# Numerical modeling of hydrogen diffusion in metals accounting for large deformations

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## ABSTRACT

While the deleterious effects of hydrogen on metals and alloys are well known, the precise role of hydrogen in the underlying microscopic mechanisms is still not well understood and as of yet, the modeling attempts on hydrogen embrittlement and hydrogen induced cracking have not led to a proper method for life-time prediction. This work aims at the development of a robust numerical strategy in order to solve the non-linear coupled problem presented in the work of Anand [1]. The numerical implementation is performed for finite element method and the analysis are done to address the issue of hydrogen transport and hydrogen-embrittlement-related failures in metals. Specifically, problems related to the mechanism of hydrogen enhanced localized plasticity (HELP) is studied and macroscale shear localization phenomenon resulting from hydrogen induced material softening is considered at the phenomenological level.

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## Introduction

Hydrogen has a big potential to be an environmentally attractive fuel for the future. However, the usage of hydrogen in a wide range of engineering components has brought a serious challenge for the distribution and storage of its molecular or atomic form. The role of hydrogen in reduction of ductility, fracture strength and crack growth has been reported in the literature for decades [2–5] and the embrittlement of metals and alloys in the presence of hydrogen cannot be neglected. Even tough the precise micro-mechanism responsible for hydrogen embrittlement is still under discussion, various possible mechanisms have been introduced, including hydride formation and cleavage [6–8], hydrogen-enhanced decohesion [9,10] and hydrogen-enhanced localized plasticity (HELP) [11–13]. The first of the three is only operative in a certain group of metals [11] and the latter two are lately considered as co-operative mechanisms [14,15]. The underlying principles in those mechanisms are not universally accepted and often controversial [16,17]. Nevertheless, HELP mechanism can be supported primarily by direct macroscopic experimental observations in a range of hydrogen concentration and temperatures, such as localized shear bands, reduction in yield stress and softening [5,18–21] which is rather easy to quantify and makes HELP mechanism appealing from the phenomenological modeling point of view.

In the literature, there are various continuum-level modeling attempts which associate hydrogen diffusion to elasto-plastic deformation. The work of Sofronis and McMeeking [22] is one of the earliest which proposed a finite element model based on Oriani's assumption of local equilibrium and predicts the effect of hydrostatic stress on hydrogen distribution in plastically deformed areas. Later, their model was extended by Krom et al. [23] where a strain rate parameter is introduced in the hydrogen transport equation. Taha and Sofronis [24] and Liang and Sofronis [4] proposed models accounting for hydrogen induced softening effects and HELP mechanism in order to predict the mechanical behavior at a crack tip. Miresmaeili et al. [25] and Takayama et al. [26] followed similar Galerkin approaches and further investigate shear localization due to HELP and crack tip opening displacement of a crack in a hydrogen environment. Among those attempts, Anand's recent work [1] is the first, where the coupling between hydrogen diffusion and large elasto-plastic deformations are formulated in a consistent thermodynamical framework and large elasticplastic deformations are described in modern $\mathbf{F} = \mathbf{F}^e\mathbf{F}^p$ kinematics.

The main objective of this paper is to numerically implement the theory of Anand and introduce proper plastic flow rates in order to successfully simulate the HELP related shear localization and softening phenomena. For that purpose, primarily Anand's theory is reformulated for isothermal conditions and further developed for hydrogen related softening in a finite strain framework in Section 2. In particular, numerical implementation of hydrogen diffusion is presented in Section 3. Section 4 demonstrates the numerical analysis of a crack in a pipe under high pressure hydrogen and a specimen with an initial hydrogen perturbation under uniaxial tensile load. Finally, Section 5 summarizes the results and concludes the paper with some final remarks.

## Diffusion model coupled with finite strain plasticity

### Balance equations

#### Balance law for hydrogen diffusion
Considering an arbitrary material point denoted by $\mathbf{X}$ in a body with volume $V$ and surface $S$, the total number of moles of hydrogen atoms per unit volume in a fixed reference configuration is represented by $C(\mathbf{X}, t)$. The change in the hydrogen concentration inside an arbitrary spatial region $\Omega$ is characterized by a flux $\mathbf{j}(\mathbf{X}, t)$ denoting the number of moles of diffusing species measured per unit area per unit time. Therefore, mass conservation dictates

$$\frac{\partial}{\partial t}\int_{\Omega} C dV + \int_{\partial\Omega} \mathbf{j}\cdot\mathbf{n}\, dS = 0 \tag{2.1}$$

applying Gauss divergence theorem on the integral over $\partial V$ leads to the local balance law for hydrogen diffusion

$$\dot{C} = -\text{Div}\,\mathbf{j} \tag{2.2}$$

Note that in this work, the hydrogen flux $\mathbf{j}$ is assumed to obey Fick's law of diffusion

$$\mathbf{j} = -m\nabla\mu \tag{2.3}$$

where $m$ is the mobility and $\mu$ is the chemical potential. Hence, the local balance equation for the concentration of hydrogen atoms finally reads

$$\dot{C} = \text{Div}(m\nabla\mu) \tag{2.4}$$

#### Balance of forces and moments
In view of Cauchy's theorem and neglecting the inertial effects, the force and moment balances for an arbitrary spatial region become

$$\int_{\partial\Omega} \mathbf{T}\mathbf{n}\, da + \int_{\Omega} \mathbf{b}\, dV = 0 \tag{2.5}$$

where $\mathbf{T}$ is the Cauchy stress tensor and $\mathbf{b}$ is the generalized body force and $\mathbf{n}$ is the outward unit normal on the boundary. Note that surface traction on an element of the surface $\partial\Omega$ is given by $\mathbf{t}(n) = \mathbf{T}\mathbf{n}$. Equation (2.5) must hold for all spatial regions and from which local balance is derived by using Gauss divergence theorem as

$$\text{Div}\,\mathbf{T} + \mathbf{b} = \mathbf{0} \tag{2.6}$$

### Constitutive equations

#### Basic kinematics
Considering $\mathbf{x}$ as the spatial point occupied by $\mathbf{X}$ at time $t$ within a homogeneous body, the motion of the body can be represented by the following smooth function.

$$\mathbf{x} = \chi(\mathbf{X}, t) \tag{2.7}$$

Accordingly the deformation gradient, velocity and velocity gradient become

$$\mathbf{F} = \nabla\chi, \quad \mathbf{u} = \dot{\chi}, \quad \mathbf{L} = \text{grad}\,\mathbf{u} = \dot{\mathbf{F}}\mathbf{F}^{-1} \tag{2.8}$$

Considering the Kröner decomposition, the deformation gradient is decomposed into elastic and plastic parts in the form

$$\mathbf{F} = \mathbf{F}^e\mathbf{F}^p \tag{2.9}$$

where $\mathbf{F}^e$ is the elastic distortion representing the stretching and rotation and $\mathbf{F}^p$ is the plastic distortion which represents the local deformations due to plastic mechanisms. Note that $\mathbf{F}^p$ is invariant with respect to rigid body motions that are carried by $\mathbf{F}^e$ and they are both invertible. From 2.8 to 2.9

$$\mathbf{L} = \mathbf{L}^e + \mathbf{F}^e\mathbf{L}^p\mathbf{F}^{e-1} \tag{2.10}$$

with

$$\mathbf{L}^e = \dot{\mathbf{F}}^e\mathbf{F}^{e-1}, \quad \mathbf{L}^p = \dot{\mathbf{F}}^p\mathbf{F}^{p-1} \tag{2.11}$$

The volume ratio is expressed as

$$J = \det\mathbf{F} > 0 \tag{2.12}$$

Assuming that plasticity is incompressible

$$J = J^eJ^p, \quad J^p = \det\mathbf{F}^p = 1, \quad J = J^e = \det\mathbf{F}^e > 0 \quad \text{and} \quad \text{tr}\,\mathbf{L}^p = 0 \tag{2.13}$$

Throughout this work, it is assumed that plastic flow is irrotational; therefore,