Stress effects on hydrogen permeation through tubular multilayer membranes: Modeling and simulation

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ABSTRACT

This paper deals with modeling and simulation of stress effects on hydrogen permeation through tubular multilayer membranes. In this case, it is shown that hydrogen concentration undergoes a stress-dependent jump across an interface between two materials, a result that follows from the continuity of the chemical potential and its dependence of the stress. A finite-element model, in which hydrogen concentration is approximated by using discontinuous functions, and the corresponding interface condition is accounted for in a weak manner, is developed and implemented using COMSOL Multiphysics. Numerical results for single-layer and multi-layer membranes are presented and discussed.

Introduction

Hydrogen permeation in metals is accompanied by lattice distortion [1], which in turn generates stress whenever a non-homogeneous hydrogen distribution is involved. This coupling between stress and diffusion gives rise to a series of interesting phenomena and hence has attracted both fundamental and applied research [2–4]. In the case of metallic membranes, the interaction between stress and diffusion has been studied both theoretically and experimentally [5–12]. In this case, the coupling between stress and permeation gives rise to uphill diffusion and stress-dependent hydrogen solubility, among other effects. It can also contribute to the premature failure of the membrane itself [13].

Metallic membranes have been used with success for hydrogen purification and separation processes. In particular, palladium-based membranes are particularly suited to this purpose due to their high permeability and good surface properties. Despite their advantages, palladium-based membranes are usually too expensive for large-scale applications. This has led to the development of multilayer membranes, where a low-cost material such as tantalum, niobium and vanadium, is coated with palladium [14].

In this paper, we consider the modeling and simulation of stress effects on hydrogen permeation through tubular multilayer membranes. Towards this end, we adopt a

continuum theory wherein the interaction between mechanics and hydrogen permeation is realized by considering the mechanical effect of an hydrogen atom within the host solid to be described by a strain linear with the change in hydrogen content, which in turn leads to a stress-dependent hydrogen chemical potential [15]. Further, we consider the permeation processes taking place at the boundaries and interfaces to be fast enough to be considered in equilibrium. Thus, the boundaries conditions employed here involve the prescription of the chemical potential, whereas the interface conditions include the continuity of the chemical potential. Altogether, hydrogen permeation is affected by stress through stress-assisted bulk diffusion and stress-dependent boundary and interfacial conditions. The resulting governing equation for the hydrogen content within the host solid is given by the diffusion equation which includes the stress effect which is supplemented by stress-dependent boundary conditions. Further, the hydrogen concentration is discontinuous across the metal/metal interfaces, with the relationship between the concentration on either side of an metal/metal interface involving the stress-dependent hydrogen solubility for each of the metal.

When applied to single-layer membranes, the model adopted here is essentially the same considered in previous works [5,8,10−12]. The relationship between the concentration on either side of an metal/metal interface can be seen as a generalization of the classical interface condition [16]. In event, we believe that our main contribution relies on the finite element formulation of the problem, specifically on the treatment of discontinuity conditions for the hydrogen content across metal/metal interfaces.

This paper is organized as follows. In Section 2 we present continuum equations, including interface and boundary conditions, that govern solute diffusion and elastic deformation within a small deformation setting. These equations are specialized for the case of a hollow cylinder in Section 3, which also contains the essential ingredients of the numerical model. Numerical results are presented in Section 4.

Preliminaries

We consider a material body $\mathscr{B}$ composed of an elastic solid and a one-component interstitial solute (solute for short). This body, from now on identified with the region occupied by its solid component in a fixed reference configuration, is the stage of two interdependent phenomena, namely solid deformation and solute diffusion. The state of $\mathscr{B}$ at time t is defined in terms of solid displacement $\mathbf{u}(\cdot, t)$ and solute concentration $c(\cdot, t)$, which respectively assign to each material point $\mathbf{x}$ in $\mathscr{B}$ the corresponding displacement and solute content per unit reference volume at time t. Within a small deformation setting, a relevant quantity is the small strain tensor $\mathbf{E}$, which is given by the symmetric part of displacement gradient $\nabla \mathbf{u}$, that is,

$$\mathbf{E} = \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^{\top}). \tag{1}$$

henceforth $\mathbf{A}^{\top}$ denotes the transpose of the tensor $\mathbf{A}$.

Bulk equations

Following Refs. [15,17], we now present the bulk equations that govern solute diffusion and solid deformation within a small deformation setting. This set of equations is comprised by the additive decomposition of the small strain tensor $\mathbf{E}$ into two contributions, one due solute change with no change in stress, $\mathbf{E}_c$, and one due to stress with no solute change, $\mathbf{E}_e$, and two field equations describing the mechanical force and solute content balances. These equations are conveniently organized as follows:

- Additive decomposition:

$$\mathbf{E} = \mathbf{E}_e + \mathbf{E}_c, \quad \mathbf{E}_c = \widehat{\mathbf{E}}_c(c). \tag{2}$$

- Mechanical equilibrium:

$$\mathrm{div}\, \mathbf{T} = 0, \quad \mathbf{T} = \frac{\partial \widehat{\psi}(\mathbf{E}_e, c)}{\partial \mathbf{E}_e}. \tag{3}$$

- Solute mass conservation:

$$\frac{\partial c}{\partial t} = -\mathrm{div}\, \mathbf{h}, \quad \mathbf{h} = -\widehat{\mathbf{M}}(\mathbf{E}_e, c)\nabla\mu, \quad \mu = \frac{\partial \widehat{\psi}(\mathbf{E}_e, c)}{\partial c} - \mathbf{T} \cdot \frac{\mathrm{d}\widehat{\mathbf{E}}_c(c)}{\mathrm{d}c}. \tag{4}$$

here and henceforth: div denotes the divergence operator; $\widehat{\mathbf{E}}_c$, $\widehat{\psi}$ and $\widehat{\mathbf{M}}$ are the constitutive response functions determining the solute-induced strain, the free-energy density and the solute mobility; $\mathbf{T}$ is the stress tensor; $\mathbf{h}$ is the solute flux relative to the solid; and μ is the chemical potential of the solvent in the solid. Notice that $(4)_2$ states that the solute flux is proportional to the gradient of the chemical potential [17], with the mobility tensor given as a function of the elastic strain and hydrogen concentration. The expression for the chemical potential in equation $(4)_3$ can be found in Ref. [17].

Throughout this paper, we shall concentrate on a simple theory for the coupling between mechanics and diffusion appropriate for isotropic solids [15,17]. This theory is based on the following assumptions:

- The solute-induced strain is purely dilatational and given by

$$\widehat{\mathbf{E}}_c(c) = \eta(c - c_0)\mathbf{I} \tag{5}$$

where $\eta > 0$ is constant and c_0 is the solute concentration in the reference configuration used to measure strain. Notice that $\eta = v/3$, where v is the volume occupied by one solute atom in the solid. When equations (5) and $(4)_3$ are combined, we obtain the contribution of only diagonal terms of the stress tensor $\mathbf{T}$ to chemical potential.

- The free-energy response $\widehat{\psi}$ is given by