

Contents lists available at ScienceDirect

Corrosion Science

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



Film rupture model for aqueous stress corrosion cracking under constant and variable stress intensity factor

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ARTICLE INFO

Article history: Received 30 April 2008 Accepted 25 August 2008 Available online 1 November 2008

Keywords:

- A. Stainless steel
- B. Modelling studies
- C. Stress corrosion
- C. Anodic films
- C. Effects of strain

ABSTRACT

A film rupture model for aqueous stress corrosion cracking is developed and used to predict kinetics of crack growth under constant and variable stress intensity factor. The model predicts that creep is necessary for sustained crack growth and creep rate limits crack velocity for constant K and dK/da loading. Contrary to recent thinking, the crack tip strain due to crack advance is viewed as a result, not a cause of crack growth. The crack tip strain gradient elevates and maintains crack tip stress as the crack propagates, which enables creep and sustained crack growth. The model provides a basis for understanding effects of positive and negative K – variation on crack growth.

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1. Introduction

Stress corrosion cracking (SCC) of light water nuclear reactor components has caused significant economic losses due to disruption in power plant availability and the costs associated with SCC mitigation and remediation. Consequently there is incentive for developing quantitative methods for evaluating the reliability and remaining SCC lifetimes of nuclear reactor components. Most recently there is renewed interest [1,2] in the effects of variations in the applied stress intensity factor, K, on crack growth. Engineering applications of crack growth predictive methods almost always involve assessments of potentially large crack extensions where K may undergo variations, either increasing or decreasing, due to crack growth through stress gradients and into regions of residual strain. However, nearly all SCC crack growth rate data obtained for reactor applications have been obtained using crack growth rate specimens subjected to relatively small K - variations in tests run under constant load, constant displacement, constant applied stress intensity factor and for relatively small crack extensions. Therefore, there is little phenomenological information to guide development of models that are capable of predicting response to variations in the applied K during crack growth.

The film rupture (FR)-active path corrosion (APC) model has been developed extensively for nuclear reactor applications [3] and is further developed here for applications involving variable *K*. The FR model [4] and other APC SCC models, such as the yield

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assisted dissolution (YAD) model [5], are derived from an assumption that cracks advance by the electrochemical dissolution of metal along a preferred corrosion path, such as a grain boundary or crystal slip plane [5-7]. Several mechanisms have been proposed whereby crack tip strain can activate and sustain the local corrosion rate by increasing the electrochemical activity of the crack surface. The YAD models consider that the electrochemical activity of the crack tip can be increased by strain-induced emergence of a higher density of active sites [8,9] such as dislocations and slip planes, by reduction of the thermal activation energy for dissolution at an active site [8], and by strain-induced emergence of higher index slip planes having larger dissolution rates [10]. In the YAD models, cracks may advance film free with constant velocity when the rate with which active sites are generated is equal to the rate with which they are electrochemically dissolved. In the FR models [11-15], an electrochemically active crack tip is maintained by rupturing crack tip oxide thereby creating new active surface area without affecting the reactivity of the film free surface. Crack growth is sustained by strain-induced periodic rupture of the otherwise protective crack tip oxide film.

Vermilyea [11–13] proposed a FR model in which transient creep provides the strain increment required to rupture the crack tip oxide. Parkins [16,17] showed that for a metal undergoing dissolution-controlled cracking, a correlation exists between maximum observed crack velocities and bare metal dissolution current densities measured on smooth electrodes under similar exposure conditions. To account for measured crack velocities at lower rates, Parkins appealed to the Vermilyea model wherein crack growth rate is determined by the relative film rupture and repair rates.

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Nomenclature crack length distance from tip of a propagating crack а r crack growth rate à reference crack growth rate characteristic of fully actià∗ ε_{f} oxide fracture strain $d\varepsilon/dt$ total strain rate within a material element vated crack tip ġ steady state crack growth rate $(\partial \varepsilon / \partial t)_r$ strain rate at fixed distance ahead of crack tip \dot{a}_s ε_{ct} $(\partial \varepsilon / \partial r)_t$ strain gradient within material element at an in- ε' t ct as subscript indicates variable evaluated at crack tip stant in time $\dot{\epsilon}'$ M molecular weight $\varepsilon'\dot{a}$ strain rate within material element due to crack adcharge of the metal cation Z density ŝ $(\partial \varepsilon / \partial r)_t$ spatial strain gradient at fixed distance ahead of ρ i_a anodic current density 3 F $\hat{\epsilon}\dot{a}$ strain rate at fixed distance ahead of crack tip due to Faraday's constant $i_a^* A^*$ reference (bare metal) anodic current density change in crack length with change rate à **ė*** active surface area fraction reference crack advance strain rate A_o^* initial value of active surface area fraction reference creep rate $\dot{\varepsilon}_{o}$ A_s^* strain hardening exponent in Ramberg-Osgood equation steady state value of the active surface area fraction n k passivation rate constant; \bar{k}, k^-, k^+ defined by equations m creep exponent in text K applied stress intensity factor k^* reference (bare metal) passivation rate constant K dK/dt Ŕ γ active area fraction generated per unit strain dK/da

Ford [18] and Ford and Andresen [19] (hereafter referred to as FA) adapted these concepts and derived what they describe as a fundamental crack growth rate equation. The FA crack growth rate equation is an explicit function of the CTSR, for which they adopted empirical equations [19]. The FA model has been reported extensively in the SCC literature. There is precedence for its use as it has been applied extensively within the boiling water reactor nuclear power industry as the basis for component design and lifetime predictions [3] and has been proposed for similar pressurized water reactor applications [20,21].

Shoji and coworkers [22,23] adopted the FA crack growth model and developed a CTSR equation that they describe as fundamentally derived [15] and capable of modelling the mechanical effects of cold work and variable *K* (so called *dK/dt* and *dK/da* effects) [24]. Combination of the Shoji equation and the FA crack growth rate equation represents a unique expression of the film rupture model that has been widely reported, used to analyze and project data trends and used as a basis for prediction of SCC crack growth rates in operating reactor environments [25]. However, Andresen and Morra [2] were unable to account for dK/da effects on environmental cracking of several austenitic stainless steel and nickel-base alloys using the FA-Shoji model. Moreover, Hall [26] detailed conceptual and mathematical problems in the development of the Shoji equation and provided an alternative CTSR equation as discussed below. The film rupture model developed here is combined with Hall's CTSR equation to provide an alternative to the FA-Shoji formulation of the FR-APC model.

2. APC model development

2.1. Active path corrosion assumptions and mechanisms

The fundamental mathematical expression of an APC model is Faraday's Law, which relates the mass of a substance released by corrosion to the electron current transferred at an electrode during electrolysis. Using Faraday's Law the instantaneous crack growth rate, $\dot{a}(t,\dot{\varepsilon}_{ct})$, can be expressed simply as a function of the instantaneous anodic current density, $i_a(t,\dot{\varepsilon}_{ct})$

$$\dot{a}(t, \dot{\varepsilon}_{ct}) = \frac{M}{z\rho F} i_a(t, \dot{\varepsilon}_{ct}),\tag{1}$$

where M is molecular weight, z is charge of the metal cation, ρ is density, F is Faraday's constant, t is time and $\dot{\varepsilon}_{ct}$ is the instantaneous strain rate evaluated at the crack tip surface.

2.2. Active surface area concept

The FR and YAD APC models are conceptually similar as both propose that yielding creates electrochemically active surface either by simply exposing fresh metal surface by rupturing protective oxide films or by creating active metal sites such as emergent high index planes and dislocations. In both cases the current density of a partially activated surface, i_a , can be expressed as a product of the current density of the active surface, i_a^* , and the activated area fraction, A^*

$$\dot{i}_a(t,\dot{\varepsilon}_{ct}) = i_a^* A^*(t,\dot{\varepsilon}_{ct}). \tag{2}$$

It is reasonable to assume that i_a has contributions from a number of discrete crack segments along the crack front that are characterized by local states of transient surface activation, A^* (local). However, the experimental work of Kolman and Scully [27] on deformation-induced oxide film rupture shows that discrete film rupture events are numerous and overlap in time for a propagating crack so that individual transient events may not be detectable in measurements of crack tip current. Kolman and Scully show that individual current transients, originating sporadically from many spatially distributed local areas along the crack front, may sum to establish a condition of "quasi-continuous" film rupture. Therefore, we treat A^* as a spatially averaged property characteristic of the crack front and having a magnitude $0 \le A^* \le 1$.

Operationally, we can obtain the magnitude of A^* from measurements on a straining electrode using

$$A^*(t, \dot{\varepsilon}_{ct}) \equiv i_a(t, \dot{\varepsilon}_{ct})/i_a^*, \tag{3}$$

where i_a is the instantaneous current density and i_a^* is a reference current density characteristic of a fully activated bare metal surface. In principle i_a^* is a material variable that is dependent on temperature and environment, is proportional to the exchange current density and is a function of the usual electrochemical variables [9]. Local areas of the surface are considered to be either activated, $i_a(local) = i_a^*$, or not activated, $i_a(local) = 0$. In what follows i_a is viewed as the dependent variable.

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