A Transient Model of Hydrogen Induced Delamination

L.N. Germanovich[1], A.V. Balueva[2]
[1]*Georgia Institute of Technology, Atlanta, USA;* [2]*Gainesville State College, Gainesville, USA*
e-mail: abaloueva@gsc.edu

In this work, we consider the diffusion-controlled axisymmetric fracture in an infinite space, and half-space. An important example of diffusion-controlled fracture growth is given by hydrogen induced cracking. In metals, hydrogen is typically dissolved in the proton form. When protons reach the crack surface, they recombine with electrons and form molecular hydrogen in the crack cavity. Then, the fracture can propagate even in the absence of any external loading, that is, only under the excessive pressure of gas hydrogen accumulated inside the crack.

Our results show that in the long-time asymptotic approximation (based on the quasi-static solution), the diffusion-controlled delamination propagates with constant velocity. We determine a maximum critical concentration that limits the use of the quasi-static solution. A transient solution, representing a short-time asymptotic approximation, is used when the concentration of gas exceeds the critical concentration. We then match these two end-member cases by using the method of Pade approximations and present closed-form solutions for both internal and near-surface diffusion-controlled crack propagation at different time scales.

1. INTRODUCTION

Hydrogen is known to reduce the fracture resistance of many metals and steels, and thereby affect the behavior of engineering structures [e.g., *Hirth*, 1984; *Panasyuk et al.*, 1987; *Hick and Alstetter*, 1992; *Zhong et al.*, 1993]. Hydrogen absorbed by a metal is usually dissolved in the lattice in the *proton* form [e.g., *Turnbull*, 1993; *Vehoff*, 1997; *Krom et al.*, 1999]. Some of the protons reach the surface of pre-existing or freshly created cracks where they react with electrons and form *molecular* hydrogen in the crack cavity [e.g., *Zapffe and Moore*, 1943; *Van Leeuwen*, 1974; *Turnbull*, 1993; *Gonzales et al.*, 1997]. Because the effective radius of hydrogen molecules usually exceeds the size of vacancies in the lattice cell, the molecular form of hydrogen is thermodynamically more stable near the crack surfaces, which leads to accumulation of gas hydrogen inside the crack. As a result of excessive hydrogen pressure, fracture often takes place even in the absence of any external loading [e.g., *Eliaz et al.*, 2004; *Turnbull*, 1993; *Vehoff*, 1997] and such a process is usually called hydrogen-induced cracking (HIC).

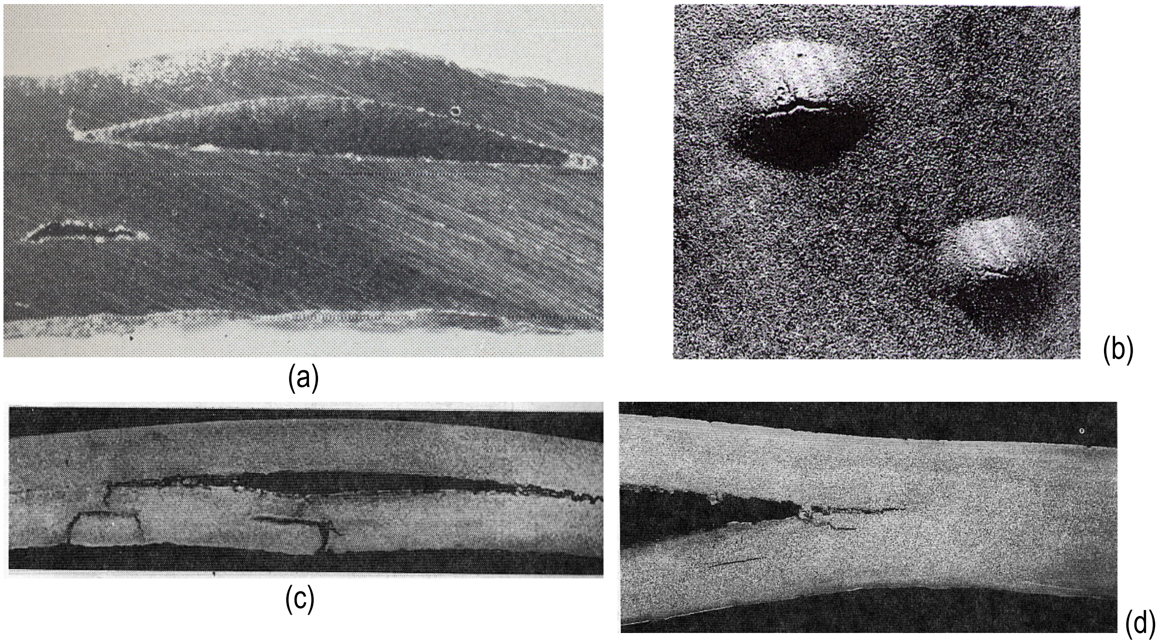


Figure 1. (a) Hydrogen induced fracture near the external surface of a thick steel pipe [after *Polyakov*, 1996], (b) hydrogen generated blisters on the [metal] surface [after *Davis*, 2000], and (b and c) hydrogen caused delaminations in a metal plate [after *Gapharov et al.*, 1998].

In this work, we consider an importan case of HIC – hydrogen-induced delamination (HID) that occures as a separation of a surface layer from the solid (Figure 1) caused by hydrogen embrittlement in metals [e.g., *Speidel*, 1984; *Turnbull*, 1993; *Vehoff*, 1997]. In particular, a common feature of HIC in pipes [*Gonzalez et al.*, 1997], is that the fractures propagate in the direction parallel to the pipe wall (as in Figures 1c and 1d). In time, such delaminations spread, damaging the pipe wall, which often results in the pipeline fracture and its premature replacement [*Gapharov et al.*, 1998]. Understanding the mechanism of HID may improve the design and safety of pipelines.

Various models of growth of *internal* cracks (i.e., far from the surface) that are pressurized by inflow of hydrogen have been considered, for example, by *Goldstein et al.* [1977 and 1985], *Panasyuk et al.* [1987], *Balueva* and Dashevski [1995], *Vehoff* [1997], and *Toribio and Kharin* [1998], *Eliaz et al.*, 2004]. A similar HID model has been considered by *Gonzalez et al.* [1997]. The model assumes that upon crack extension, the volume of the crack cavity increases resulting in the decrease of hydrogen pressure, which causes the crack to arrest. As the cavity continues to be filled with hydrogen, the crack propagation continues as well. *Gonzalez et al.* [1997] also conducted an experimental study with carbon steel in the form of pipe using ultrasonic inspection to measure crack sizes. Using the steady-state approximation for hydrogen diffusion, *Gonzalez et al.* [1997] obtained a closed-form solution for the crack growth rate, which agreed well with their experimental results corresponding to the initial stages of the crack growth. However, their model agrees less satisfactory with the experimental results for large crack sizes, which may not be very surpricieng. Indeed, as pointed out by by *Goldstein et al.* [1977], a theory based on the steady-state approximation ceases to be accurate for large crack sizes because the diffusion process becomes "slower" than the fracture growth. A transient HID model is needed for large crack sizes, when the delamination may become more dangenerous because due to the interaction with the free surface, hydrogen inflow may not not be needed anymore for the fracture growth and the growth may become unstable.

In this paper, a transient model of a penny-shaped delamination controlled by a diffusion process is considered. We obtain the delamination size, velocity of growth, and time of incubation. We also present a quantitative analysis of the results for hydrogen diffusion and metal embrittlement.

2. NEAR – SURFACE DELAMINATION

2.1. Delamination kinetics

Let a half-space $z < 0$ (e.g., a substrate of the base metal), saturated uniformly by fluid with concentration c_0, be covered by a thin infinite layer of thickness h. Suppose a circular delamination of the initial radius, a_0, appears in the interface, $z = 0$, at $t = 0$ (Figure 2). The covering layer is assumed to be thin compared to a_0 ($h << a_0$). As delamination develops, the crack opening, w, under the fluid pressure, p (Figure 2), can be determined in the asymptotic approximation of thin plates [e.g., *Timoshenko and Goodier*, 1970] as $w(r) = pa^4(1 - r^2/a^2)/(64D_0)$, where $D_0 = Eh^3/[12(1 - \nu^2)]$ is the plate flexural rigidity, E and ν are the Young modulus and Poisson ratio of the delaminated material. In the axisymmetric case, the potential energy of bending of a circular plate is $U = -\pi p^2 a^6/(384 D_0)$. In the framework of beam (plate) asymptotic approximation [e.g., *Rice*, 1968], only the bending part of the strain energy contributes to the energy release rate, G. Hence, the rate of energy absorption by the growing delamination per unit length of the crack front can be expressed as $G = -1/(2\pi a)\partial U/\partial a$. Therefore, because for a stably growing fracture, $G = 2\gamma$, the connections between the fracture energy, γ, crack radius, a, crack volume, V, and the fluid pressure, p, can be written as

$$2\gamma = \frac{p^2 a^4}{128 D_0}, \quad V = 2\pi \int_o^a w(r) r\, dr = \frac{\pi}{192} \frac{p a^6}{D_0} \tag{2.1}$$

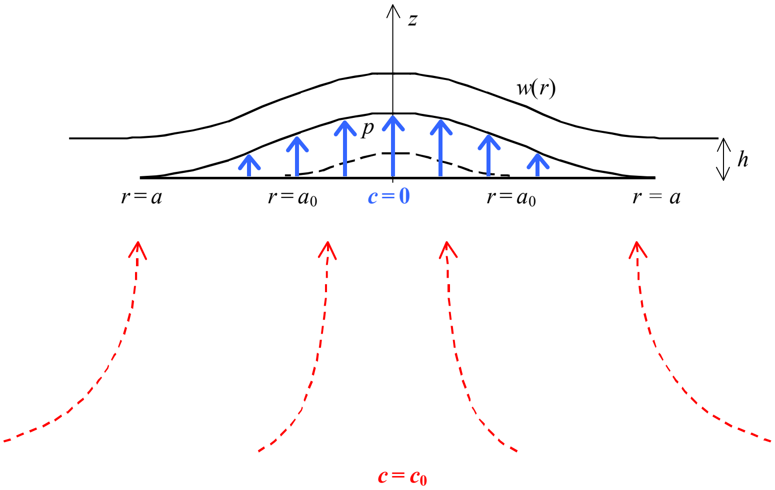


Figure 2. Delamination growth under the pressure of the fluid accumulated in the crack.

The constitutive relation, $p = f(\rho)$, between the fluid pressure, p, inside the crack and fluid density, $\rho = m/V$, can be written in the form of

$$p = f\left(\frac{1}{V}\int_0^t Q(t)dt\right), \qquad Q(t) = 2\pi\int_0^{a(t)} q(r,0,t)\,rdr \tag{2.2}$$

where $Q(t)$ is the full fluid flux into the crack, the first integral in (2.6) represents the total gas mass accumulated inside the crack by time t, $q(r, z, t)$ is the diffusion flux. In many cases [e.g., *Goldstein et al.*, 1985], the hydrogen gas in the crack can be modeled as an ideal gas in isothermal conditions. Then, the constitutive relation $p = f(\rho)$ becomes a linear function $p = RT\rho$, where $R = 8.314$ J/(mole×°K) is the ideal gas constant and T is the gas temperature (in °K). Substituting expressions (2.1) for crack volume and critical energy release rate into (2.2), we arrive at the main kinetic equation

$$a^2(t)\gamma = \frac{3}{4\pi}RT\int_0^t Q(t)dt \tag{2.3}$$

for the transient analysis of the quasi-stationary growth of a penny-shaped delamination. In this consideration, we further need one more relationship between functions $a(t)$ and $Q(t)$ that can be obtain by considering hydrogen (proton) diffusion in metall latice towards the propagating fracture (Figure 2).

2.3. Diffusion flux into delamination

If the flux density, q, is the result of diffusion into delamination, it can be found from the usual boundary value problem for the hydrogen concentration, c, in metal (expressed in moles per unit volume due to the chosen dimenison of R) :

$$\begin{cases} \partial c/\partial t = D[\partial^2 c/\partial z^2 + (1/r)\partial/\partial r(r\partial c/\partial r)] \quad (z < 0,\ t > 0,\ r \geq 0) \\ c = c_0 \quad (z \to -\infty,\ r \geq 0,\ t > 0),\ \ c = c_0 \quad (z < 0,\ r \geq 0,\ t = 0) \\ \partial c/\partial z = 0 \quad (z = 0,\ r > a(t),\ t > 0),\ \ c = 0 \quad (z = 0,\ 0 \leq r \leq a(t),\ t > 0) \end{cases} \tag{2.4}$$

where D is the diffusion coefficient of atomic hydrogen in metal. Then, $q = -D\partial c/\partial z$ ($z = 0$, $0 \leq r < a$) and the full flux into the delamination can be determined from $Q(t)$ in (2.2). If $Q(t)$ is expressed through $a(t)$, after substituting $Q(t)$ into (2.3), the main kinetic equation for $a(t)$ will be derived.

In the case of hydrogen embrittlement, the second condition in (2.4), $c(r, 0, t) = 0$ at $0 \leq r \leq a(t)$, means that if the pressure inside the delamination crack is not too large, the crack can be modeled by an ideal sink so that the *molecular* gas hydrogen accumulates inside the crack [e.g., *Goldstein et al.*, 1977 and 1985; *Gonzales et al.*, 1997; *Eliaz et al.*, 2004], while the concentration of atomic hydrogen there is zero.

In general, the boundary value problem (2.4) can only be solved numerically [*Eliaz et al.*, 2004]. However, there are at least two extreme cases when asymptotic consideration allows one to determine $c(r, \mathrm{z}, t)$ and, then, the parameters of the delamination growth in the closed form. These are the cases of short, $t << a^2/D$, and long times, $t >> a^2/D$, where $a^2/D = t_d$ is the diffusion time scale, which is of the order of the relaxation time required to establish the equilibrium steady state for the hydrogen diffusion around the delamination.

3. ASYMPTOTIC SOLUTION FOR LONG TIMES

3.1. Asymptotic solution for long times

If delamination growth is slow enough so that the delamination time, t, is much smaller than the diffusion (relaxation) time scale, $t_d = a^2/D$, required to establish the steady state in the delamination proximity, at each moment, t, the diffusion flux, q, into the delamination can be found from the solution of the corresponding *steady-state* diffusion problem. We will refer to this as to the asymptotic solution for long times. Then, for each time, t, we should find the concentration function, $c(r, z, t)$, which is harmonic ($\nabla^2 c = 0$) in the half-space, $z < 0$, and satisfies the boundary conditions from (2.4).

The solution of this boundary value problem is well known [e.g., *Sneddon*, 1972]. In aprticular, the expression for the flux density, $q = -D\partial c/\partial z$ ($z = 0$, $0 \leq r < a$), is given by $q(r, 0, t) = (2/\pi)c_0 D[a(t)^2 - r^2]^{--1/2}$, so that the volumetric flow rate in (2.2) into the crack can be written as $Q(t) = 4c_0 Da(t)$. Substituting this $Q(t)$ into (2.3), we finally obtain the kinetic equation for the delamination growth driven by the ideal gas and controlled by diffusion:

$$a^2(t)\gamma = \frac{3}{\pi} RTDc_0 \int_0^t a(t)dt \tag{3.1}$$

At the first stage, that is, during the incubation period, $0 < t < t_i$, the fluid diffuses into the delamination, accumulates inside it, and creates the pressure sufficient for starting the delamination growth. Since at this stage, $a(t) = a_0 = \text{const}$ for $t \le t_i$, we can write from (3.1) the expression for the incubation time, t_i, as follows:

$$t_i = \frac{\pi\gamma a_0}{3RTD\,c_0} \tag{3.2}$$

The velocity, da/dt, of crack growth is obtained by differentiating both parts of equation (3.1) with respect to t: $da/dt = 3RTDc_0/(2\pi\gamma) = a_0/(2t_i)$. This expression reveals that the delamination develops with a *constant* velocity. Such a stationary process of the crack growth is provided by the balanced increase of the gas pressure caused by the fluid diffusion into the crack and by the gas pressure decrease, caused by the increase in the volume of the growing crack. Of course, the velocity is not exactly constant but only with the accuracy of the higher assypmtoti terms that are neglected. In this approximation, time is considered to be a parameter and enters the equation set not through diffusion process, assumed to be fast, but through the kinetic equation (3.1).

3.2. Asymptotic solution for short times

The stationary solution for fluid flux into the crack is valid for long times, $t >> a^2/D$, and provides an upper estimate for the growth time, $t(a)$. Further insight can be gained by turning to a transient solution for the fluid flux. The leading asymptotic term for the fluid diffusion into the delamination for a short time, $t << a^2/D$, is given by the one-dimensional approximation in z-direction (Figure 2), so that the fluid diffusion in the radial direction is only given by the quantities of the higher order with respect to t [*Germanovich and Kill'*, 1985; *Germanovich*, 1986]. Accordingly, the first-order solution of the transient problem (2.4) can be found by considering the latter as independent of r and is well known [e.g., Carslaw and Jeager, 1992]. Then, for $t < t_i$ and $z \le 0$,

$$c(z,t) = \begin{cases} -c_0 \operatorname{erf} \dfrac{z}{2\sqrt{Dt}} & (r < a_0), \quad \operatorname{erf}(x) = \dfrac{2}{\sqrt{\pi}} \displaystyle\int_0^x e^{-\eta^2} d\eta \\ c_0 & (r > a_0) \end{cases} \tag{3.3}$$

Once the delamination front reaches position $a(t)$, the concentration function $c = a(r, t)$ is asymptotically given by the same expression (3.3) where t should be replaced by $t - \tau(r)$, where $\tau(r)$ is the time at which the crack radius, a, reaches the value of r; so that $\tau(a) = a^{-1}(\tau)$ is the function inverse to $a(t)$. Then, the flux into the delamination is given by $q = -D\partial c/\partial z$ ($z = 0,\ 0 \le r < a,\ t > 0$), so that substituting expression (2.2) for $Q(t)$ into (2.3), we obtain the kinetic equation for $a(t)$ in the short time asymptotic approximation:

$$a^2(t) = \frac{3c_0}{2\pi\gamma} RT\sqrt{\pi D}\left[a_0^2\sqrt{t} + 2\int_{a_0}^{a(t)} \sqrt{t-\tau(a)}\; a\,da \right] \tag{3.4}$$

Noticing that $a(t) = a_0$ during the incubation period, $t < t_i$, we find from (3.4) the incubation time, t_i. By this time, a sufficient gas pressure is accumulated in the delaminated space (opening) and the fracture starts growing at

$$t_i = \frac{4\pi\gamma^2}{9c_0^2 R^2 T^2 D} \tag{3.5}$$

Substituting t_i from (3.5) into (3.4) and integrating by parts, (3.4) can be reduced to the well known Abel integral equation of the second kind [*Polyanin and Manzhirov*, 1998], which has the following solution:

$$a^2(t) = a_0^2\left[F(t) + \frac{\pi}{t_i}\int_{t_i}^{t} F(\tau)\exp\frac{\pi(t-\tau)}{4t_i}\,d\tau \right] \tag{3.6}$$

where $F(t) = (1/2)\{[2t^{1/2} - (t - t_i)^{1/2}]/t_i^{1/2} - (t/t_i)\arcsin[(t_i^{1/2}/t^{1/2})]\} + \pi/4$. From (3.6), the delamination growth velocity is $da/dt = (1/2)[a_0^2/a(t)][dF/dt + (\pi/4)(1/t_i)(a^2(t)/a_0^2)]$. In this case, the delamination velocity is not constant.

4. EXAMPLES

Figure 3 shows the dependence of the normalized incubation time, $t_i' = t_i / t_0$, on the dimensionless fluid concentration in the material, $c_0' = c_0 / c_*$, where $c_* = 2\gamma/(a_0 LRT)$. Curves 1 and 2 in Figure 3 correspond to the incubation times in the long and short time approximations, respectively. Line 3 shows the Pade asymptotic approximation.

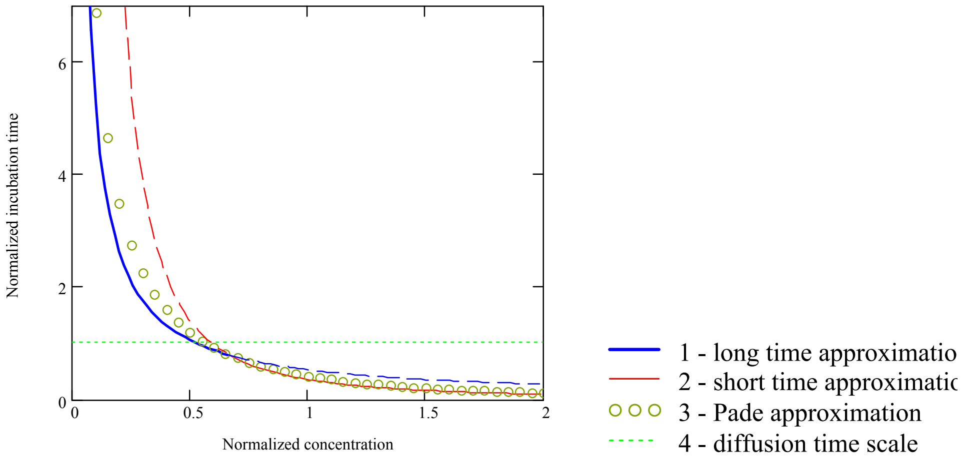
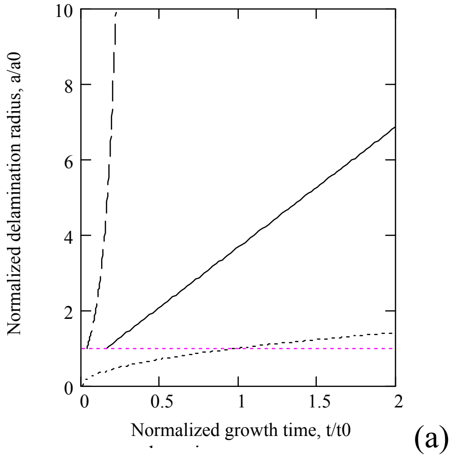


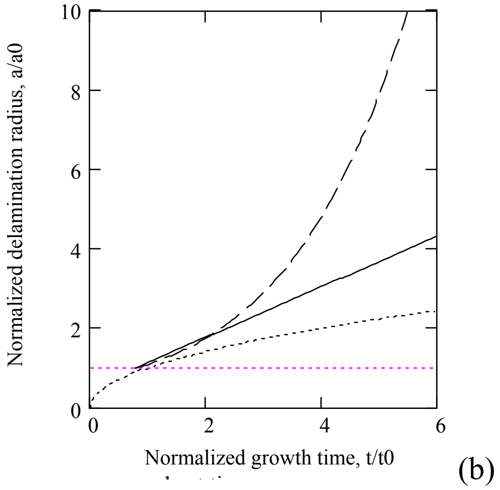
Figure 3. Dependence of dimensionless incubation time, $t' = t_i/t_0$, on normalized hydrogen concentration, $c' = c_0/c_*$, in the approximation of long (curve 1) and short (curve 2) times.

Figure 4 shows dependence of the normalized delamination radius, $a' = a/a_0$, on the normalized growth time, t'/t_0, in the long (lines 1) and short (lines 2) time approximation for three values of the initial fluid concentrations: (a) large, $c_0' = 3.33$, (b) intermediate, $c_0' = 2/3$, and (c) small, $c_0' = 0.133$.

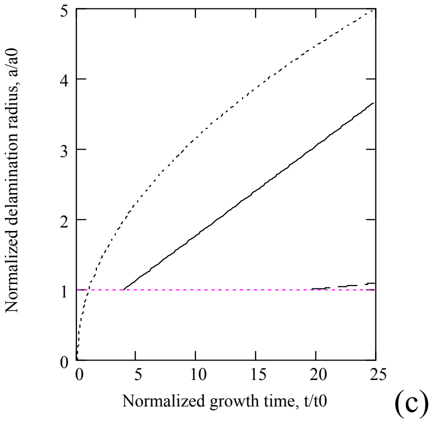
$c_0' = 5c_m' \approx 3.33$



$c_0' = c_m' = 2/3$



$c_0' = c_m'/5 \approx 0.133$



Legend:

—— 1 - long time
— — 2 - short time
------ 3 - relaxation time

Figure 4. Dependence of the normalized delamination radius, $a' = a/a_0$, on the normalized growth time, t'/t_0, in the long (lines 1) and short (lines 2) time approximation

5. CONCLUSIONS

Our results show that in the long-time asymptotic approximation (based on the quasi-static solution), the diffusion-controlled delamination propagates with a finite velocity, which remains constant during the growth. In this paper, we determine a maximum critical concentration that limits the use of the quasi-static solution. A transient solution, representing a short-time asymptotic approximation, is used when the concentration of gas exceeds the critical concentration. We then match these two end-member cases by using the method of Pade approximations and present closed-form solutions for both internal and near-surface diffusion-controlled crack propagation at different time scales.

An application of the developed asymptotic modes is demonstrated on the example of propagation of the near-surface and internal cracks driven in metal by the diffusion of the atomic hydrogen (protons). We show that for typical properties of low alloy steels in hydrogen embrittlement conditions, the approximation of long times is usually valid. However, depending upon the parameters, the metal durability (life-to-failure) varies rather considerably, i.e., from hours to decades.

The obtained results reveal some intriguing features worth checking experimentally. For example, the main kinetic equations for growth of the *near-surface* (delamination) and *internal* fractures [*Goldstein et el.*1985] are essentially identical, despite the difference in the problem geometry. Consequently, although the driving pressures for identical axisymmetric fractures located far and close to the half-space boundary are very different, their radii and velocities are exactly the same. This, perhaps, indicates that the approach adopted in this work is sufficiently robust to simulate delaminations of not necessarily small thicknesses.

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