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Effect of mechanical loading on the galvanic corrosion behavior of a magnesium-steel structural joint

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ABSTRACT

Here a time dependent numerical model aimed to investigate the role of mechanical deformation on the corrosion behavior of galvanic joint is developed. The influence of mechanical loading on the corrosion behavior of the AE44 (Magnesium alloy) and mild steel galvanic joint immersed in a 1.6 wt% NaCl solution is explored across a wide range of combined mechanical and electrochemical conditions. It is shown that the onset of plastic deformation during mechanical loading greatly accelerates the galvanic corrosion behavior. The overall numerical approach developed here provides a robust framework for understanding the role of mechanical deformation on the corrosion behavior.

1. Introduction

Magnesium alloys are of increasing interest due to their low-density, moderate specific strength and stiffness, recyclability, and high damping among other properties [1–3]. In particular, these properties are of direct relevance to automobile, rail, and aerospace applications, and thus, have attracted the interest of the civilian and defense transportation sectors for fuel use reduction. However, the wide-scale applicability of magnesium alloys has been limited due to its poor corrosion resistance. The inability of magnesium alloys to form a stable passive layer in combination with high electrochemical activity, makes them highly susceptible to corrosion damage [4]. At present, this matter is further complicated through unavoidable engineering design, where Mg alloys are frequently coupled to electrochemically dissimilar structural components. In service, these couples lead to preferential galvanic corrosion which is accelerated by mechanical loading in harsh corrosive environments, (such as: sulfides, hydroxides, bicarbonates, CO/CO₂, chlorides and ammoniacal solutions), thereby undermining the structural integrity of transport platforms by drastically decreasing failure life [5–8]. In general, there is a lack of fundamental understanding on the role of mechanical deformation plays in the corrosion behavior of dissimilar metal joints containing Mg based alloys. Therefore, understanding the role of mechanical loading on the galvanic corrosion behavior is highly relevant to the long-term goal of increasing the applicability of magnesium alloys. Moreover, there are no prior comprehensive studies that examine the combined effects of

mechanical loading and corrosion behavior on the structural integrity of galvanic joints.

The recent advances in numerical methods such as finite element methods (FEM) [9–14], boundary element methods (BEM) [15,16], cellular automata [17–19] and peridynamics [20] are increasingly utilized in investigating general galvanic corrosion. For instance, the corrosion behavior between AZ91 coupled to a steel fastener creating a galvanic joint was investigated using BEM [15,16] which found that the predicted current density distribution was in close agreement with experimental measurements. Deshpande [9,10] utilized the arbitrary Lagrangian-Eulerian (ALE) framework to track the evolution of the corrosion front, i.e. electrolyte/electrode surface as the mass redistribution took place, for a Mg alloy and mild steel couple. This resulted in a close agreement between the predicted pit depth measurements and experimental findings. Furthermore, Cross et al. [11] observed a qualitative agreement between experimental and numerical findings on the effectiveness of various coatings for steel. These examples clearly highlight the robust nature of the numerical methods, such as finite elements that allow accurate modeling of the corrosion behavior. Therefore, employing numerical methods to examine the corrosion behavior for a galvanic joint subjected to mechanical loads is highly relevant in improving the life of structural joints that are subjected to harsh conditions. Thus, in present study, the role mechanical deformation on the corrosion behavior between AE44 (Mg alloy) and mild steel galvanic joint immersed in 1.6 wt% NaCl solution was investigated using a novel modeling framework. The goal of the finite element based model, which

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are described in greater detail in the subsequent section, is to develop a comprehensive understanding of complex interactions between the mechanical deformation and electrochemistry by examine a wide spectrum of combined mechanical and electrochemical conditions (magnitude of tensile stress, electrolyte depth and area ratio). Then, the effect of tensile load on the corrosion behavior of the galvanic joint by examining the initial current density and the corrosion pit depth profile will be discussed. Rather than modeling individual corrosion processes, instead the overall electrode potential for the anodic surface as whole is shifted based on the localization of elasto-plastic deformation. Through this model, which captures most relevant phenomena, we show that the increase in pit depth due to the presence of tensile loads far exceeded the theoretical estimates (Faraday's equation) based on the initial current density for the galvanic joint. This typifies the transformation of the corrosion behavior from a predictable (steady-state) to unpredictable (rapid) nature due to the introduction of mechanical stresses. Furthermore, the effect of electrolyte depth and the surface area ratio of the electrodes were systematically reexamined in the presence of mechanical loading. Overall, the model offers technique to assess the failure life of the galvanic joint under combined mechano-electrochemical effects by quantifying the stress intensity factor ahead of the corrosion pit.

2. Model development

The galvanic corrosion simulations were performed using the finite element method within the commercial code COMSOL [21]. First, the coupling of the electrochemical-mass transport problem with the corrosion front movement (i.e., galvanic corrosion without mechanical load) was modeled using the following governing equations that are deduced from the mass balance principle. The mass transfer of ionic species in an aqueous environment is the summation of various contributions such as diffusion, electro-migration and convection that can be expressed in the following manner (Nernst-Planck equation):

$$\frac{\partial c_i}{\partial t} = -D_i \nabla^2 c_i - z_i F u_i \nabla \cdot (c_i \nabla \varphi) + \nabla \cdot (c_i V) \quad (1)$$

where F , φ and V are the Faraday's constant, electric potential (measured with respect to standard calomel electrode, SCE) and electrolyte velocity, respectively. Further, c_i , z_i , u_i and D_i are the concentration, charge number, mobility and diffusion coefficient for the i^{th} ionic species taking part in the electrochemical reaction, respectively. The generalized Nernst-Planck equation (Eq. (1)) can be simplified further for the case of steady state conditions ($\partial c_i / \partial t = 0$) based on the following assumptions:

- The electrolyte solution is well-mixed resulting in a lack of concentration gradient of the ionic species;
- The electrolyte solution is incompressible; and
- The electro-neutrality condition is satisfied in the electrolyte solution ($\sum z_i c_i = 0$). This reduces the mathematical description for the galvanic corrosion (Eq. (1)) to a Laplace equation that depends on the local electrolyte potential field (φ)

$$\nabla^2 \varphi = 0 \quad (2)$$

This equation can be solved numerically with the help of appropriate boundary conditions for the electrochemical domain (refer Fig. 1). For instance, at the electrode-electrolyte interface the gradient of the local electrolyte potential field along the electrode surface normal ($\nabla_n \varphi$) is directly proportional to the current density(j).

$$j = -\sigma \nabla_n \varphi \quad (3)$$

where σ is the conductivity of the electrolyte (for 1.6 wt% NaCl solution $\sigma = 2.5 \text{ S/m}$). The electrolyte domain was assumed to be electrically isolated except for the solution/electrode interfaces that were set as a free boundary, and the bottom of the electrodes were assumed to be

ground (Fig. 1). For the mild steel and AE44 galvanic couple, it was assumed that anodic dissolution of Mg would take place on the AE44 surface (Eq. (4a)), while predominantly hydrogen evolution from the reduction of water (cathode reaction) would occur on the mild steel surface (Eq. (4b)) [22,23].



Next, the cathodic current density evolution for the mild steel surface was inferred from the experimental polarization behavior observed in [9]. The anodic current density evolution for the AE44 surface was described using the Tafel equations [24] for charge transfer polarization.

$$\eta = A \log \left(\frac{j}{j_0} \right) \quad (5)$$

where j_0 is the corrosion current density (refer Table 1), η is the over-potential and A is the Tafel slope for the electrode (refer Table 1). The corrosion current density and anodic Tafel slope for AE44 was obtained by using Levenberg Marquardt least square curve fitting within the Matlab framework on the experimental polarization behavior presented in [9] (also refer Table 1). The evolution in current density at the anode was related to the corrosion rate (C_R) with the help of Faraday's law given by the following expression.

$$C_R = \frac{M}{zF\rho} j_{\text{galvanic}} \quad (6)$$

where F is the Faraday's constant, M is the molar mass of the corroding material (24.305 g/mol for Mg), z is the number of electrons that are lost by the corroding surface (in the case of Mg it is 2), ρ is the density of the corroding material (1820 kg/m³ for Mg) and j_{galvanic} is the galvanic current density in A m⁻².

Next, the effect of mechanical stresses on the electrochemical process was modeled using a shift in the anodic corrosion potential due to elastic and plastic deformation [25]. Note that in this analysis, it is assumed that mechanical deformation results in a decrease in electrode potential for the anode surface. In the presence of elastic deformation, the shift in the electrode potential for an anode ($\Delta\varphi_{a,\text{eq}}^e$) can be given by

$$\Delta\varphi_{a,\text{eq}}^e = -\frac{\Delta P V_m}{zF} \quad (7)$$

where V_m is the molar volume of the electrode ($7.13 \times 10^{-6} \text{ m}^3/\text{mol}$ for Mg) and ΔP is the magnitude of hydrostatic stress. Under the plastic deformation, however, the change in electrode potential ($\Delta\varphi_{a,\text{eq}}^p$) is governed by the effective plastic strain, ε_p [25] which is given by

$$\Delta\varphi_{a,\text{eq}}^p = -\frac{RT}{zF} \ln \left(\frac{\nu \bar{\alpha}}{N_0} \varepsilon_p + 1 \right) \quad (8)$$

where ν is the orientation-dependent factor (0.45 from [25]), $\bar{\alpha}$ is a material constant that relates the evolving plastic strain to the dislocation activity (in the case of Mg, $\bar{\alpha} = 1.67 \times 10^{11} \text{ cm}^{-2}$) and N_0 is the initial dislocation density ($4 \times 10^9 \text{ cm}^{-2}$ [26–28]). Physically, the initial dislocation density represents the effect of material processing; however, this does not incorporate the effect of grain size and second phase particle distribution on the material response. Note that the effective plastic strain evolution and hydrostatic stress fields were computed by performing solid mechanics simulations (see Fig. 1) using an isotropic elastic-plastic material model [29]. Specifically, the von Mises yield criterion along with an isotropic hardening model was utilized. Thus, the overall electrode potential for an anode subjected to elasto-plastic deformation can be expressed by

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