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An arbitrary Lagrangian–Eulerian model for modelling the time-dependent evolution of crevice corrosion

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ABSTRACT

An arbitrary Lagrangian–Eulerian (ALE) model is proposed to model the time-dependent evolution of a crevice corrosion that can achieve the simulation of the transient potential distribution, the dynamic concentration profiles and the time-dependent geometry deformation for a corroding crevice. The time-dependent deformation of the crevice geometry, which is caused by metal dissolution and corrosion product precipitation, is implemented via the ALE method. The model reasonably predicted the dynamic pH distribution inside the crevice. It is also predicted that the deposition of corrosion product accelerates crevice corrosion. The current model would be helpful in elucidating the mechanism of crevice corrosion and other forms of related localised corrosion.

1. Introduction

Crevice corrosion is an especially dangerous type of localised corrosion for its unfeasible detection, which can cause sudden and often catastrophic failure [1–5]. It may occur on the surface of all kinds of metallic alloys as a result of small occluded cells under crevices [2,6]. During the crevice corrosion process, as the crevice mouth narrows gradually, the mass transfer between crevice solution and bulk solution becomes more difficult, resulting in the formation of more aggressive crevice solution [7–9]. A good understanding of the crevice corrosion mechanism cannot only improve our ability to predict and prevent crevice corrosion, but also provide further insights into other forms of localised corrosions [10], such as pitting corrosion [11,12] and stress corrosion cracking [13,14].

Crevice corrosion is an extremely complex phenomenon [15–18]. As far as crevice corrosion of iron in chloride solution is concern, the iron inside the crevice dissolves releasing Fe^{2+} ions. Then Fe^{2+} ions react with water to generate H^+. With the development of crevice corrosion, cations concentrate inside the crevice. In order to maintain charge balance of the crevice solution, the anions outside the crevice (such as Cl^-, OH^-) immigrate into the crevice and concentrate inside the crevice. Subsequently, the crevice solution becomes saturated with ferrous chloride [19]. Consequently, after a series of complex physical and chemical processes, corrosion product precipitates at the crevice mouth. The deposition of corrosion product changes the geometry and the crevice corrosion behaviour. It is known that an active local corrosion cell can be sealed off by a special restrictive geometry via limiting the mass transfer between crevice solution and bulk electrolyte. Thus, the common features of restrictive geometry and local acidity lead to the viewpoint that crevice corrosion is a general phenomenon of “occluded cell corrosion” [20].

Up to now, crevice corrosion has been studied experimentally using methods such as freezing methods using a micro-electrode [21], simulation of corrosion in the occluded space [22], cyclic potentiodynamic polarisation [23], the Potentiodynamic–Galvanostatic–Potentiodynamic technique [24] and dynamic electrochemical impedance spectroscopy [25]. However, the prediction of the crevice damage via those methods still remains difficulties. With the development of computational mathematics and computer technology, mathematical modelling, which is important for predicting corrosion rate, has also been extensively used to analyse the crevice corrosion. Sharland and Tasker [26] established a model to predict the steady-state solution chemistry and electrode kinetics within an active corrosion cavity. The model includes a solid hydroxide precipitation reaction. They found the corrosion rate depends on many parameters, such as external electrode potential and crevice dimensions. Sharland et al. [27] developed a mechanistic model of the propagation stage of an established crevice. The model considers the time evolution of the solution chemistry and electrochemistry within a corroding cavity. Vuillemin et al. [28] published a work to simulate steady state crevice corrosion propagation by considering the formation of solids and gases. Kennell et al. [4,10] developed a critical crevice solution and *IR* drop crevice corrosion model. They reported that cathodic reactions are likely to occur towards the tip of the crevice. In order to quantitatively assess the risk of the corrosion of pipelines under the disbonded coatings, Song [29–33] developed several crevice corrosion models

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Nomenclature

a	anode
c	cathode
b_i	Tafel slope of species i involved electrochemical reaction (V/decade)
c_i	concentration of species i (mol/m^3)
$c_{i,0}$	concentration of species i in bulk electrolyte (mol/m^3)
D	distance from crevice mouth (cm)
D_i	diffusion coefficient of species i (m^2/s)
$\mathbf{E}$	electric field strength (V/m)
E_i	electrode potential (V vs. SCE)
E_i^0	equilibrium electrode potential (V vs. SCE)
F	Faraday's constant (96,485 C/mol)
i_j	current density of electrochemical reaction j (A/m^2)
i_M	anode current density of metal M (A/m^2)
i_i^0	exchange current density (A/m^2)
i_p	current density at artificial boundary (A/m^2)
$\mathbf{J}$	current density vector (A/m^2)
J_i	mass diffusion flux (mol/(m^2 s))
k	rate coefficient of corrosion product deposition (m^{10}/(mol^3 s))
k'	reaction constant of corrosion product deposition (mol/m^3)
$\vec{k}_i$	forward reaction constant for hydrolysis reaction of species i (the unit is related to hydrolysis reaction)
$\overleftarrow{k}_i$	backward reaction constant for hydrolysis reaction of species i (the unit is related to hydrolysis reaction)
$K_{\mathrm{eq},i}$	equilibrium constant for the hydrolysis reaction of species i
M_{CP}	molecule weight of corrosion product (kg/mol)
M_i	molar mass of species i (kg/mol)
$\mathbf{n}$	normal vector
n_j	number of electron transferred of reaction j
N	grid number near crevice mouth
$\mathbf{N}_i$	molar flux of species i (mol m^2/s)
R	universal gas constant (8.3145 J/(Kmol))
R_i	reaction rate of species i (mol/(m^3 s))
R_i^e	Electrochemical reaction rate of species i (mol/(m^2 s))
s	area of corrosion product deposition (m^2)
T	temperature (K)
u_i	mobility of species i (mol m^2/(J s))
$\mathbf{v}$	moving velocity vector (m/s)
V	moving velocity caused by corrosion product deposition rate (m/s)
x	x co-ordinate in spatial frame (m)
X	X co-ordinate in reference frame (m)
y	y co-ordinate in spatial frame (m)
Y	Y co-ordinate in reference frame (m)
z_i	charge number of species i
Greek	
ε	relative permittivity
$\bar{\varepsilon}$	coverage rate of corrosion product deposition at the crevice mouth
$\bar{\varepsilon}_0$	initial coverage rate of corrosion product deposition at the crevice mouth
θ	porosity of corrosion product deposition
θ_0	initial porosity of corrosion product deposition
ϕ	potential (V vs. SCE)
v_{ij}	stoichiometric coefficient of species i in reaction j
ρ	charge density (C/m^3)
ρ_{CP}	density of corrosion product (kg/m^3)
ρ_i	density of species i (kg/m^3)
σ	electric conductivity (S/m)
Γ	boundary

of metals with different influenced factors, such as the oxygen diffusion, the solution resistance and the variation of the disbondment gap.

However, since the appearance of corrosion product inside or outside a crevice will automatically have an impact on the crevice geometry, steady state simulations together with transient simulations without considering time-dependent geometry deformation of crevice cannot precisely describe the whole crevice corrosion process. Vuillemin et al. [28] suggested that one of the challenges in the simulation of crevice corrosion propagation is the rigorous treatment of non-aqueous species (gas or solid), since the appearance of a new phase in a crevice will automatically have an impact on its geometry. To overcome this challenge, moving mesh method would be a promising approach. At present, there are several interesting works about modelling corrosion processes on the basis of moving mesh method [34,35]. Xiao and Chaudhuri [36] presented a FEM-based corrosion model. Their work aim to provide a "deterministic" modelling approach that can systematically identify, couple and characterise multiple physical and (electro)chemical phenomena under a modular framework. Deshpande [37] presented a moving mesh model to predict the corrosion rate of galvanic couples. The numerical corrosion rates were in good agreement with those obtained from the experimental techniques. Subsequently, this model was used to capture the evolution of cross-sectional microstructure to investigate its effect on corrosion behaviour of magnesium alloys [38]. Onishi et al. [39] presented a novel numerical method to analyse time-dependent localised corrosion by combining the finite volume method and the voxel method. The method successfully reproduced the experimental results of a capillary electrophoresis problem and several examples of crevice corrosion. Sarkar et al. [40] presented a numerical framework to simulate corrosive dissolution over long periods of time by using an adaptive meshing strategy, with which the evolution of the ionic fields, current density, and movement of the anodic front can be calculated. They demonstrated that such a mathematical/numerical framework can capture the complex dynamics of electrochemical behaviour in the corroding systems. However, most of those models are developed to simulate the movement of anodic front. As far as we know, there have been no transition models that can be used to study the effect of the deposition from the corrosion product on the crevice corrosion behaviour.

During crevice corrosion, the precipitation of corrosion product and the dissolution of crevice walls naturally occur, and that leading to the timely deformation of crevice geometry. Since the crevice geometry is one of the main factors to influence the development of crevice corrosion, analysing the deformation of crevice geometry would help to understand crevice corrosion [28,41]. On the basis of our published work [42,43], a mathematical model is established in this paper to illustrate the time-dependent evolution of geometry corrosion by considering the crevice deformation caused by metal dissolution and corrosion product deposition.

2. Simulation

The numerical results predicted by the current model are validated against the experimental results of Alavi and Cottis [44]. In their experiment, a plate of AISI 304 stainless steel, which was