Computational Materials Science 122 (2016) 219–228

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Modelling the coupling between hydrogen diffusion and the mechanical behaviour of metals

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ARTICLE INFO

Article history:
Received 8 February 2016
Received in revised form 23 May 2016
Accepted 24 May 2016

Keywords:
Hydrogen embrittlement
Hydrogen diffusion equation
Analogy thermal/diffusion models
Finite element method
HELP mechanism
Abaqus

ABSTRACT

It is well known that hydrogen can have a detrimental effect on the mechanical properties of metals. The aim here is to provide a fully coupled model of the HELP (Hydrogen Enhanced Local Plasticity) mechanism with hydrogen transport. Using the similarities between the heat and mass diffusion equations, a coupled temperature–displacement procedure has been adopted to allow the coupling between hydrogen diffusion and the mechanical behaviour of the material to be simulated. The diffusion equation takes into account the fact that hydrogen atoms reside in interstitial sites and in trapping sites such as dislocations. In the simulations presented here it is assumed that concentration of hydrogen at the dislocations is in equilibrium with the concentration in the matrix interstitial sites. The mechanical behaviour of the material is represented by an isotropic hardening law in which the flow stress decreases with increasing hydrogen content in the matrix which is evaluated by solving the fully coupled mechanical diffusion equations. We use the model to analyse the response of a plane strain component which contains deep and sharp doubled-edged notches. For highly constrained components of this type the hydrostatic component of stress scales with the local yield strength of the material. A high local hydrostatic stress would result in a high hydrogen concentration, but a high hydrogen concentration results in softening, i.e. a low yield strength, and therefore a low hydrostatic stress. These conflicting relationships result in a balance being achieved between hydrostatic stress, hydrogen concentration and yield strength, i.e. the response does not become unstable. Also there is a high degree of kinematic determinacy in the way which the component deforms, i.e. the strain pattern in the presence of hydrogen is very similar to that when there is no hydrogen. A consequence of these two effects is that softening of the constitutive response due to the presence of hydrogen, does not lead to localization of strain and a macroscopic brittle response. Softening must be combined with other degradation process for the material to embrittle.

1. Introduction

Experimental studies and fractography analysis of iron and steels indicate a deleterious influence of hydrogen on their mechanical properties. There is extensive evidence documented in the literature on the effect of hydrogen in degrading the properties of high strength steel; resulting in reduced ductility, strength and toughness, accompanied in most cases by intergranular brittle fracture [1–4]. However, opinion is divided concerning the mechanisms that are responsible for this embrittlement. Numerous theories have been suggested and reviewed. For non hydride forming materials they can be grouped into two main primary mechanisms: hydrogen induced decohesion (HID) and hydrogen enhanced local plasticity (HELP). The HID mechanism finds its roots in the work by Scott and Troiano [1] and Oriani and Josephic [5] which postulates that hydrogen, which accumulates at a crack tip, reduces the cohesive strength of atomic bonds giving rise to a reduced fracture toughness. The HID theory has been generalized by assuming that hydrogen lowers the cohesive strength of a range of internal interfaces such as grain boundaries or carbide–matrix interfaces. The HELP mechanism implies that hydrogen enhances the dislocation mobility by reducing the elastic interaction energy between dislocations. As a consequence, hydrogen reduces the applied stress necessary to move a dislocation through a field of obstacles. This phenomenon promotes material softening. It is worth mentioning that the HID mechanism at the atomic level, i.e. hydrogen decreasing the atomic cohesion, has not been proved experimentally. However, atomistic simulations have revealed that

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http://dx.doi.org/10.1016/j.commatsci.2016.05.030
0927-0256/

the atomic cohesion decreases with increasing hydrogen content. There have been experimental studies of the effect of hydrogen on the crack tip opening angle, which reduces with increasing hydrogen content for a Si-doped iron single crystal. The crack tip becomes sharper in the presence of hydrogen and the fracture surface appears brittle [6]. The HELP mechanism is supported by the experimental studies of Robertson using an in situ charging cell in an environmental TEM [7], in which it is shown that hydrogen enhances the dislocation mobility in stainless steel. However, recent theories recognise that the HID and HELP mechanisms might act in synergy. Novak et al. [8] present work on hydrogen-induced intergranular failure in AISI 4340 steel, in which they assume that failure initiates by decohesion at grain-boundary carbides. Hydrogen promotes the mobility of dislocations that pile up at the matrix carbide interface and enhancing the local stress there. Recently, the authors [9–11] have modelled the behaviour of a carbide-rich region in a dissimilar weld interface in the presence of hydrogen. This carbide rich region provides a low energy path for crack growth. An Abaqus finite element model of the carbide-rich microstructure was created by image processing which converts a TEM image into a finite element mesh. The presence of fine carbides plays an important role in the constitutive response of these materials. Simulations show that in regions where the hydrogen content is high the matrix surrounding the carbides softens and plastic flow is localized. Moreover, the presence of hydrogen lowers the cohesive strength, giving rise to microcrack formation at the carbide–matrix interface, leading to microvoid formation. As deformation proceeds the pores enlarge and link to form cracks, which generates the failure surface. This failure process is supported by experimental observation of failed components.

In order to fully model these and other failure mechanisms that have been proposed in the literature we need to solve the hydrogen diffusion equation to determine how hydrogen is distributed inside the material and couple this with the mechanical constitutive response of the material. The aim of this paper is to explain in details how to solve coupled hydrogen diffusion-mechanics problems in a finite element context by using the commercial finite element software Abaqus. We use the similarity between heat transport and diffusion in order to adopt the coupled tempera ture–displacement procedure available in Abaqus to effectively solve diffusion-mechanics problems. This requires the implementation of a user subroutine (UMATHT) for the diffusion equation. Here we concentrate on coupling the elasto-plastic response with models of diffusional transport and use the resulting routines to analyse a range of problems that have been designed to provide new insights into the HELP mechanism. In a subsequent paper we combine these routines with those for interface decohesion, modelled using cohesive elements, to model situations where diffusion, plastic deformation and decohesion (e.g. at grain boundaries or carbide/matrix interfaces) combine to determine the component response.

The structure of this paper is as follows. Firstly, we give a detailed description of the hydrogen diffusion equation. We then discuss the analogy with the heat transfer problem and the implementation of the equation in a UMATHT subroutine in Abaqus. We include details of the implementation. We solve first a simple diffusion problem for which an analytic solution is available, and then analyse diffusion in an elastic plate with a hole and compare the solution with analytical results. Both these simulations serve to validate the models and the code. We then analyse the response of a double-edged notched plane strain component whose isotropic hardening law is a function of the hydrogen content. We show the effect of plastic strain on the concentration of hydrogen in the traps and in the lattice and evaluate whether for this type of component plastic flow localizes, leading to failure at low macroscopic plastic strain, i.e. whether failure can occur exclusively by a HELP process.

2. Hydrogen diffusion equation

It is well known that hydrogen being the smallest element diffuses more easily through metals than any other element. Even at low concentration, hydrogen often leads to the embrittlement of metals for reasons that are not yet well understood, but are certainly related to the speed at which it can diffuse to highly stressed regions. Hydrogen atoms move through a metal by normal interstitial site (NILS) diffusion or dislocation transport. Hydrogen atoms reside either at NILS or at trapping sites such as dislocations, grain boundaries, carbide/matrix interfaces, microvoids and other defects. The vast majority of sites are the NILS and the minor fraction of sites are the traps. Here we consider a lattice consisting of two kinds of sites for occupancy by hydrogen. Hydrogen trapping at defects has a large effect on the diffusion process. Hydrogen diffuses so easily that even shallow traps can produce a significant effect on the diffusivity. One of the first theories on the mobility of dissolved hydrogen in an iron lattice containing trapping sites was given by McNabb and Foster [12]. They introduced a diffusion equation solved with terms for sources and sinks. Oriani [13] reformulated the work by McNabb and Foster introducing the assumption of local equilibrium between the mobile and the trapped populations for a restricted domain of degree of trap coverage. Sofronis and McMeeking [14] formulated the hydrogen transport problem coupled with large deformation elastic–plastic behaviour of a material based on Oriani's theory. In [14] the authors incorporated the effect of hydrostatic stress and trapping sites at dislocations, which increase in number as the material deforms plastically. Krom et al. [15] demonstrated that the hydrogen transport model proposed in [14] does not provide a correct hydrogen balance. Hence in [15] a modification on the hydrogen diffusion model is introduced which includes a factor depending on the strain rate. The strain rate factor decreases the hydrogen concentration in lattice sites due to the filling of trap sites.

2.1. Implications of Oriani's assumptions

Oriani's theory is based on considering a lattice consisting of two kinds of sites for occupancy by hydrogen. He postulates that the vast majority of sites are the ordinary or "normal interstitial" sites. The minor fraction of sites called "trapping" sites provides an energetically favourable environment for occupancy by the hydrogen atoms. We consider here the case in which the number of trapping sites is small. The trap population then does not reduce significantly the cross-section for diffusion in the normal lattice. We denote the hydrogen concentration in the lattice as C_L (number of H atoms per unit volume) and use C_X to denote the concentration associated with the hydrogen in the traps. In this paper we consider only one kind of trap, i.e. hydrogen trapped at dislocations. The total concentration of hydrogen is given by $C_T = C_L + C_X$, where

$$C_L = \beta\theta_L N_L \tag{1}$$

$\beta$is the number of interstitial sites per solvent atom, $0 \leqslant \theta_L \le 1$, is the fraction of lattice sites occupied by hydrogen atoms and

$$N_L = \frac{\rho N_A}{A_r} = \text{const.} \tag{2}$$

is the number of atoms of solvent per unit volume (atoms/m^3). The density and relative atomic mass of the lattice element are ρ and A_r respectively and N_A is Avogadro's number. Similarly C_X is the hydrogen concentration trapped at dislocations: