ANALYSIS OF THE *K*-DOMINANCE CONDITION IN HYDROGEN ASSISTED CRACKING

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ABSTRACT

This paper summarizes the achievements of a basic research line in the field of environmentally assisted cracking in general and of hydrogen degradation in particular. It deals with the meaning and significance of the fracture mechanics approach to hydrogen assisted cracking, analyzing the question of *K*-dominance not only over the purely mechanical aspects, but also over the environmental (physico-chemical) events influencing the whole coupled process of hydrogenation and failure.

Two key factors able to violate the uniqueness of the crack growth kinetics curve $v = v(K)$ are discussed: the role of far field (the stress-strain field which is not *K*-dominated) and the effect of the history of hydrogenation and crack growth. Stress-strain assisted diffusion of hydrogen is considered as the rate-controlling factor of hydrogen assisted cracking under sustained or quasi-static loading conditions. The far field is shown to have a minor effect on near-tip hydrogen diffusion. It can only widen the scatter band of crack growth rates in the near-threshold portion of the $v(K)$-curve.

With regard to the effect of history, the study reveals that hydrogenation and crack growth are coupled processes, one influencing the other, so the crack growth kinetics curve $v = v(K)$ is not unique as an intrinsic material property must be. However, a special regime of steady-state crack growth is seen to exist in which hydrogen assisted cracking turns out to be a *K*-dominated process, and the corresponding plot of the steady-state v against K acquires the uniqueness of a material's characteristic curve which may be used in engineering to provide more conservative evaluation of material resistance and structural integrity.

1 INTRODUCTION

Engineering design frequently involves problems of environmentally assisted cracking (EAC) in materials and structures, a phenomenon which appears in diverse forms such as stress corrosion cracking, hydrogen assisted cracking (HAC), etc. In this framework, the fracture mechanics approach has proved to be effective for material evaluation and structural integrity assessment. In the domain of linear elastic fracture mechanics under small scale yielding, the stress intensity factor K is the only parameter governing the stress-strain state in the vicinity of the crack tip. The key of the fracture mechanics approach to EAC is the crack growth kinetics curve: a plot of crack growth rate v vs. stress intensity factor K, between the threshold K_{th} and the fracture toughness K_c.

The idea of *uniqueness* of $v(K)$-curves and thresholds K_{th} as intrinsic characteristics of {material-environment} systems forms the backbone of the approach and ensures the soundness of applications in engineering design. This uniqueness ensures the *similitude* of crack behaviour in test specimens and in structural components in service, thereby providing *transferability* of laboratory testing data to real engineering structures. If the $v(K)$-curve including K_{th} is indeed unique for a given {material-environment} couple, any discrepancy between predicted and real behaviour should be attributed to roughness in the analysis or experimental scatter but not to the concept. Otherwise, conceptual weakness makes the predictions less reliable and calls for more constraints on testing and evaluation to obtain data of the crack growth resistance parameters in a specific aggressive environment under consideration.

The reliability of the fracture mechanics approach to EAC in engineering design was reviewed by Toribio and Kharin [1]. The results showed that the same stress intensity factor does not always yield equal crack velocities in otherwise identical couples {material-environment}. This uncertainty of the basic fracture mechanics characteristics of EAC produces loss of confidence in materials evaluation and structural integrity assessment.

To solve the described problem, a procedure is proposed by Toribio and Kharin [2] for engineering safe design against EAC. For a given {material-environment} system, the aim is to find the *worst* crack tip situation producing the fastest crack growth rate v_m attainable at each K. The corresponding master curve $v_m(K)$ is the envelope of all possible $v(K)$-curves for a {material-environment} couple and represents an intrinsic characteristic of the system: the weakest resistance to EAC.

The present paper reviews some research work by the authors on the meaning and significance of the fracture mechanics approach to the particular phenomenon of HAC (very important in engineering). The final aim is to elucidate the question of K-dominance not only over the mechanical aspects of the phenomenon but *also* over the environmental (physico-chemical) events affecting the coupled process of hydrogenation and failure.

2 FRACTURE MECHANICS APPROACH TO HAC

To have K-dominance over the whole HAC process, and thus uniqueness of the $v(K)$ curve as an attribute of a given {material-hydrogen} system, two preconditions must be fulfilled (cf. Toribio and Kharin [3] for details and nomenclature):

/1/ K-controlled uniqueness of critical hydrogen concentration C_{cr} which must be reached at some point of the fracture process zone to cause local fracture and crack advance;

/2/ K-dominance over the process of hydrogenation in the fracture process zone ahead of the crack tip, i.e., uniqueness of the evolution of the distribution of hydrogen concentration $C(x,t)$.

Since the critical concentration C_{cr} in the vicinity of the crack tip inherits the property of K-dominance from the stress-strain fields, the two preconditions /1/ and /2/ formulated above are equivalent to the following ones:

/1*/ K is the only variable which controls the near-tip stress-strain fields (i.e., $\sigma = \sigma(K, \boldsymbol{r})$ and $\varepsilon_p = \varepsilon_p(K, \boldsymbol{r})$) in a domain embracing the physical process zone, thus dominating the mechanical aspects of both pre-damage and stress-strain affected hydrogenation;

/2*/ in addition, K also governs the environment parameters which control hydrogenation in the very close vicinity of the crack tip (*local* environment parameters) which determine the hydrogen concentration evolutions $C(x,t)$ in the fracture process zone.

Finally, the two preconditions may be formulated in terms of fracture mechanics concepts as:

/1**/ K-controlled *mechanical autonomy* of the crack tip region, i.e., K-dominance over the stress-strain field, which is the keystone of linear elastic fracture mechanics under small scale yielding;

/2**/ K-controlled *environmental autonomy* of the crack tip region, i.e., K-dominance over the physico-chemical events, namely the boundary conditions and the hydrogen diffusion in the vicinity of the crack tip.

While the first precondition is always fulfilled in the framework of linear elastic fracture mechanic analyses under small scale yielding, the achievement of the second one is not guaranteed in all cases, because many causes may destroy the K-dominance in HAC and thus the uniqueness of the crack growth kinetics curve $v=v(K)$. In further sections of this paper, the effects of far field and history are discussed in depth.

However, in spite of the fact that the $v(K)$-curve is not always K-dominated (the second precondition could fail), on assuming sustained loading conditions (as in the approach presented in this paper) the threshold K_{th} always meets the two preconditions of K-dominance, because the limit of crack non-propagation is associated with the unique steady-state distribution of concentration $C_\infty(\boldsymbol{r})$ near a tip of a stationary crack reached at $t \rightarrow \infty$. This stationary solution of the equation of stress-strain assisted diffusion has the same self-similitude properties as the stress-strain field. Therefore, only the first precondition (mechanical autonomy) is necessary to guarantee the uniqueness of the threshold state which is always K-dominated when linear elastic fracture mechanics, small scale yielding and sustained loading are considered.

3 THE EFFECT OF FAR FIELD ON *K*-DOMINANCE IN HAC

The matter of K-dominance with regard to hydrogen diffusion in the near-tip region is addressed by Toribio and Kharin [4]. It is the question of the accuracy of the approximate hydrogen concentration distribution C_a governed by the K-dominated component of the near-tip stress-strain field (i.e., by the *asymptotic* term σ_a) to represent the actual concentration C_f being driven by the complete mechanical field containing the whole series expansion (i.e., influenced by the non-autonomous *far field* σ_f). To this end, the discrepancy $\mathcal{E} = C_a - C_f$ between the two concentration distributions is the subject of interest to estimate the effect of the far field (the remote stress-strain field which is not K-controlled) on hydrogenation of the fracture process zone.

As explained in detail in [4], the following equation can be derived regarding the discrepancy:

$$\frac{\partial \mathcal{E}}{\partial t} = D\,[\,\nabla^2 \mathcal{E} - \mathbf{M}_f \bullet \nabla \mathcal{E} - N_f \mathcal{E}\,] + \nabla D \bullet [\,\nabla \mathcal{E} - \mathbf{M}_f \mathcal{E}\,] + Q \tag{1}$$

where an additional source-type term arises (cf. [4]):

$$Q = \begin{cases} D\Omega\, \nabla(\sigma_f - \sigma_a) \bullet \nabla C_a \neq 0 & \text{at } r > R_{SIF} \\ 0 & \text{at } r \leq R_{SIF} \end{cases} \tag{2}$$

This expression represents the source term for diffusion of discrepancy $\mathcal{E}$ which is the difference between the approximate K-driven concentration distribution and the exact far-field affected one. With nil initial and boundary conditions for $\mathcal{E}$, its absolute value increases from zero with time since the source Q produces this imaginable "diffusable substance" in the solid. This "error" is generated by the source Q operative at long distances, and it diffuses from there towards the fracture process zone. The effect of the far field on K-dominance over hydrogen accumulation in the fracture process zone becomes more severe when a greater amount of "error" $\mathcal{E}$ can reach the near-tip region by "virtual" diffusion from the distant source (2) situated at long distance.

After estimation of the sizes of the crack tip zones and analysis of near-tip hydrogen diffusion [4], it was possible to obtain the time evolution of the asymptotic (K-driven) concentration C_a at the outer border of the fracture process zone where the effect from the source Q appears first as:

$$C_a\,(x{=}R_{FPZ},\, t) = C_a(\tau) = C_\infty(R_{FPZ})\ \mathrm{erfc}\left(\frac{1}{2\sqrt{\tau}}\right) \tag{3}$$

where *erfc*(•) is the complementary error function and τ the dimensionless time $\tau = Dt/R_{FPZ}^2$.

Fig. 1 shows a plot of this function $C_a(\tau)$ (solid line). Approximately for $\tau \geq 130$, the concentration C_a in the fracture process zone exceeds 95% of the steady-state level C_∞. From then on, its further variation lies within a 5%-scatter band near this hydrogenation limit, i.e., the increase of K-driven concentration in the fracture process zone falls into this 5% strip from the time moment t_{SS} (where *ss* indicates steady-state) given by:

$$t_{\mathrm{SS}} = 130\,\frac{R_{\mathrm{FPZ}}{}^2}{D} \tag{4}$$

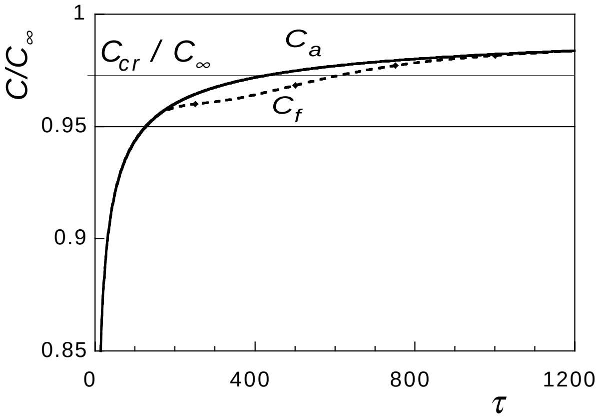

Fig. 1. Plot of hydrogen concentration evolutions with time: asymptotically-driven (K-controlled) concentration C_{a} (solid line) and a representative of the family of far-field affected concentration C_{f} (dashed line). An example of near-threshold critical concentration level C_{cr} below the steady-state one C_{∞} at K_{th} is also shown.

Thus the far field can affect the K-driven hydrogenation of the fracture process zone well after the transient concentration falls within a 5% scatter band near the steady-state hydrogen distribution in that area. Within the narrow 5%-width strip in the vicinity of the steady-state limit C_{∞} which defines K_{th} the supposition about K-control over hydrogen diffusion may be erroneous, as shown in Fig. 1 where the dashed line represents schematically one of the possible C_{f}-curves. This fact can affect the $v(K)$-curve only in its near-threshold part, but without affecting the threshold itself which remains K-controlled, as demonstrated in [4].

4 THE EFFECT OF HISTORY ON *K*-DOMINANCE IN HAC

Neither of the two processes of crack propagation and hydrogen diffusion should be considered separated from the other, since the movement of a crack tip is relevant to hydrogen accumulation in the fracture process zone. In terms of the theory of boundary value problems this situation is qualified as diffusion with moving boundary. Thus the crack growth *history* appears as a factor capable of affecting near tip diffusion and consequently the kinetics of HAC, apart from the stress-strain field represented by K. This again questions the idea of exclusive K-dominance over HAC.

The effect of history of the coupled hydrogenation-cracking process on K-dominance during HAC is addressed in previous research work by Toribio and Kharin [5,6]. The first paper [5] develops theoretical bases and a qualitative analysis demonstrating the coupling between hydrogenation and crack growth, so that the crack growth rate in HAC generally is not governed solely by K. The quantitative consideration is given in the second paper [6] where the K-dominance condition is analyzed, showing the significance of the history effect which destroys K-

control over crack growth rate, although a special regime of steady-state crack growth is seen to exist for which hydrogen assisted cracking becomes a K-dominated process.

When a moving crack is considered, the diffusion problem may suitably be considered in a movable coordinate system (x, y) *attached to the crack tip* so that $x = x_1 - a(t)$, $y = x_2$. This will cause transformation of the diffusion equation associated with the coordinate system (x_1, x_2) pinned to the solid. Then the total time derivation of concentration is:

$$\overset{\bullet}{C} = \frac{\partial C}{\partial t} - \frac{\mathrm{d}a}{\mathrm{d}t}\frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} - v\frac{\partial C}{\partial x} \tag{5}$$

By analogy with the actual thermodynamic one, the *fictitious driving force* for diffusion may be formally considered:

$$\mathbf{X}_{\mathrm{D}}^{*} = \mathbf{X}_{\mathrm{D}} - \frac{RT}{D}\,\boldsymbol{v} \tag{6}$$

This formulation of the diffusion-cracking problem is completely closed provided the stress intensity factor K is somehow known. For such a case, if C is a solution of the diffusion equation parametrically dependent on crack growth rate v, the latter can be found as:

$$v = v\,(K, t) \tag{7}$$

Therefore, the crack growth rate must not be the same at equal values of K, but varies with the total HAC process time t. The crack growth rate v for a given K could be obtained as [6]:

$$v\,(K, t) = -\frac{D}{x_{\mathrm{c}}}\,\ln\left[\frac{C_{\mathrm{cr}}\,(K, x_{\mathrm{c}})}{C_0\,K_{\mathrm{S}}\,(x_{\mathrm{c}}, K)\ \mathrm{erfc}\left(\dfrac{x_{\mathrm{c}}}{2\sqrt{Dt}}\right)}\right] \tag{8}$$

which has physical sense only after some incubation period t_{in} has elapsed, i.e., at $t \geq t_{\mathrm{in}}$.This happens for:

$$t_{\mathrm{in}} = \frac{1}{4D}\left[\frac{x_{\mathrm{c}}}{\mathrm{erfc}^{-1}\,(C_{\mathrm{cr}}/C_{\infty})}\right]^2 \tag{9}$$

where the function $\mathrm{erfc}^{-1}(\bullet)$ is the one inverse to $\mathrm{erfc}\,(\bullet)$ and $C_{\infty} = C_0\,K_{\mathrm{S}}\,(x_{\mathrm{c}}, K)$ is the exact steady-state solution for hydrogen diffusion near a stationary crack attained at long time $t \to \infty$.

From relation (8) it follows that the crack growth rate at fixed K is a rising function of time which starts to increase from $v = 0$ at $t = t_{\mathrm{in}}$ and asymptotically approaches some *steady-state* value v_{SS} as $t \to \infty$:

$$v_{\mathrm{SS}} = -\frac{D}{x_{\mathrm{c}}}\,\ln\left[\frac{C_{\mathrm{cr}}\,(K)}{C_0\,K_{\mathrm{S}}\,(K)}\right] = \frac{D}{x_{\mathrm{c}}}\,\ln\left[\frac{C_{\infty}\,(K)}{C_{\mathrm{cr}}\,(K)}\right] \tag{10}$$

For a given {material-environment (hydrogen)} couple, the steady-state crack growth rate (10) is a single-value function of K, and thus the crack growth kinetics curve as a plot of v_{SS} vs. K possesses the uniqueness of a material's characteristic curve. This stage of the HAC process, the steady-state crack growth, appears to be really K-dominated.

5 CONCLUSIONS

The meaning and significance of the linear elastic fracture mechanics approach to hydrogen assisted cracking was revised for small scale yielding and stationary cracks under sustained or quasi-static loading.

The role of *far field* on near-tip diffusion of hydrogen is of minor importance and can affect the $v(K)$-curve only in its near-threshold part, but it has no effect on the threshold itself which is still governed by K.

The effect of *history* appears in the form of coupling between hydrogen diffusion and crack growth and loss of K-dominance over the crack growth rate v, although a K-controlled steady-state value v_{SS} does exist.

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