Journal of Zhejiang University SCIENCE A
ISSN 1009-3095 (Print); ISSN 1862-1775 (Online)
www.zju.edu.cn/jzus; www.springerlink.com
E-mail: jzus@zju.edu.cn

Review:

Advances in research of stress-assisted corrosion fatigue problem*

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Received Nov. 30, 2006; revision accepted Dec. 15, 2006

Abstract: Ceramic materials are notable for their rigidity, insulation and resistance to hostile environment. Nevertheless, if a stressed ceramic component is exposed to chemical attack, it may suffer from a form of delayed fracture known as static fatigue. From the point of view of a designer, it is clearly desirable to determine the behavior of sub-critical crack growth; the crack path and crack growth rate, as a function of material properties and loading conditions are of particular interest. This paper presents a review of advances in stress assisted corrosion problem in history and its corresponding numerical approaches in the last decades, and finally, comes up with consideration and crucial suggestions for future work.

Key words: Stress corrosion, Finite element method (FEM), Fracture, Static fatigue, Meshless local Petrov-Galerkin (MLPG)
doi:10.1631/jzus.2007.A0221 **Document code:** A **CLC number:** R683

INTRODUCTION

People find that when many glasses and ceramic materials are subject to static loading as well as a chemically reactive environment, they often experience delayed failure due most likely to stress-assisted chemical reactions at the cracks: pre-existing surface flaws grow to a critical length, at which point unstable rapid crack propagation ensues, leading to catastrophic failure. For a given material and ambient environment, the lesser the applied stress, the longer the time to fracture (Mould and Southwick, 1959a; 1959b). When the applied stress decreases to a threshold level, known as a fatigue limit or a stress corrosion limit, failure of the solid can be avoided.

During the past century, experiment and mechanism studies for this particular issue were extensively carried out. Delayed fracture of brittle materials was first reported by Grenet (1889) in glass. Later, Orowan (1944) reported such sub-critical crack growth by observing the strength reduction of a glass in air, compared to that measured under vacuum, which he attributed to a reduction of fracture energy. Following this work, different systematic studies were conducted on glass, single-crystal alumina, and mica (Charles and Hillig, 1961; Bailey and Kay, 1967; Wiederhorn, 1967; 1975; Wiederhorn *et al*., 1974; Freiman *et al*., 1985). Those studies showed that a unique relationship could be established, for a given environment, between the crack tip velocity and the stress intensity factor, e.g., results from experiments on some ceramic materials suggest that crack tip velocity can be an exponential linear function of stress intensity factor.

Unfortunately, the mechanisms are still not completely understood so far. However, it is generally believed that, in corrosive environment, the pre-existing flaws induced by abrasion, cutting, and machining, nothing but water plays a key role in effectively increasing the rate of slow crack growth. Dai *et al*.(1995) reported the water corrosion behavior of fluoroaluminate glass. Chevalier *et al*.(1999) conducted crack propagation tests for crack velocities of 10^{-12}~10^{-15} m/s in several environments, including air, water (in the temperature range of 3~85 °C), secondary vacuum (10^{-5} mbar), and even silicon oil, and confirmed that the stress corrosion by water

* Project supported by the Special Scholar Grant of Zhejiang Ocean University (No. 20051217), and Education Department of Zhejiang Province (No. 20061137), China

molecules is the key mechanism for the crack propagation in 3Y-TZP ceramics.

Michalske (1983) proposed a molecular mechanism for stress corrosion, which involves a chemical interaction between strained crack tip bonds in vitreous silica and water molecules from the environment. They showed how such a molecular interpretation of the stress-corrosion process leads to an understanding of the effectiveness of water as a stress-corrosion agent along with the ability to qualitatively predict the effects of various non-aqueous stress corrosion agents. Fig.1 sketches the process of water molecule attacking the Si-O-Si bond at crack tip and forming surface hydroxyl groups. Salganik *et al*.(1997) presented similar molecule level model to investigate the effects of material structure on the crack growth and calculated crack velocity as a functions of stress intensity factor, temperature and humidity around the crack tip.

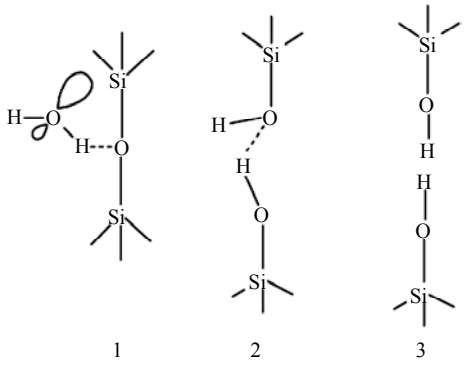

Fig.1 Representation of proposed reaction between water and strained Si-O-Si bond at crack tip. Reaction steps involve (1) adsorption of water to Si-O bond, (2) concerted reaction involving simultaneous proton and electron transfer, and (3) formation of surface hydroxyl groups

However, another understanding of a material's delayed failure is that this behavior is attributable to a process involving chemical dissolution of material from the region near the tips of small pre-existing cracks in the solid (Charles and Hillig, 1961; Hillig and Charles, 1965; Wiederhorn, 1975; Chuang and Fuller, 1992). Consequently, the morphology of the surface flaws is altered and their tips are allowed to move forward. The rate of the kinetic changes on the surface depends on the environmental species and the local stress states which in turn are functions of the applied stress and the continuously changing flaw geometry. The loss of material causes cracks to progressively sharpen and increase in length until catastrophic fracture occurs. By this mechanism, the behavior of crack propagation is determined together by chemical reaction environment in the flaw, stress states, and geometrical condition near the crack tip. The first attempt to deal with static fatigue based on this mechanism was made by Charles and Hillig (1961), who proposed a model based on an absolute reaction-rate theory, which is well-known in physical chemistry. They considered an elliptical-shaped cavity embedded in a homogeneous, isotropic, and linear elastic solid subjected to a remote tensile load applied in a direction perpendicular to its major axis. They also built into the model an implicit assumption that the geometry of the cavity effectively retained an elliptical shape after chemical dissolution. This postulate reduced the complexity of the mathematical analysis and permitted the treatment to be focused on the points at the cavity apex and its immediate vicinity. What actually happened elsewhere along the reactant surface was neglected. As a result, the Charles and Hillig's model predicted the existence of a critical applied stress, defined as a fatigue limit, at which the stress concentration factor at the cavity apex stays invariant at all times and sharpening or blunting of the notch tip takes place when the applied stress lies above or below this fatigue limit. This model was extended by Chuang and Fuller (1992) by calculating the initial rate of material loss on the crack surface. They confirmed the existence of fatigue limit but pointed out that the crack surface was unlikely to remain elliptical through its propagation and consequently predicted four possible behaviors of evolution of flaws.

NUMERICAL APPROACHES

The rates of at least some chemical reactions involving solids are dependent on the mechanical stress levels at or near the reaction boundary. The reactions, in turn, can change the local stress field distribution and introduce both long- and short-term time dependencies in the mechanical strength. For example, ozone-cracking of rubber is stress enhanced. Galvanic stress corrosion of alloys can also be considered to match this description. In most cases, the mechanism is only partially understood.

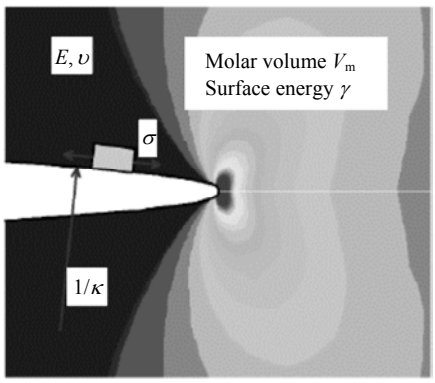

Fig.2 Sketch map for Hillig and Charles's model of stress corrosion problem. E, υ are elastic modulus and Poisson's ratio, respectively; κ is curvature of surface and σ is the tangential stress along the crack surface

Hillig and Charles (1961; 1965) presented a constitutive law to characterize the resulting rate of loss of material on crack surface. They proposed that the mechanism leading to the ultimate failure of a brittle substance maintained at a constant nominal applied stress is due to the enhancement of pre-existing stress concentrators in the form of fissures. Changes in the tip geometry of these fissures can result from a stress-sensitive corrosion reaction. This reaction is considered to be a surface-controlled, thermally activated reaction for which the rate is determined by the local 'thermodynamic driving force' and the local energy barrier. Both the driving force and energy barrier depend on the local stress state and curvature.

We have noticed that Hillig and Charles's model is the only one that has been widely applied in numerical simulation of crack propagation. For better understanding in this model, we would like to describe some fundamental governing equations with plane assumption. For instance, in plane strain problem for convenience, the rate of corrosion can be expressed as a velocity of attack normal to the surface by

$$v_n = 2A\sinh\frac{\Delta F}{2RT}\exp\frac{-\Delta F^*}{RT} \approx A\exp\left[-\frac{\Delta F^* - \Delta F/2}{RT}\right], \tag{1}$$

where v_n is the normal velocity of the surface in the unstressed reference configuration, ΔF and ΔF^* are the free energy of the corrosion reaction and the activation free energy, respectively, A is constant, and R and T have their usual thermodynamic significance. The reaction free energy can be analyzed into the following components:

$$\Delta F = \Delta F_0 - \gamma V_m \kappa + \sigma^2 V_m / 2E', \tag{2}$$

where γ is the surface free energy, κ is the curvature, V_m is the molar volume, and E' is the elastic modulus in plane strain problem. The second term on the right assumes that surface geometry remains constant along the length of the flaw. Now we expand ΔF^* in powers of stress σ to yield

$$\Delta F^* = \Delta F^*(0) + \sigma\frac{\partial \Delta F^*}{\partial \sigma} + \frac{\sigma^2}{2}\frac{\partial^2 \Delta F^*}{\partial \sigma^2}, \tag{3}$$

where $\sigma=\sigma_{ij}t_it_j$ is the tangential stress at the void surface. Then it is convenient to re-write Eq.(1) as:

$$v_n = -v_0 \exp(\Psi / RT), \tag{4}$$

where v_0 is the corrosion rate on an unstressed planar surface (v_n is negative because the surface is receding) and

$$\Psi = V_m(\alpha\sigma + \beta\sigma^2 / E' - \gamma\kappa), \tag{5}$$

where

$$\alpha = -\frac{\partial \Delta F^* / \partial \sigma}{V_m}, \quad \beta = \frac{V_m - E(\partial^2 \Delta F^* / \partial \sigma^2)}{2V_m}. \tag{6}$$

From Eq.(6), one may see α is a dimensionless phenomenological constant such that $\alpha V_m = (\partial \Psi/\partial \sigma)_{\sigma=0}$ is the activation volume for the reaction; β is another dimensionless constant, which accounts for both a quadratic term in stress in the Taylor expansion of the activation energy about σ=0 and also for the strain energy released as atoms are removed from the surface.

In this model, it is assumed that chemical reactions at the solid/vapor interface limit the rate of material removal, so that it is not necessary to account explicitly for processes involving diffusion of material to or away from the reaction site, nor is it necessary to model adsorption or desorption of chemically reactive species at the surface. Eqs.(4) and (5) indicate that the reaction is driven by a difference in chemical potential between the material near the solid

surface and the reaction product, and that the rate of reaction is determined by a combination of the driving force and the activation energy associated with the chemical process. In the absence of stress, this causes a flat surface to recede at uniform rate v_0, which is generally a function of temperature. Surface curvature and stress modify the chemical potential of atoms near the void surface, and also influence the activation energy.

The essential feature of this micro-mechanical model is that the rate of material loss from a surface is influenced by both the stress-acting tangent to the surface and also by surface curvature. Stress generally tends to increase the rate of material loss, while the curvature of a concave surface reduces it. The competition between these two effects may be characterized by a dimensionless parameter

$$\Sigma = \sigma_{\text{tip}} /(\gamma\kappa_{\text{tip}}), \tag{7}$$

where σ_{tip} is some measure of the stress near the cavity tip, γ is the free energy per unit area of the unstressed surface, and κ_{tip} is the surface curvature near the crack tip. For large Σ, the effects of stress dominate over curvature, so that the ellipse tip propagates more rapidly than the flanks. This causes the ellipse to sharpen, and eventually results in the formation of a crack that triggers brittle fracture. For small Σ, the effects of stress are negligible. Material at the tip of the ellipse then dissolves more slowly than material near the flanks, and the ellipse is blunted, eventually evolving to a rounded cavity. The critical value of Σ that discriminates between blunting and sharpening gives the static fatigue limit for the solid. Charles and Hillig (1961) estimated the critical Σ by assuming that the crack remains elliptical throughout its evolution.

Chuang and Fuller (1992) extended the Charles-Hillig (1961) model to compute the initial rate of dissolution of material from the entire surface of the ellipse. Under a uniform applied stress σ^{∞}, the analytical solution of the stress field of an elastic medium containing a 2D elliptical void is available, so is the curvature distribution along the elliptic surface. Then with some typical values of material properties and temperature, the initial corrosion rate of the surface can be obtained by using Eqs.(4) and (5). Chuang and Fuller confirmed the existence of critical load values for sharpening but showed that an initially elliptical flaw was unlikely to remain elliptical throughout its evolution, and instead predicted four possible regimes of behavior for the crack: (1) gross blunting where the rate of dissolution of material from the crack flanks exceeds the rate of removal near the tip so that the ellipse approaches a circular shape; (2) enhanced blunting where the material just adjacent to the crack tip is removed faster than material at the crack tip itself, resulting in blunting near the apex; (3) necking, where the material removal rate is a minimum just adjacent to the crack tip, resulting in a neck-like crack forming near the apex; (4) gross sharpening, where material is removed most rapidly near the crack tip. In addition they showed that, for a typical reaction theory based constitutive law of corrosion, a second material parameter m, where $m=\alpha^2E'/(\beta\kappa_0\gamma)$, plays an important role in governing the behavior of the crack. In general, the expressions for both the driving force for material removal and also the associated activation energy contain linear and quadratic terms in stress. For large m, the linear term dominates, while for small m, the quadratic term is dominant. Chuang and Fuller (1992)'s computations suggest that there exists a threshold value for m, which controls a transition from enhanced blunting behavior (regime *b*) to neck-like crack growth (regime *c*).

Both of those calculations related with Charles and Hillig's constitutive law thus either were based on a simple geometrical description of the crack, or predicted results based on the initial rate of material loss from the crack surface. Then it is very natural that a time dependent numerical simulation is highly expected to determine the behaviors of crack propagations with various combinations of parameters.

Tang *et al*.(2000) developed a numerical method, where a front-tracking finite element method (FEM) was used, to compute the evolution of a crack-like defect that propagates by stress driven corrosion in an isotropic, linear elastic solid. Depending on material properties, loading, and temperature, they observed three possible behaviors for the flaw: (1) gross blunting at the crack tip, (2) stable, quasi-steady state notch-like growth, and (3) unstable sharpening of the crack tip. The range of material parameters and loadings that cause each type of behavior was computed. The results also confirmed the existence of a threshold stress level (known as the fatigue limit) that leads to crack sharpening and ultimately to catastrophic fracture. Contrary to earlier predictions, however, the simulations showed that the fatigue threshold is determined not only by the driving force for crack

extension but also by the kinetics associated with the chemical reaction at the crack tip. The results suggested that the fatigue threshold is likely to decrease as temperature is reduced. Finally, they computed the steady state crack tip velocity as a function of applied load in the regime of steady state crack growth. Their predicted crack growth law is in good qualitative agreement with experiment, but uncertainties in material data made quantitative comparison difficult.

The transition from one behavior to another depends on notch geometry, stress, temperature, and on the material parameters characterizing the chemical reaction and elastic properties of the solid. These calculations therefore provide an estimate for the static fatigue threshold. In addition, one can estimate the time required for the notch to become a crack, and thereby obtain estimates for static fatigue periods above the static fatigue threshold. One difficulty in actually applying this model is that the notch geometry evolves as a function of time and loading, and must be computed as part of the solution. Estimates based on assumed flaw geometries prove to be highly inaccurate, and consequently do not match experimental observations. Until recently, such computations were beyond the scope of numerical methods. In the last couple of years, Tang (2002) and Tang *et al*.(2004) reported the results of numerical simulations of the growth and deflection of a stress-corrosion crack on the interface between two brittle solids.

CONSIDERATIONS FOR FUTURE WORK

Regarding the mechanisms of stress-assisted corrosion problem, some recent models of static fatigue (e.g. Lawn, 1975; Cook and Liniger, 1993; Cook, 1999) have adopted the view that the sub-critical cracks in a brittle solid are atomically sharp. In this case, the crack is assumed to propagate as a result of chemically assisted bond rupture ahead of the crack tip. In these models, the crack can only propagate if the energy release rate G induced by remote loading exceeds the intrinsic material toughness (the toughness is twice the surface energy γ for an ideally brittle solid). The static fatigue limit therefore coincides with what would normally be considered to be the point of fracture in an ideally brittle material. If $G>2\gamma$, the excess energy is assumed to drive a thermally activated mechanism responsible for rupturing bonds at the notch tip. The rate of bond rupture may be limited either by the rate of material transport to the notch tip, or by the kinetics of the reaction itself. In the reaction-controlled limit, the rate of bond rupture is related to the energy release rate through a phenomenological rate law, based on reaction rate theory, in the same spirit as the original Charles-Hillig model. The result is a predicted crack growth rate of the form $v=v_0\sinh(V_m(G-2\gamma)/RT)$, where V_m is an activation volume, R is the gas constant, T denotes temperature, and v_0 is a temperature dependent constant.

The chemically assisted bond rupture models of subcritical notch crack are widely used, and predict trends that are in good agreement with experiment. It is not clear that they contain a fundamentally better description of the atomic scale processes that cause static fatigue than the early Charles-Hillig models, however. The bond rupture model is critically dependent on the existence of an activation energy barrier for bonds to break ahead of a notch (lattice trapping). Early models indicated that such an energy barrier does exist (Thomson *et al*., 1971), but more modern atomistic simulations show that the existence of lattice trapping is strongly sensitive to the choice of atomic potentials (Curtin, 1990; Berensetin and Hess, 2003). Lattice trapping is not always observed in molecular statics computations of perfectly crystalline solids. Whether lattice trapping is significant in amorphous materials remains an open question. These issues do not arise for the Charles-Hillig model, although this viewpoint also includes an activation energy barrier associated with material dissolution, in this case the material at the eroding surface is subjected to much lower stress levels. However, the Charles-Hillig model is not without problems of its own. For example, while appropriate chemical reactions have been identified that may drive the bond-rupture model (Michalske and Freiman, 1982), this is not the case for material dissolution. The original Charles-Hillig model also idealizes the precursors to fracture as smooth notches. Opinion is divided as to whether sub-critical cracks in ceramics and glasses are truly sharp at the atomic scale; experiments have reported that both blunt notches and sharp cracks may be present in actual specimens (Bando *et al*., 1984; Lawn *et al*., 1985; Wiederhorn *et al*., 2003).

Clearly, further work is required to resolve these issues. In all probability, the two viewpoints are not mutually exclusive: brittle fracture in an actual material could be triggered either by pre-existing sharp

cracks that increase in length by subcritical (lattice-trapping limited) growth, or by pre-existing notches that evolve to form sharp cracks. On forming a sharp tip, notches may transition from Charles-Hillig type growth either to sub-critical crack growth limited by lattice trapping under appropriate loading conditions, or directly to brittle fracture.

Concerning the numerical approaches applied in the issue, a question arises very naturally: since the typical crack length shows the scale of several micros, crack tip curvatures is predicted in the order 10^{11} m^{-1}, and the crack tip stress is correspondingly high. Then the validity of a continuum linear elastic solution in this regime is highly questionable. Moreover, the re-meshing techniques with acceptable large time step size in finite element simulation do not prove its capability of estimating the crack tip curvatures accurately and smoothly. The curvatures, especially that of the crack tip, which evolve as a function of time and loading, and must be computed as part of the solution, turned out to be remarkably sensitive to the mesh size along the flaw, so that, inevitably, the results obtained had much fluctuation in both predicted crack tip curvatures and propagation velocities.

In the last decade, the meshless local Petrov-Galerkin (MLPG) method was employed widely and intensively. MLPG was first introduced by Atluri Zhu (1998a; 1998b), wherein the weak forms of governing equations are based on local sub-domains, instead of whole domain in question. Subsequently the test function domains are also intentionally localized to sub-domains and, the space for test function may be completely different from that of trial function. MLPG is well known as a truly meshfree method, in which two characteristics are guarantied: one is a non-element interpolation technique, and the other is a non-element approach for integrating the weak form. Most of the element-free methods are based on the non-element interpolation techniques, such as the Shepard interpolation technique (Shepard, 1968) and other approximations mentioned above, which do not need any elements for constructing the interpolation functions for the unknown variables. However, most of the meshless methods such as EFG, PKPM, and hp-clouds method, still require a global background mesh for numerical integration of the global weak form. Soon after MLPG method was introduced, it was intensively developed and implemented in many areas (Atluri *et al*., 1999a; 1999b; 2000; Atluri and Zhu, 2000a; 2000b). Atluri and Shen (2002a; 2002b) in their pioneering work have depicted the framework and application of MLPG method systematically, and successfully developed some fast and robust approaches. It should be noted that the MLPG concept is independent of a meshless interpolation technique, and can be combined with any meshless interpolation technique, such as PUM, or PKPM. Furthermore, it can be shown that almost every other meshless method proposed in literature could be considered as a special case of MLPG. Meshfree method has been proved to be an effective alternative way in many areas, ranging from solid mechanics to fluid mechanics (Wu *et al*., 2005), from static field to dynamic analysis (Qian *et al*., 2004), from linear assumption to non-linear problem (Tang *et al*., 2003). And it has also been recognized as a better way in describing the crack related numerical simulations.

ACKNOWLEDGEMENTS

The authors thank Prof. Haojiang Ding for his helpful discussion and suggestion.

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