

INTERACTION BETWEEN HYDROGEN DIFFUSION AND CRACK GROWTH IN HYDROGEN ASSISTED CRACKING

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ABSTRACT

Hydrogen diffusion and crack growth are coupled processes in hydrogen assisted cracking so that the particular histories of evolution of both stress intensity factor K and crack size a influence crack growth rate v at the same instantaneous K -value. Consequently, the crack growth kinetics curve $v = v(K)$ in general does not possess intrinsic character as a *unique* material's characteristic curve. However, a special regime of steady-state crack growth is seen to exist for which hydrogen assisted cracking turns to be a really K -dominated process. The correspondent plot of steady-state v - K acquires the uniqueness of a material's characteristic curve which may be used in engineering to provide conservative assessment of materials resistance and structural integrity under hydrogen assisted cracking conditions.

KEYWORDS

Hydrogen assisted cracking, crack growth rate, K -dominance, hydrogen diffusion

INTRODUCTION

The fracture mechanics approach is widely used to account for hydrogen assisted cracking (HAC) in assessment of integrity of materials and structures (Gangloff, 1988). It is based on the crack growth kinetics (CGK) curve "crack growth rate (CGR) v vs. stress intensity factor (SIF) K ", $v = v(K)$, as an intrinsic characteristic of the material-environment system. This suggests that equal SIF values induce the same CGRs in a given material under fixed environmental conditions irrespective of geometry and mode of loading of a specific body, i.e., the CGK curve possesses the *uniqueness* of a constitutive-type relationship. In other words, HAC is considered to be a K -dominated process.

Although ample experimental data support this approach (cf. Gangloff, 1988), some experimental observations raise doubts on the intrinsic character of CGK curves for environmentally assisted cracking in general (Kharin and Toribio, 1996) and reveal uncertainty of cracking characterization, since the same SIF values do not always result in equal CGRs for a given material-environment couple.

Experienced deviations are rather systematic in contrast to obvious statistical scatter of test data. Thus, fracture mechanics appears to be not fully capable of treating HAC even under small scale yielding and the matter of K -dominance over the entire HAC process becomes an open issue.

As a matter of fact, crack propagation and hydrogen diffusion are coupled in HAC and consequently the process history turns to be a factor capable of affecting near tip hydrogenation and thus the kinetics of HAC, as discussed in a previous work (Toribio and Kharin, 1996a). The objective of the present paper is to study hydrogen diffusion in the vicinity of a growing crack and to elucidate the role of history in HAC. This would provide more insight into the matter of uniqueness of the CGK curve, i.e., about its intrinsic character dependent —by hypothesis— solely on the specific material-environment couple.

HYDROGEN DIFFUSION ANALYSIS FOR A GROWING CRACK

Crack size is assumed to be a smooth function of time $l = l(t)$, the CGR being its ordinary time derivative. To study near-tip hydrogenation, it is suitable to consider a movable rectangular coordinate system (x, y) attached to the crack tip. This will cause transformation of the equation of stress-strain affected hydrogen diffusion (Kronshtal and Kharin, 1992; Toribio and Kharin, 1996b) associated with arbitrary stationary coordinates (x_1, x_2) pinned down to a solid. The latter system may be attached so that $x = x_1 - l(t)$, $y = x_2$. Then the total time derivative of hydrogen concentration C is:

$$\dot{C} = \frac{\partial C}{\partial t} - \frac{dl}{dt} \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} - v \frac{\partial C}{\partial x} \quad (1)$$

which yields the following equation of stress-strain assisted diffusion in moving coordinates (Toribio and Kharin, 1996a):

$$\frac{\partial C}{\partial t} = -\nabla \cdot \left[\left(\frac{D}{RT} X_D - v_x \right) C \right] \quad (2)$$

where D is the hydrogen diffusion coefficient, R the ideal gas constant, T the absolute temperature and X_D the thermodynamic driving force for diffusion. The vector $v_x = v j_x$ is independent of spatial coordinates and colinear with the x -axis whose unit vector is j_x .

Equation (2) takes the same form as that for a steady coordinate system by using formally the fictitious diffusion driving force:

$$X_D^* = X_D - \frac{RT}{D} v_x \quad (3)$$

It may be expressed by analogy with the true driving force X_D given in terms of hydrogen solubility factor K_s (cf. Kronshtal and Kharin, 1992; Toribio and Kharin, 1996b) responsible for stress and strain effect on diffusion:

$$X_D^* = -RT \nabla \left[\ln \left(\frac{C}{K_s} \right) + \frac{v}{D} x \right] = -RT \nabla \left[\ln \left(\frac{C}{K_s^*} \right) \right] \quad (4)$$

where the formal solubility-like term K_s^* is:

$$K_s^* = K_s^*(\sigma, \varepsilon_p, v) = K_s(\sigma, \varepsilon_p) \exp \left(-\frac{v}{D} x \right) \quad (5)$$

where σ and ε_p are the hydrostatic stress and the equivalent plastic strain.

On the basis of relations (3) and (4) and assuming a constant diffusion coefficient, further transformations of equation (2) yield:

$$\frac{\partial C}{\partial t} = D \left[\nabla^2 C - M^* \cdot \nabla C - N^* C \right] \quad (6)$$

where the coefficients M^* and N^* are correspondingly the following:

$$M^* = \nabla \left[\ln K_s^*(\sigma, \varepsilon_p, v) \right] \quad \text{and} \quad N^* = \nabla^2 \left[\ln K_s^*(\sigma, \varepsilon_p, v) \right] \quad (7)$$

Assuming K -dominance over the near-tip stress-strain field under small scale yielding, the equation coefficients (7) in the vicinity of the crack tip depend parametrically on SIF and CGR (apart from spatial coordinates):

$$M^* = M^*(K, v) \quad \text{and} \quad N^* = N^*(K, v) \quad (8)$$

Therefore, concentration of hydrogen near a moving crack tip determined by the equation (6) of stress-strain assisted diffusion should have not only SIF, but also CGR, among its governing parameters, i.e., $C = C(x, t; K, v)$. For HAC, v becomes one more unknown *a priori* variable in the coupled problem of hydrogen diffusion and crack growth. To close the system of equations of this problem, a criterion of crack growth is needed. It may be formulated in general form as:

$$C(x_c, t; K, v) = C_{cr}(K, x_c) \quad (9)$$

i.e., crack extends so that hydrogen concentration C reaches a critical value C_{cr} at some specified distance $x = x_c$ from the tip (cf. Toribio and Kharin, 1996a, b).

On establishing the right-hand part of relation (9) this formulation of the coupled diffusion-cracking problem is completely closed if SIF is somehow known, e.g., is maintained constant as in some fracture mechanics test specimens (cf. Ahmad and Ashbaugh, 1982). For this case, if the left-hand part of expression (9) is a solution of the diffusion equation (6) parametrically dependent on v , the CGR comes from equation (9) as:

$$v = v(K, t) \quad (10)$$

That is, CGR must not be the same at equal values of SIF, but varies with the HAC process time t .

However, in a majority of situations SIF depends on both applied load and current crack length. Then the problem is similar to the one of $K = \text{const}$, but now the left-hand part of equation (9) to determine CGR becomes not a function parametrically

dependent on K and v , but a functional over the whole history of the process. Correspondingly, instantaneous CGR $v(t)$ as a solution of this coupled diffusion-cracking problem also becomes a functional dependent on the process history from its very beginning in a particular solid with its individual previous variations of $K(t')$ and $v(t')$ during time $t' < t$. It looks then quite doubtful that resulting CGR could take the same values at equal SIF values if they were approached along different process histories. On the contrary, variability of CGR at the same SIF must be unavoidable. The question is would it be significant quantitatively or a variety of diffusion controlled CGK curves (10) can be always enclosed within a reasonably narrow scatter band around some definite single curve $v = v(K)$.

SIGNIFICANCE OF THE HISTORY EFFECT ON CRACK GROWTH

Clear parallelism may be noticed between the key equation (6) for a moving crack and that derived earlier for diffusion near a stationary crack tip (Toribio and Kharin, 1996b). The only difference is that in the latter case the coefficients (7) depend on the actual solubility factor $K_s(\sigma, \epsilon_p)$. If the variation of CGR (10) with time is slight enough, then under sustained SIF the coefficients (7) may be assumed to be nearly time independent, too. This allows one to solve equation (6) approximately using the same formalism as for stationary crack under sustained load (Toribio and Kharin, 1996b), as follows:

$$C(x, t; K, v) = C_e K_s^*(x; K, v) \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}}\right) \quad (11)$$

where C_e is the equilibrium hydrogen concentration in a stress-strain free metal provided by the crack tip environment (environmental hydrogen activity), and $\operatorname{erfc}(\bullet)$ the complementary error function. Taking into account expression (5):

$$C(x, t; K, v) = C_e K_s(x; K) \exp\left(-\frac{v}{D}x\right) \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}}\right) \quad (12)$$

Using this solution in the left-hand part of the crack growth criterion (9), the CGR may be obtained for a given SIF:

$$v(K, t) = -\frac{D}{x_c} \ln \left[\frac{C_{cr}(K, x_c)}{C_e K_s(x_c, K) \operatorname{erfc}\left(\frac{x_c}{2\sqrt{Dt}}\right)} \right] \quad (13)$$

which has physical sense only after some incubation period t_{in} have elapsed, i.e., at $t \geq t_{in}$. For earlier times $t < t_{in}$ it gives negative CGR values when expression in square brackets in formula (13) exceeds unit or, equivalently:

$$\operatorname{erfc}\left(\frac{x_c}{2\sqrt{Dt}}\right) < \frac{C_{cr}}{C_\infty} \quad (14)$$

where:

$$C_\infty = C_e K_s(x_c, K) \quad (15)$$

is the steady-state solution for hydrogen diffusion near a stationary crack (Toribio and Kharin, 1996b) attained at $t \rightarrow \infty$.

Relation (14) yields:

$$t_{in} = \frac{1}{4D} \left[\frac{x_c}{\operatorname{erfc}^{-1}\left(C_{cr}/C_\infty\right)} \right]^2 \quad (16)$$

where $\operatorname{erfc}^{-1}(\bullet)$ is the function inverse to erfc .

From expression (13) it follows that CGR at fixed SIF is a rising function of time which starts to increase from $v = 0$ at $t = t_{in}$ and approaches asymptotically some steady-state value v_{ss} as $t \rightarrow \infty$. Expression (13) renders:

$$v_{ss} = -\frac{D}{x_c} \ln \left[\frac{C_{cr}(K)}{C_e K_s(K)} \right] = \frac{D}{x_c} \ln \left[\frac{C_\infty(K)}{C_{cr}(K)} \right] \quad (17)$$

The validity of relations (11)-(13) and (16)-(17) relies strongly on the assumption about negligible time dependence of the diffusion equation coefficients (7), which presumes the same regarding CGR, i.e., $v(t) = \text{const}$ with reasonable accuracy. However, the obtained result does not confirm this hypothesis, since time variation of CGR according to (13) does not seem to be as slight as necessary for acceptable accuracy of expression (11) and the subsequently derived formula (13) for CGR itself.

Nevertheless, in spite of the roughness of this solution for the coupled diffusion-cracking problem, it clearly proves the intrinsic variability of CGRs values at a fixed SIF level. Hence, in general, CGR is not a single-value function of SIF, and CGK curve does not possess the intrinsic uniqueness as a characteristic curve of a material-environment system.

It is interesting to discuss the two limit situations. For applied K -values close to the threshold SIF K_{th} (at which $v = 0$), critical concentrations approach the steady-state level: $C_{cr}/C_\infty \rightarrow 1$ as $K \rightarrow K_{th}$. On the other hand, as $K \rightarrow K_c$, the latter being the fracture toughness of the material in air, less hydrogen is required to cause crack growth, i.e., $C_{cr} \rightarrow 0$, and $C_{cr}/C_\infty \rightarrow 0$, too.

Thus, the ratio C_{cr}/C_∞ is a monotonous diminishing function of SIF, and the same does the incubation time according to expression (16): for SIF values $K_1 < K_2$ corresponding $t_{in1} > t_{in2}$. At the same time, according to expression (17), the steady-state CGR is likely a rising function of SIF. This kind of K -dependence of both incubation time and CGR (theoretically predicted) agree fairly well with available experimental observations (Landes and Wei, 1973; Hudak and Wei, 1979) of HAC under constant SIF, sketched in Fig. 1 in a plot representing CGR vs. time for two different constant levels of K .

Therefore, although alterations of CGR may be generally different at variable SIF this particular case of $K = \text{const}$. provides evidence that CGK curves for a particular HAC process in a given material-environment system may diverge widely, in a noticeably band below steady-state CGR v_{ss} at the least.

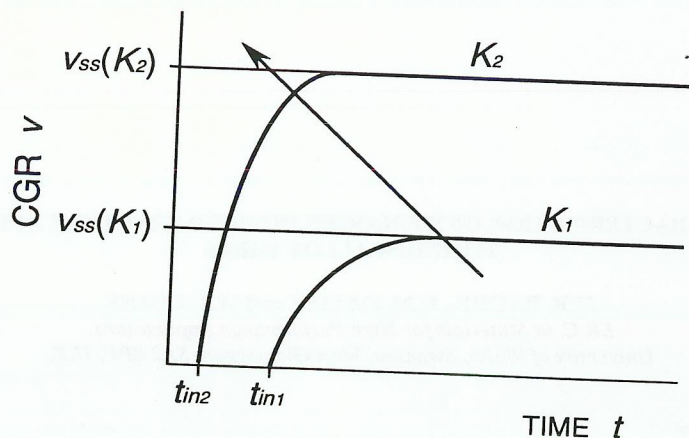


Fig. 1. Schematic variability of CGR in HAC tests with constant SIF according to available experimental data (Landes and Wei, 1973; Hudak and Wei, 1979) and theoretical analysis of the present paper (here $K_2 > K_1$ and the arrow indicates the tendency of shift of $v(K, t)$ -curves with increase of sustained SIF level).

ON THE STEADY-STATE REGIME OF HAC

Meanwhile, a way of HAC testing does exist which provides uniqueness of the CGK curve as a material's property, i.e., which ensures K -dominance over all the constituents of the HAC process. Namely, time dependent fractures of different natures (in particular, HAC) often stabilise and reach the regime of steady-state crack growth if controlling parameters of the process, such as SIF and hydrogen activity at the crack tip, are kept constant for sufficiently long time (cf. Kanninen and Popelar, 1985).

During such a steady-state regime of crack growth all near-tip processes are time independent when viewed by an observer fixed to moving crack tip. This obviously happens in HAC as follows from the existence of the steady-state solution (i.e., with $\partial C / \partial t = 0$ and $\partial v / \partial t = 0$) of the above considered coupled diffusion-cracking problem at constant SIF and crack tip environment. Indeed, diffusion equations for a crack moving with $v = \text{const}$ and for a stationary crack become nearly the same in the steady-state case, with insignificant distinction of solubility coefficients K_s^* and K_s regarding their spatial variation, cf. expression (5). Thus, the steady-state solution of equation (6) for moving crack may be obtained using the method used for a stationary one (Toribio and Kharin, 1996b) as follows:

$$C_{ss}(x_c; K, v) = C_e K_s^*(x_c; K, v) \quad (18)$$

Comparing this with the estimate (11) of the non steady-state solution of the same equation (6), one may conclude that the estimation (11) approaches at $t \rightarrow \infty$ the exact steady-state concentration distribution in the vicinity of a moving

crack tip. Consequently, with steady-state concentration from (18) and expression (5) for K_s^* , crack growth criterion (9) yields the steady-state CGR which is exactly the same as derived above (17) from estimation (13) of transient behaviour of CGR. Thus the approximate nonsteady solution (13) has the right asymptotic behaviour, too.

It is evident that the steady-state CGR (17) depends —apart from SIF— only on the characteristics of the material and the crack tip environment, such as diffusion coefficient D , specific scale x_c , hydrogen activity in near tip environment represented by C_e , and others which might be involved if the dependence of solubility coefficient K_s on stress and strain were considered explicitly (Kronsthal and Kharin, 1992). This CGR is a single-value function of SIF, and the CGK curve as a plot of steady-state values of CGR vs. SIF possesses the uniqueness of a material's characteristic curve. This stage of HAC process, the steady-state crack growth, appears to be really K -dominated.

CLOSURE

The coupled analysis of the interaction between hydrogen diffusion and crack growth reveals that K -dominance over the near-tip elastoplastic stress-strain field does not guarantee that the HAC process is K -controlled in general, i.e., a given SIF does not render a single value of CGR. Accordingly, plots of $v(K)$ -dependence cannot be considered as intrinsic curves which characterise HAC in particular material-environment systems. Instantaneous CGR at each reference SIF value turns to be a variable depending on initial conditions and subsequent history of cracking process in terms of time variations of $K(t)$ and $v(t)$.

However, a lot of test data and engineering experience confirm reasonable uniqueness of CGK curves and applicability of the fracture mechanics approach to HAC for a wide variety of situations, although numerous ambiguities have also been well documented, too (cf. Kharin and Toribio, 1996). This indicates, that intrinsic not K -controlled scatter of CGR under certain circumstances sometimes can be rather narrow so that the uniqueness of CGK curve may be granted with reasonable accuracy, e.g., within the band of statistically random scatter associated with cracking.

Furthermore, a special regime of steady-state crack growth does exist for which the distribution of diffusible hydrogen around the crack tip is totally determined by the SIF (apart from characteristics of the specific material and environment) and the CGR is consequently K -controlled. In this special case, whichever criterion of local fracture —dependent on the concentration— combined with the steady-state solution of the coupled diffusion-cracking problem yields a steady-state CGR v_{ss} being a single-value function of the SIF for a given material and specific environmental variables governing crack tip hydrogenation. This is a valuable attribute of the steady-state CGK curve which allows it to be adopted as a checkpoint to provide conservative evaluation of material susceptibility to HAC and structural integrity assessment.

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