general conditions are as yet unavailable. The reason is that, in a general case, several phenomena have to be considered and modelled simultaneously: (i) Hydrogen diffusion, driven by gradients in stress, temperature and hydrogen concentration. (ii) Metal-hydride phase transformation, i.e. precipitation and dissolution of hydrides. (iii) Stress-directed orientation of hydride precipitates, which induces a very strong directional dependence (anisotropy) for the expansion and strength of hydrided material. (iv) Expansion of the material, caused by hydrogen in solid solution within the metal as well as hydride precipitates, and resulting in internal (misfit) stresses. (v) Cracking and fracture of hydrided material.

Although sub-models exist in the literature for the aforementioned phenomena, there are very few reported attempts to combine these sub-models into comprehensive computational models for hydride induced fracture. This work is an attempt in that direction. Here, we integrate phenomenological sub-models for hydrogen diffusion, metal-hydride phase transformation, stress-directed hydride orientation, hydrogen induced expansion, and fracture of hydride-embrittled materials by coupling the governing equations for the involved phenomena. The resulting system of multi-field partial differential equations (PDEs) is then solved numerically by use of the finite element method. The outcome is a flexible numerical model, which can be used to study a wide range of problems involving temperature- and/or stress-directed transport of hydrogen in combination with hydride precipitation and fracture. The model, which uses a continuum mechanics approach with relevant length scales from microns to centimetres, may also help to bridge the gap between mesoscale models and engineering approaches to fracture in hydride forming metals.

Part I of this two-part paper deals mainly with the formulation and numerical implementation of the model. In addition, results of model validation against experiments on hydrogen transport, metal-hydride phase transformation and stress-directed hydride orientation are presented. Part II of the work [6] is focussed on fracture, and includes details on the fracture model and its validation against tests and experiments on initiation and propagation of hydride induced cracks. Part I of the paper is organized as follows: Section 2 presents a set of sub-models, applied for modelling the fundamental phenomena discussed in the introduction. Section 3 deals with the numerical implementation and integration of the sub-models. The end result is a three-dimensional finite-element based solution to the space–time variations of hydrogen and hydride concentration, hydride orientation, stress and strain. In Section 4, key elements of the model are validated against well-defined separate effect tests and experiments on the behaviour of hydrogen and hydrides in zirconium alloys. Discussions of the model, including comparisons with other modelling approaches, are given in Section 5, and the conclusions are finally summarized in Section 6.

2. Model description

2.1. Representation of hydrided material

In optical microscopy, precipitates of δ-hydride in α-phase Ti and Zr alloys usually appear as thin discs or platelets, and in polycrystal materials, they exist both within the grains and on grain boundaries [1]. These macroscopic hydrides (2–10 μm wide and 0.1–0.2 μm thick) are reported to consist of smaller precipitates (0.1–0.2 μm wide and 30–50 nm thick), which are closely stacked

embrittlement may become severe, since crack growth is possible through a network of interconnected hydrides that provide a brittle crack path through the material. This mode of fracture is very sensitive to the hydride orientation with respect to the direction of tensile stress [7]. At low hydride concentration, a continuous crack path through the brittle hydride phase cannot be formed. Still, the material may lose much of its macroscopic ductility, since the hydrides hinder plastic deformation (dislocation movements) and/or promote initiation and link-up of voids in the material [8].

In addition to these embrittling effects, hydride formation may also challenge the structural integrity through slow, sub-critical fracture processes. This is because hydrogen atoms in solid solution diffuse towards regions with high hydrostatic stress, which over time may lead to excessive local hydride formation ahead of stress-concentrators, such as notches and cracks. Ultimately, the locally embrittled and stressed region may fail, leading to a short (10–20 μm) increment of crack growth. The process then repeats itself, thus giving rise to stable crack growth, the rate of which is basically controlled by the rate of hydrogen diffusion to the peak stress region.

Hydride formation may thus jeopardize the structural integrity of the material in many ways. Accurate computational models are therefore needed, by which hydride induced failures can be predicted, given the loading and environment conditions of a specific component over time. As of today, such models are usually restricted to a specific failure mechanism in a specific material.