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ENVIRONMENTALLY ASSISTED CRACK GROWTH IN
STRUCTURAL ALLOYS: PERSPECTIVES AND
NEW DIRECTIONS

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Robert P. Wei and Richard P. Gangloff

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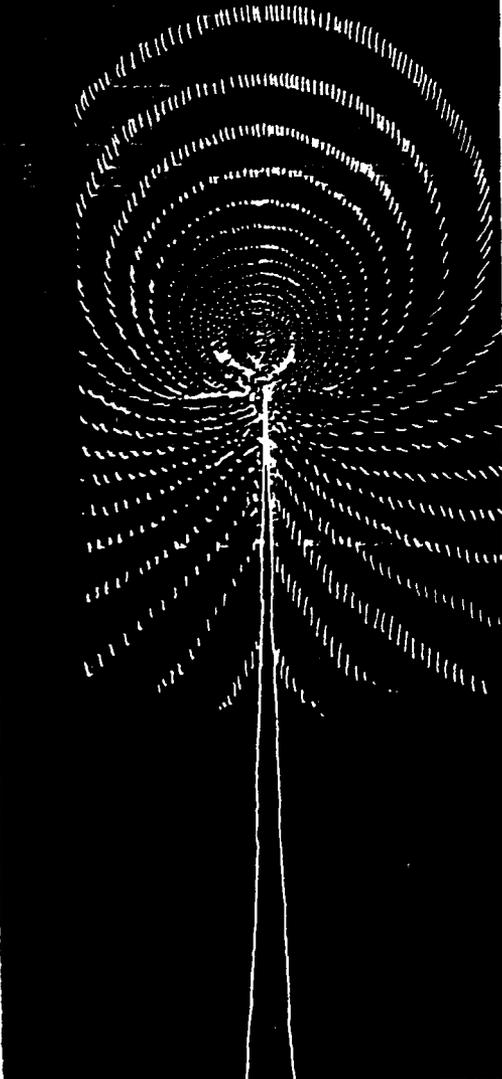| 1a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED		1b. RESTRICTIVE MARKINGS	
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Environmentally assisted crack growth (namely, stress corrosion cracking and corrosion fatigue) in alloys is one of the principal determining factors for durability and reliability of engineering structures. Over the past 20 years, activities in this area have transformed from principally that of screening and of qualitative characterizations of the phenomena, to that of quantitative assessment and scientific understanding. This work has enabled the recent development of life prediction procedures.

In this paper, the contributions of fracture mechanics in this transformation are reviewed. Current mechanistic understanding of environmentally assisted crack growth by hydrogen embrittlement is summarized, and is placed in perspective. Applications to mitigate stress corrosion and corrosion fatigue cracking in marine environments are summarized. Some outstanding issues and new directions for research are discussed.

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ENVIRONMENTALLY ASSISTED CRACK GROWTH IN STRUCTURAL ALLOYS: PERSPECTIVES AND NEW DIRECTIONS

Robert P. Wei[1/] and Richard P. Gangloff[2/]

INTRODUCTION

Environmentally assisted cracking of structural alloys (incorporating the well known phenomena of stress corrosion cracking and corrosion fatigue) is well recognized as an important cause for the failure or early retirement of engineering structures. Stress corrosion cracking (SCC) was first recognized as a technological problem in the last half of the 19th century as "season cracking" in cold drawn brass [1], with corrosion fatigue (CF) being recognized in the early 1900's [2,3]. Brown, tracing the historical background through 1972, concluded that "SCC, once thought confined to a few systems (combinations of metals and environments), must now be regarded as a general phenomenon which any alloy family may experience, given the wrong combination of heat treatment and environment" [1].

Bolstered by defense related interests and by safety issues in the energy industry, there was a decade of unusually high research activity from the early 1960's to the early 1970's [1]. Because of these interests and of the impact of the "energy crisis", work continued through the 1970's in support of offshore oil exploration and various alternative energy systems, such as, coal gasification and liquefaction, and solar energy. Coincidental to these activities, fracture mechanics was under-

going considerable development and maturation, and became increasingly accepted as an important tool in structural analysis and materials research.

In this paper a heuristic summary of the key developments in environmentally assisted crack growth in structural alloys is given, and key issues and new directions for research are discussed. The intent of this summary is to highlight the significant milestones and the contributions of fracture mechanics. As such, it does not include a complete chronology of all of the developments and contributions to this field. A complete view of these developments over the past 20 years may be gleaned from several monographs and from the proceedings of a large number of international conferences and symposia [1,4-23].

Chronologically, it is convenient to think of three periods, 1966 to 1972, 1972 to 1978, and 1978 to the present. Much of the groundwork in the United States was established during the first of these periods under two major programs, one sponsored by the Advanced Research Projects Agency of the Department of Defense (ARPA Coupling Program on Stress Corrosion Cracking) and the other by the Air Force Materials Laboratory [1,24]. It was during this period that the fracture mechanics methodology was first introduced.

Activities in the 1972 to 1978 period were devoted principally to phenomenological characterizations of cracking response, and to the development of empirically based design and failure analysis methods. Initial efforts toward quantitative mechanis-

tic understanding were launched, and science based approaches began to be formulated.

The past decade, beginning with 1978, is a period of transition. Scientific understanding of environmentally assisted crack growth and engineering application of this understanding are placed on a quantitative footing. Significant advances have been made, and are possible with the development of more sophisticated analytical instrumentation and experimental techniques. More emphasis is now needed to translate the improved understanding into methods for quantitative design and to improve structural reliability.

INITIATION vs PROPAGATION (Dawn of an Era)

A key turning point in the study of environmentally assisted crack growth and in the approach to design occurred in 1965. Brown and his coworkers [25,26] at the Naval Research Laboratory were encouraged to investigate the stress corrosion cracking susceptibility of titanium alloys by using specimens which were deliberately precracked in fatigue. Due to this initial crack, the alloy was highly susceptible and fractured in a matter of minutes, even though it appeared immune to stress corrosion cracking when stressed in the smooth (uncracked) state in the same electrolyte.

As a result of these findings, a major shift in emphasis was made from testing of smooth and mildly notched specimens to that of cracked bodies. Fracture mechanics was introduced as a basis for analyzing environmentally assisted cracking in a paper by H.

H. Johnson and P. C. Paris [27] in the First National Symposium on Fracture Mechanics in 1967. Experimental support for the use of the crack tip stress intensity factor (K) to describe the mechanical driving force was provided by Smith, Piper and Downey [28] for stress corrosion crack growth (Fig. 1), and by Feeney, McMillan and Wei [29] for corrosion fatigue (Fig. 2). A formal discussion of the use of crack tip stress intensity to describe the mechanical driving force for crack growth was given by Wei [30].

Early users of this emerging technology for environmental cracking investigations include Steigerwald [31], Hanna, Troiano and Steigerwald [32], Johnson and Willner [33], and Hancock and Johnson [34] for stress corrosion cracking (or crack growth under sustained loading), and *Bradshaw and Wheeler* [35], Hartman [36], and Li, Talda and Wei [37] for corrosion fatigue. Some early work predated the usage of the K concept. The effort associated with the ARPA Coupling Program made extensive use of fracture mechanics, and contributed to the development of fracture mechanics based technology for materials evaluation and for design [1].

PHENOMENOLOGICAL CHARACTERIZATION (1966 to 1972)

Two important approaches emerged from the early activities on stress corrosion cracking using precracked specimens [30,38]; namely the threshold and the kinetics approaches. The choice of a particular approach was determined in part by tradition and design philosophy, and in part by practical considerations of

experimentation and cost.

The simpler and less expensive approach involves the measurement of time-to-failure for precracked specimens under different applied loads, and the determination of a so-called threshold stress intensity factor (designated as K_{Iscc}), below which (presumably) no failure would occur by stress corrosion cracking [25,30,38,39]. The level of K_{Iscc} in relation to K_{Ic}, the plane strain fracture toughness of a material, provided a measure of stress corrosion cracking susceptibility. The use of the threshold approach was favored for material selection and for safe-life design.

The second approach was more complex, and involved the determination of crack growth kinetics [30,38]. It required the measurement of crack growth rate (da/dt) under controlled environmental conditions and as a function of the mechanical crack driving force, which is characterized by the stress intensity factor K. This approach required greater effort and more sophisticated instrumentation, and was favored for mechanistic studies and for fail-safe design. A similar distinction in approach existed for corrosion fatigue [40].

Both of these approaches were widely used. The period from 1966 to 1972 was principally devoted to phenomenological characterizations of materials response and to the development of empirically based design approaches. The final report for the ARPA Coupling Program, published as a monograph [1], reflects the typical efforts during this period.

It was during this period that a number of key studies [30,33-37,41-47] took place which helped to set the stage for the development of quantitative understanding of environmentally assisted crack growth over the next decade. These studies also showed the importance of the kinetics approach, and served to establish its use in subsequent investigations. Some of the important findings are as follows:

o There was an increasing awareness of the importance of the kinetics approach, and a recognition of the fact that stress corrosion crack growth progresses in three stages (Fig. 3) [31,41]. The particular significance of the K-independent Stage II in terms of mechanistic understanding of crack growth response was also recognized. The approach was used extensively by Speidel [42] to study the influence of halide ions on stress corrosion crack growth in high strength aluminum alloys, and by Wei and his coworkers [43-44] to examine hydrogen embrittlement of high strength steels.

o The effectiveness of using crack growth rate as a means for understanding the mechanisms for environmental enhancement of crack growth under sustained loading was demonstrated by Johnson and coworkers [33,34], and of fatigue crack growth by Bradshaw and Wheeler [35], Hartman [36], and Wei et al [37]. Through studies of the influences of different environments and of the inhibiting effect of trace amounts of oxygen, these researchers demonstrated the importance of surface reactions as a part of the embrittlement sequence (Fig. 4).

o Pressure and temperature were recognized as important probes for identifying the processes that control environmentally assisted crack growth under sustained loading [31,33,34,45, 46] and in fatigue [31,35,37,47].

o Early recognition of the relationship between stress corrosion and corrosion fatigue crack growth, and modeling of corrosion fatigue crack growth as a superposition of fatigue and stress corrosion cracking processes [48].

As an illustration of the phenomenological characterization effort, typical Stage II crack growth rate data for a high strength (AISI 4340) steel, stressed in various environments, are shown as a function of temperature in Fig. 4 [44,49]. These data demonstrated that the crack growth response can vary widely with environmental conditions, and can depend uniquely on temperature. Fractographic evidence suggested that the micromechanism for crack growth in these diverse, hydrogen producing environments was the same. Thus, the different response had to be attributed to one of several chemical processes in the overall chain.

Based on these initial findings, it was recognized that further progress in understanding environmentally assisted crack growth was not possible without integrating, at least, the fracture mechanics based kinetic measurements with independent measurements of the kinetics of participating chemical processes.

DEVELOPMENT OF A SCIENTIFIC APPROACH (1972-1978)

Beginning in 1972, a scientific approach to the study of environmentally assisted crack growth in high strength alloys began to be developed, and evolved over the following years [40,43,44,50,51]. The approach was grounded in linear fracture mechanics and was predicated on the recognition that environmentally assisted crack growth is the result of a sequence of processes and is controlled by the slowest process in the sequence [40,50].

The processes that are involved in the environmental enhancement of crack growth in high-strength alloys by hydrogen and hydrogenous gases (such as H_2O and H_2S) and by aqueous environments are as follows, and are illustrated schematically in Fig. 5 for the case of gaseous environments [40,50]:

1. Transport of the deleterious environment to the crack tip.

2. Reactions of the deleterious environment with newly produced crack surfaces to effect localized dissolution and to produce hydrogen.

3. Hydrogen entry (or absorption).

4. Diffusion of hydrogen to the fracture (or embrittlement) site.

5. Partitioning or distribution of hydrogen among various microstructural sites.

6. Hydrogen-metal interactions leading to embrittlement at the microstructural sites.

The actual processes depend on the mechanism of crack growth enhancement; namely, active path dissolution or hydrogen embrittlement. For a dissolution mechanism (see (6)), only the first two steps in the sequence need to be considered, and the

anodic (dissolution) reactions in the second step are directly responsible for crack growth enhancement. On the other hand, if hydrogen embrittlement is the responsible mechanism, then the reaction step serves only as the source for hydrogen; the remaining processes (3 through 6) must be considered. These steps are identical for aqueous and gaseous environments.

The overall crack growth response is governed by the rate controlling process in conjunction with the mechanical driving force, which is characterized here by either the local stress or the crack tip stress intensity factor, K [27,31,40].

Embrittlement, or the final step in the sequence is a function of microstructure. The extent of embrittlement, or the rate of cracking along each microstructural path, depends on the local hydrogen concentration, which depends in turn upon the external conditions (i.e., pressure, pH, electrode potential, temperature). Cracking along the various microstructural paths can take place concurrently (in parallel) with the overall crack growth rate given as the weighted average of the individual rates.

Clearly, the understanding of crack growth required identification and quantification of the rate controlling process. The evolving scientific approach, therefore, focused upon well defined and coordinated chemical/electrochemical, mechanical and metallurgical experiments.

It was recognized from the outset that the concept of rate controlling processes applied to both stress corrosion (or sustained-load) and corrosion fatigue crack growth, and that considerations of both modes of crack growth would afford a syner-

gism in exploring the fundamental issues. Because of the ready linkage of a K-independent regime (Stage II) of crack growth to the underlying rate controlling process, early efforts were directed at sustained-load crack growth. Detailed studies of the kinetics and mechanisms of reactions of water with iron and steel, and of Stage II crack growth were carried out as a function of temperature [51-55]. These efforts led to an unambiguous identification of the iron-water oxidation reaction as the rate controlling process for crack growth in high-strength steels in 1977, and a realization of the very limited extent of these reactions [6-10] (see Fig. 6).

With additional work on surface reactions and on crack growth, a broader based understanding of rate controlling processes emerged during the remainder of the 1970's, and modeling of sustained-load crack growth was begun. In 1977, this approach was used to examine the role of water vapor in enhancing fatigue crack growth in high-strength aluminum alloys [56], and the role of hydrogen sulfide in enhancing fatigue crack growth in steels [57]. These efforts led to the modeling of corrosion fatigue crack growth [58,59], which led in turn to the recognition and modeling of transport-controlled crack growth under sustained loading [60].

CHEMICAL AND MICROSTRUCTURAL MODELING

The modeling effort, which began in the late 1970's, emerged as an important adjunct of the scientific approach over the past 10 years. It has provided guidance and a formalized framework

for examining the fundamental issues, and has served as a basis for the utilization of data in design. Chemical and microstructural modeling of stress corrosion and corrosion fatigue crack growth are briefly summarized to provide perspective and to serve as a basis for discussing directions for future research.

Modeling of crack growth may be sub-divided in terms of loading conditions, and additionally according to the various processes that affect crack growth (see Fig. 5). Assuming hydrogen embrittlement as the responsible mechanism, modeling may then be grouped in terms of hydrogen supply (transport, reactions and diffusion), hydrogen distribution within the material (partition), and embrittlement reactions (which determine the rates of cracking along the various microstructural paths), and incorporates suitable methods for obtaining an average or macroscopic crack growth rate. Most of the effort during this period has concentrated on chemical modeling, and reflects developments in understanding the chemical and fracture mechanics aspects of environmentally assisted crack growth. More recently, this effort has been extended to include the influence of microstructure.

Sustained-Load Crack Growth

Initial modeling of sustained-load crack growth was largely phenomenological and was limited to the case of hydrogen-supply controlled Stage II crack growth in the lower temperature region (Region A in Fig. 6). The principal thrust was directed at obtaining reliable chemical reaction and crack growth data to

confirm the concept of rate controlled crack growth and to iden-
tify the controlling process. Obvious deviations of the crack
growth response curves (Fig. 6) from those of single rate con-
trolling processes and the observed changes in fracture paths (or
micromechanisms) led to the consideration of transfers of control
and of the role of microstructure.

Models Based On Hydrogen Supply. Models for Stage II crack
growth were proposed on the basis of extensive data on the kinet-
ics of surface reactions and crack growth for high-strength
steels in water/water vapor, hydrogen and hydrogen sulfide
[45,46,50,52-62]. When the reactions are slow (e.g., in hydrogen
and in water), Stage II crack growth rate is controlled by the
rate of surface reactions. On the other hand, for very rapid
reactions (e.g., in hydrogen sulfide), the growth rate is deter-
mined either by the rate of transport of the gases to the crack
tip, or by the rate of diffusion of hydrogen to the embrittlement
site. The models, expressing the specific dependence on gas
pressure (p_o) and temperature (T), are as follows [44,63]:

Transport Control: $\quad (da/dt)_{II} = C_t p_o / T^{1/2}$ \hfill (1)

$\qquad\qquad\qquad$ (for Knudsen flow [64])

Surface Reaction
$\qquad$ Control: $\quad (da/dt)_{II} = C_s p_o^m \exp(-E_s/RT)$ \hfill (2)

Diffusion Control: $\quad (da/dt)_{II} = C_d p_o^{1/2} \exp(-E_d/2RT)$ \hfill (3)

The constants C_i contain chemical and physical quantities that
relate to gas transport, surface reaction, etc., and reflect the
susceptibility of specific alloys to embrittlement by specific

environments (viz., the embrittlement reaction term). E_s and E_d are the activation energies for surface reaction and hydrogen diffusion respectively. Good agreement with experimental observations in the low temperature region (Region A) is indicated in Fig. 6.

In these models, a single process is assumed to be in control, and the terms C_i are assumed to be sensibly constant. It is recognized that transfer of control from one process to another may occur as the environmental conditions are changed. The consequences of this transfer have been discussed [63]. In formulating these phenomenological models, simple (single step) reactions were implicitly assumed. Because the reactions tend to be more complex, these models are viewed as starting points for developing further understanding of environmentally assisted crack growth.

Partitioning of Hydrogen and Surface Phase Transformation. The observed decrease in sustained-load crack growth rate with increasing temperature in the "high temperature" region (Region C in Fig. 6) for carbon martensitic steels (such as, AISI 4340 steel) has been analyzed based on surface chemistry [46,65,66]. Similar analyses have been attempted to account for the much steeper decrease in crack growth rate for 18Ni maraging steels [67]. These analyses, however, have ignored the important role of microstructure and micromechanism in hydrogen embrittlement, or have made unrealistic assumptions regarding surface coverage by hydrogen. The models proposed on the basis of these analyses, therefore, cannot explain the observed changes in fracture

mode with temperature [49].

To quantitatively account for the role of hydrogen-micro-structure interactions, a "hydrogen partitioning" model was developed [49]. The model suggested that the rate of hydrogen assisted crack growth is determined by two factors: (i) the rate of hydrogen supply to the fracture process zone, and (ii) the partitioning of hydrogen amongst different microstructural elements or traps (principally between prior-austenite grain boundaries and the martensitic matrix). The partitioning of hydrogen is controlled by hydrogen-trap interactions and determines the contribution by each element to the overall crack growth rate, which is the weighted average of rates of cracking along the different microstructural paths. This model is illustrated schematically in Fig. 7 for hydrogen assisted crack growth in a high strength steel. Detailed considerations and a derivation of the model are given in [49].

At low temperatures (in Region A of Fig. 6), hydrogen would reside primarily at grain boundaries and slip planes. Crack growth would tend to be predominantly intergranular (IG), and would conform to Eqns. (1) to (3). With increasing temperature (into Region C), the hydrogen supply processes remain in control, but hydrogen concentration at the grain boundaries and in the slip planes decreases and more hydrogen would reside in the martensite lattice. This temperature induced partitioning of hydrogen leads to increasing amounts of microvoid coalescence (MVC) and to slower crack growth rates. The change in crack

growth rate with temperature reflects a transfer of cracking mechanisms, rather than (or in addition to) a change in the process of hydrogen supply. The predicted temperature and pressure dependences for Stage II crack growth in high-strength steels were in good agreement with crack growth data for an AISI 4340 steel in hydrogen and in hydrogen sulfide (see, for example, Fig. 8) [49].

For the 18Ni maraging steels, a phase transition model was proposed to account for the abrupt decrease in crack growth rate that had been observed at the higher temperatures [50,68]. The model was based on the suggestion by Hart [69] that solute atoms can undergo a phase transformation at certain temperatures and pressures, and conformed with experimental observations.

Clearly, a number of factors can influence the kinetics of environmental crack growth. The hydrogen partitioning and surface phase transition models have provided some insight, and a clear indication of the need for a broadly based understanding, including that of the embrittlement mechanisms.

Fatigue Crack Growth

Based on the understanding developed for sustained-load crack growth, models for surface reaction and transport controlled fatigue crack growth were developed [58,59], and have been used successfully to explain the observed dependence of fatigue crack growth rates on cyclic load frequency and pressure in gaseous environments [56-59]. Insight obtained from this effort

has been applied to the consideration of corrosion fatigue in aqueous environments.

Superposition Model. Modeling was based on the proposition that the mechanical and environmental contributions could be decoupled such that the rate of crack growth in a deleterious environment, $(da/dN)_e$, may be written as the sum of three components, Eqn. (4) [40,70,71].

$$(da/dN)_e = (da/dN)_r(1 - \phi) + (da/dN)_c\phi + (da/dN)_{scc} \qquad (4)$$

In this equation, $(da/dN)_r$ is the mechanical fatigue rate; $(da/dN)_c$ is the "pure" corrosion fatigue rate; ϕ as the fractional area of crack that is undergoing pure corrosion fatigue; and $(da/dN)_{scc}$ is the contribution of sustained-load crack growth. These rates may be composed of contributions from several concurrent micromechanisms. For simplicity, the sustained-load growth term is not included in the following discussions. Eqn. (4) is rewritten as follows [66]:

$$(da/dN)_e = (da/dN)_r (1 - \phi) + (da/dN)_c\phi$$
$$= (da/dN)_r + [(da/dN)_c - (da/dN)_r]\phi \qquad (5)$$

or,

$$(da/dN)_{cf} = [(da/dN)_c - (da/dN)_r]\phi \qquad (6)$$

where $(da/dN)_{cf}$ denotes the incremental increase in growth rate above the reference level resulting from the embrittling environment.

In the limit for $\phi = 0$, or for a test in an inert environment, $(da/dN)_e = (da/dN)_r$ and corresponds to pure fatigue. For $\phi = 1$, corresponding to chemical reaction saturation [53,54], $(da/dN)_e = (da/dN)_{e,s} = (da/dN)_c$, and measured growth rates correspond to pure corrosion fatigue rates. In essence, the parameter ϕ represents material response to changes in environmental conditions. It is directly related to its counterpart, the fractional surface coverage (θ), in chemical modeling; i.e., $\phi = \theta$ [58,59]. The maximum in corrosion fatigue crack growth rate, therefore, corresponds to the maximum extent of chemical reaction ($\theta = 1$).

Chemical Modeling

Important understanding of corrosion fatigue crack growth response in gaseous environments was developed through chemical modeling (58,59) and through experimental verification of the role of gas transport and surface reactions on $(da/dN)_{cf}$ [56,57,72]. Similar understanding is being developed for aqueous environments [17,23-26].

Assuming that the environmental enhancement of fatigue crack growth results from embrittlement by hydrogen produced by the reactions of hydrogenous gases with freshly produced crack surfaces, models for transport and surface reaction were developed [58,59]. An analogous model for electrochemical reaction controlled crack growth was proposed for steels in aqueous environments, where the kinetics of reaction are assumed to be slow [73-75]. In these models, the environmental contribution is

assumed to be proportional to the extent of chemical or electro-chemical reaction per cycle, which is given by the fractional surface coverage θ, and the crack growth rate $(da/dN)_{cf}$ is given as by Eqn. (6) with $\phi = \theta$.

Models for diffusion controlled growth (76) and strain in-duced hydride formation [77-79] have also been suggested. The latter model relates to metallurgical changes and the consequent effect on crack growth rates, and is considered later. Diffusion controlled crack growth occurs when the preceding transport and surface reaction processes are rapid, and must be considered outside of the context of limited surface coverage per cycle.

<u>Transport controlled growth</u>. For highly reactive gas-metal systems, crack growth is controlled by the rate of transport of gases to the crack tip [58,59]. The surface coverage (θ) is linearly proportional to pressure (p_O) and inversely proportional to frequency (f). The environmental contribution to fatigue crack growth is given by the following relationships:

$$(da/dN)_{cf} = [(da/dN)_c - (da/dN)_r] \cdot [(p_O/f)/(p_O/f)_s] \qquad (7a)$$
$$\text{for } (p_O/f) < (p_O/f)_s$$

$$(da/dN)_{cf} = [(da/dN)_c - (da/dN)_r] = \text{constant} \qquad (7b)$$
$$\text{for } (p_O/f) \geq (p_O/f)_s$$

The term $[(da/dN)_c - (da/dN)_r]$ is the maximum enhancement in the rate of cycle-dependent corrosion fatigue crack growth, and is a consequence of the finite extent of surface reaction (i.e., $\theta \rightarrow 1$) [56,57]. The saturation exposure, $(p_O/f)_s$, is a function

of gas pressure, temperature, and molecular weight of the gas, and of stress intensity level, load ratio, yield strength and elastic constants; through their influences on crack geometry and gas transport [58,59,72].

Surface and electrochemical reaction controlled growth. With less reactive systems, crack growth is controlled by the rate of surface or electrochemical reactions at the crack tip. For simple first-order reactions, the crack growth rate in gaseous environments is given by Eqn. (8) in terms of pressure, frequency and the reaction rate constant k_c [58,59].

$$(da/dN)_{cf} = [(da/dN)_c - (da/dN)_r] \cdot [1 - \exp(-k_c p_o/f)] \qquad (8)$$

A more general interpretation of surface coverage can be made to accommodate multi-step reactions, with the actual response reflecting the nature and kinetics of the individual reaction steps.

For aqueous environments, $(da/dN)_{cf}$ may be expressed as an analogue to Eqn. (8) [75]:

$$(da/dN)_{cf} = [(da/dN)_c - (da/dN)_r] \cdot [q/q_s] \qquad (9)$$

where q is the amount of electrochemical charge transferred per cycle; q_s is the "saturation" amount or that required to complete the reactions; and the ratio q/q_s is identified with θ.

Diffusion controlled growth. When transport and surface reaction processes are sufficiently rapid, the crack growth rate is determined by the rate of diffusion of hydrogen from the crack

tip to the "fracture process zone". According to Kim (76), $(da/dN)_{cf}$ is given by Eqn. (10):

$$(da/dN)_{cf} = A_o exp(-H_B/RT) \cdot (p_o D/f)^{1/2} \Delta K^2 \qquad (10)$$

where A_o is an empirical constant, H_B is the binding enthalpy of hydrogen to dislocations, and R is the universal gas constant, D is the hydrogven diffusivity, and f is the frequency.

In the transport and reaction controlled models, a growth rate dependence upon ΔK^2 is implicitly assumed to reflect the expected proportionality between the sizes of the "hydrogen damaged" zone and the crack-tip plastic zone [59,72]. This dependence is explicitly incorporated in the diffusion controlled model [76]. The temperature dependence is reflected through its influence on the reaction rates, the fatigue process, the mechanical properties and on gas transport [58,59,73,74]. If the reaction mechanisms remain unchanged, the maximum enhancement in rate (or $(da/dN)_c$) is expected to remain constant. The temperature dependence for corrosion fatigue would be reflected principally through its frequency dependence, with the maximum remaining constant.

Experimental support. The transport and surface (and electrochemical) reaction controlled models have been examined by coordinated studies of the kinetics and mechanisms of gas-metal reactions, and of corrosion fatigue crack growth response as functions of pressure, temperature, and loading time (or frequency). Good agreement between these models and the experimental data on crack growth and surface and electrochemical reactions

has been obtained (see Figs. 9 to 11, for example). The trans-port controlled case is represented by aluminum and titanium alloys in water vapor (Fig. 9), and steels in hydrogen sulfide (Fig. 10) at low pressures; the reaction controlled case, by high strength steels in aqueous electrolytes (see Fig. 11, for example [80]).

It is recognized that the form of the crack growth rate response depends on the kinetics and on the specific nature and mechanism(s) of the reactions, and may reflect both transport and reaction control. For the case of reaction controlled crack growth, the response may reflect the fact that the reactions do not follow simple first-order kinetics, and the presence of more than one reaction step. For example, for the case of 7075-T651 aluminum alloy (Fig. 10), the additional enhancement at the higher pressures is surface reaction controlled and is attributed to a slow step in the reactions of water with segregated magne-sium [81]. Similarly, the increase in rate observed on the 2-1/4Cr-1Mo steel in hydrogen sulfide at the higher pressures is surface reaction controlled, and is identified with the slower second step in the reactions of H_2S with iron [57,60]. A similar situation exists for crack growth of high-strength steels in water vapor and in aqueous solutions. The situation in water vapor may be further complicated by capillary condensation at the crack tip [53,73,74].

Evidence for diffusion-controlled crack growth is provided by data on titanium alloys (see Fig. 12, for example). At the higher frequencies, $(da/dN)_{cf}$ was found to be inversely propor-

tional to the square root of frequency [82]. This dependence, coupled with the known reactivity of titanium, is consistent with diffusion control. The abrupt decrease in growth rates at the lower frequencies is attributed to a hydride mechanism that depends on both strain and strain rate [29-31,36]. There is, however, no quantitative model for hydride induced crack growth.

Microstructural Modeling

The important role of different micromechanisms was discussed by Gerberich and Peterson (83). The role of micromechanism (or of microstructure) is explicitly incorporated in Eqns. (4) and (6). The implications of the model are as follows: (i) the partitioning of hydrogen to the various microstructural sites need not be uniform, and (ii) the fractional area of fracture surface (ϕ) produced by pure corrosion fatigue is equal to the fractional surface coverage (θ) for chemical reactions. The relationship between the microstructural and environmental parameters (ϕ and θ) was examined by Ressler (84) and by Gao et al (85).

For an AISI 4340 steel in water vapor (585 Pa) at room temperature, Ressler (84) determined the corrosion fatigue crack growth rate as a function of frequency (Fig. 13). Fractographic data show a change in fracture surface morphology with decreasing frequency from a predominantly transgranular mode (relative to the prior-austenite grains) to one that is predominantly intergranular, Fig. 14. By identifying the intergranular failure mode with pure corrosion fatigue and the transgranular mode with

mechanical fatigue, the fraction of pure corrosion fatigue (ϕ), was estimated from the microfractographs. A comparison was made between ϕ and θ, based on independent surface reaction measurements [51] and an adjustment of exposure to account for capillary condensation in the fatigue crack (Fig. 15). Agreement is excellent. A similar good correlation was reported by Gao et al [85] for a 7075-T651 aluminum alloy.

These results indicate the important role of microstructure, and of the interactions between the environmental and microstructural variables.

More work is needed to broaden the scope of this understanding, and to provide statistically reliable support. Nevertheless, the framework for understanding has been set in place.

IMPLEMENTATION OF THE FRACTURE MECHANICS APPROACH (1980s)

Significant complexities must be overcome in implementing the fracture mechanics approach for the quantitative prediction of component life to control environmentally assisted crack growth. During the 1980s, two critical issues emerged. Firstly, the principle of fracture mechanics similitude (that is, equal subcritical crack growth rates are produced by equal applied stress intensities) may be violated because of the effects of crack closure and variations in environmental chemistry within the crack. Secondly, the large number of relevant variables and their time-dependent interactions greatly complicate life predictions, particularly for lower strength materials which were not extensively characterized during the 1970s and where linear frac-

ture mechanics may not suffice.

Mechanistic understanding and scientific modeling provide the means for characterizing subcritical cracking in terms of a scalable crack tip driving force, for extrapolating short term data to predict long term component behavior, and for defining the effects of mechanical, chemical and metallurgical variables.

Fracture Mechanics Similitude

The use of fracture mechanics similitude to scale environmentally assisted crack growth rates, for different crack sizes and loadings, is permitted only if the following two conditions are met: (1) the applied driving force parameter (e.g., stress intensity factor) uniquely defines the stresses, strains and strain rates near the crack tip; and (2) the composition and conditions of the environment at the crack tip are constant for a given applied driving force parameter, irrespective of crack size and opening shape. Investigations of environmentally assisted crack growth at low stress intensities, in low strength or anisotropic materials, for small crack geometries and in complex embrittling environments demonstrate that the validity of similitude cannnot be always assumed [16-18,86-88]. The applied stress intensity basis shown in Figs. 1 and 2 may need to be modified to reflect the actual crack driving force, as described in the following subsections.

Crack Closure Problem. Premature contact of fatigue crack surfaces during unloading or crack closure reduces the effective crack driving force relative to the applied value (e.g., ΔK).

For corrosion fatigue, likely crack closure mechanisms include environmentally assisted crack deflection, intergranular cracking and enhanced surface roughness, enhanced plasticity, fluid pressure, and corrosion product wedging [89,90].

The effect of corrosion product wedging is shown in Fig. 16. Here, the reduction in fatigue growth rates at low mean stress is attributed to wedging by a thin surface oxide produced by fretting of the fracture surfaces in moist or oxygenated environments [91]. This closure mechanism is relevant when the crack tip opening displacements are small, and may become more pronounced because of enhanced (crevice) corrosion within fatigue cracks. The effect was documented for cathodically polarized steels in seawater [92,93].

For laboratory specimens, crack closure effects are accounted for through measurements of specimen compliance [89]. The implications of environment induced closure for component behavior, however, are unclear and require attention. Even though elementary micromechanical models have been proposed for specific closure mechanisms, the environment sensitive processes are poorly understood [90]. The problem is exacerbated by the time dependent nature of corrosion processes.

Small Crack Problem. Because of the importance of early crack formation and growth to component fatigue life, considerable emphasis has been placed on the fracture mechanics of small cracks through the 1980s [87]. Generally, stress corrosion and corrosion fatigue cracks sized below 1 to 5 mm have been found to grow unexpectedly rapidly relative to long cracks at the same K

level, and to grow below the threshold K level for long cracks [87,90,94,95]. This behavior must be understood in order to apply the fracture mechanics approach to predict environmental cracking life in components.

The observed crack size effect may be caused by inappropriate formulation of the mechanical driving force and by differences in crack tip environment. A variety of mechanisms and bounding crack sizes were identified [96]. For fatigue in benign environments, increased crack tip plasticity, underdeveloped crack wake closure, three dimensional crack shape and large scale yielding can increase growth rates of small cracks. Interactions with grain boundaries, on the other hand, can arrest growth. The same mechanisms may affect environmentally assisted cracking through changes in crack-tip environment, environmental modification of crack closure, or strain enhanced film rupture [90,95,97-99].

The occluded environments within short cracks can differ from the bulk and from those within long cracks. For the cases examined to date, the short crack environments appear to be more deleterious. Data for a high strength steel in aqueous chloride solution (Fig. 17) show that stress intensity similitude was not obeyed, with K_{Iscc} decreasing with crack size below about 1 mm [100]. Values for K_{Iscc} at the smallest crack size were about 1/3 those obtained with specimens containing 15 to 30 mm long cracks. This crack size effect is chemical in origin, and has been predicted successfully through calculations of solution pH, electrode potential and total rate of hydrogen production within

cracks of varying length, and the use of an empirical relation-
ship between K_{Iscc} and adsorbed hydrogen concentration [100-102].

Gangloff showed that small corrosion fatigue cracks (below
about 3 mm) grew at rates up to 10^3 times faster than long cracks
at equivalent applied ΔK [103,104]. This effect is illustrated
in Fig. 18 by comparing data for 25 to 40 mm long cracks with
those of 0.1 to 3 mm long elliptical surface and through-thick-
ness edge cracks for a high strength steel in aqueous 3% NaCl
solutions. The concomitant absence of a crack size effect in
vacuum and moist air further demonstrates the chemical origin of
the breakdown in similitude. The chemical crack size effect has
been confirmed by literature data, and by experiments with lower
strength steels. The effect, however, decreased from a factor of
10^3 to 2 as the yield strength of the steel decreased [95].

The effect of crack geometry shown in Fig. 18 is qualita-
tively understood in terms of a complex interaction between mass
transport by diffusion, ion migration and convection, and elec-
trochemical reactions [94,104,105]. Initial effort at modeling
this effect has been made, but needs to be supported by critical
experiments [104-106]. Possible variations in crack chemistry
and their effects on the growth of longer cracks must be considered.

In essence it is necessary to examine the coupled reaction
and transport problem as a whole in dealing with the apparent
violation of similitude. Turnbull has attempted to do this by
modeling steady state reactions and mass transport in simple
trapezoidally shaped cracks [107,108]. This work must be modi-
fied to include the important transient reaction kinetics dis-

cussed in a previous section, and must be related to the microme-
chanics of crack advance. Such an approach will provide the
foundation for developing quantitative predictive methods for
long term component service.

Life Prediction for Environmental Cracking

The time dependence of environmental crack growth, the many
relevant variables and the complexities affecting similitude
hinder fracture mechanics life prediction. None-the-less, the
phenomenological and scientific foundations have been developed
during the past 15 years. The challenge is to refine and inte-
grate this understanding to produce methods which overcome the
weaknesses of smooth specimen, time-indifferent design rules.
Significant progress has been reported in this regard for stress
corrosion and corrosion fatigue in nuclear systems, based on film
rupture mechanistic modeling [109].

Recent advances and future directions for life prediction of
steels cracking by hydrogen embrittlement are reviewed for static
and cyclic loading.

Stress Corrosion Cracking. Stress corrosion cracking is
sensibly predicted based on the threshold stress intensity
(K_{Iscc}) concept, within the bounds summarized elsewhere [110].
Two variables critically affect K_{Iscc}: steel yield strength and
hydrogen uptake.

Lower bound relationships between K_{Iscc} and yield strength
are presented in Fig. 19, based on over 500 measurements reported
during the past 20 years [111]. The beneficial effect of de-

creasing strength is shown for five specific environments. For constant strength, K_{Iscc}, correlates with the steady state concentration of dissolved hydrogen, produced by gaseous chemical or aqueous electrochemical reactions on the input surface of a steel foil and measured by a permeation experiment, as illustrated in Fig. 20 [111,112]. Such data, when employed with permeation based sensors of hydrogen uptake in plant components [113], enable conservative life predictions for environmental cracking under monotonic loading. This approach may be compromised by cyclic loading, unique microstructures and new environment chemistries [110,111].

Corrosion Fatigue. Significant progress was achieved in the 1980s for fracture mechanics predictions of long term corrosion fatigue in offshore structural applications, particularly large welded joints between low strength carbon steel tubulars [109,114]. The elements of this approach are reviewed.

(1) Data base. Corrosion fatigue cracking in the low alloy steel/aqueous environment system occurs at stress intensities well below K_{Iscc} and within the regime relevant to tubular joint performance, as illustrated by the data and arrow in Fig. 21 [93,115-119]. Crack growth is enhanced by reduced loading frequency and increased cathodic polarization; such effects must be understood to predict the long term (10^8 cycles at 0.1 Hz) life of cathodically protected tubulars.

(2) Hydrogen embrittlement models. The aim of mechanistic modeling is to predict corrosion fatigue crack growth rate as a

function of stress intensity, stress ratio, environment composition, microstructure, electrode potential and loading frequency. No single, quantitative model exists. The foundation has been established, however, by research on crack tip reactions and the frequency dependence of corrosion fatigue [44,59,75], on crack closure [89], on steady state crack chemistry modeling [100,102,105-108], and on hydrogen embrittlement [115,120]. Mechanisms for near-threshold corrosion fatigue have been proposed also [90,91,93,94,115,121], however, quantitative formulations and experimental evaluations are lacking.

(3) Stress intensity and life predictions. To date, only approximate K solutions for complex tubular joints have been developed [122]. Such equations were integrated with laboratory crack growth rate data (Fig. 21) to yield the prediction shown in Fig. 22 [122].

(4) Full scale component testing. Sophisticated capabilities exist in several countries to conduct fatigue and corrosion fatigue experiments with large welded tubular joints, and to monitor crack growth continuously by electrical potential techniques [123-125]. As shown in Fig. 22, fatigue life data on full-scale joints are in excellent agreement with fracture mechanics predictions for cycle dependent cracking in moist air. Measurements of surface crack shape development are also consistent with model predictions.

Limited corrosion fatigue experiments on full-scale joints demonstrate the deleterious effect of the seawater environment,

potential problems due to cathodic protection and the inability of simple cycle based fatigue design rules to adequately describe tubular life [109]. Modeling of these effects requires improved crack growth rate data, mechanistic models and understanding of similitude, particularly for near threshold corrosion fatigue.

OUTSTANDING ISSUES AND NEW DIRECTIONS

The foregoing sections provide a perspective summary of the modeling effort to quantitatively connect chemical and physical processes with environmental crack growth response. From an initial effort that was narrowly focused on the rate controlling processes for crack growth in steels exposed to gaseous environments, ensuing studies broadened the understanding to cracking in high-strength aluminum and titanium alloys, and extended the approach to the complex problem of crack growth in aqueous environments. The simplified models serve as a framework for research and design, as illustrated for offshore structures. They serve also as a basis for furthering the understanding of environmentally assisted crack growth. Results from these studies will aid in the development of alloys and of methods to minimize sensitivity to environmentally assisted cracking, and of procedures for making reliable predictions of long-term service. Fracture mechanics has played a major role in the development of this understanding. Significant issues remain to be resolved.

(i) Detailed understanding of the kinetics and mechanisms of surface/electrochemical reactions with clean metal surfaces is needed, over a broad range of environmental conditions: (a) to

establish the form and quantity of hydrogen that is produced, and the fraction that enters the material to effect embrittlement, (b) to explore and confirm the indicated transfer of rate controlling processes with changes in environmental conditions, and (c) to determine the relationship between the kinetics of surface reactions and crack growth response to improve the predictive capability of crack growth models.

(ii) For environmentally assisted crack growth in aqueous environments, the traditional electrochemical measurement of polarization response is inadequate. A new technique for measuring the kinetics of equilibration reactions at the crack tip has been developed. These measurements, however, must be coupled with a detailed understanding of the reaction mechanisms and modeling of the crack tip chemistry.

(iii) Greater effort is needed to understand the factors and processes that control or inhibit electrochemical reactions of bare, straining metal surfaces with electrolytes.

(iv) Quantitative understanding of the physical-chemical interactions between hydrogen and metal (i.e., the embrittlement mechanisms) is needed to establish the roles of microstructure and of other metallurgical variables in determining the rate of crack growth, or the degree of susceptibility.

(v) Better understanding of the influences of alloying and impurity elements, and of microstructure is needed. It is essential to determine whether such influences on environmental cracking result from alterations of the reaction kinetics (chemical effects), from their influence on the mechanical properties of

alloys (physical effects), or both (physical-chemical effects).

(vi) Better understanding of the processes that control environmental crack initiation and early growth (viz., threshold and Stage I crack growth) is needed.

(vii) Mechanistic descriptions of crack chemistry, transient reactions and micromechanical embrittlement must be integrated to produce a predictive model of environmentally assisted crack growth rate. The model must quantitatively predict both specimen and component cracking performance, and must be amenable to experimental confirmations and mechanistic refinements.

(viii) Experimental research and modeling must be exploited to develop in-situ monitors of environmental crack growth in complex components. Sensors for hydrogen uptake from service environments must be further developed and implemented.

(ix) New experimental methods must be developed for direct measurement of environmental damage processes at the crack tip.

(x) Research on environmentally assisted cracking must be extended to include novel monolithic and composite materials.

Summary

Experimental and analytical work in the past decade has contributed significantly to both the phenomenological and mechanistic understanding of environmentally assisted crack growth. Crack growth response reflects the complex interplay among chemical, mechanical and metallurgical factors, and is dependent on the rate controlling processes, and on the micromechanisms for crack growth and the mechanisms of the relevant chemical reac-

tions. On the basis of this understanding, modeling of environmentally assisted crack growth, under sustained-load and in fatigue, has been made. This modeling effort has placed the study of this technologically important problem on a sound footing, and provides a framework for new understanding and for the development and utilization of data in design. To make significant further advances in understanding, continued emphasis on multidisciplinary approaches which incorporate chemistry, physics, materials science and fracture mechanics, and long-term support are essential.

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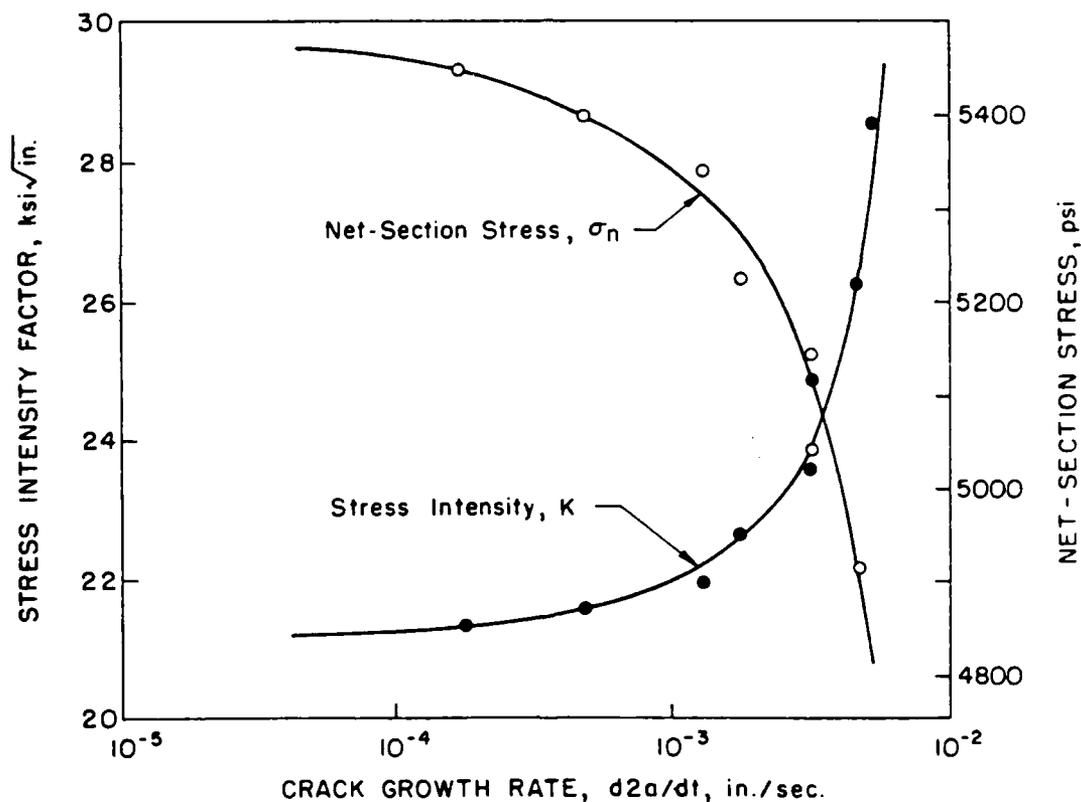
Fig. 1 - Stress intensity factor and net-section stress versus crack growth rate for a wedge-force-loaded specimen [28].
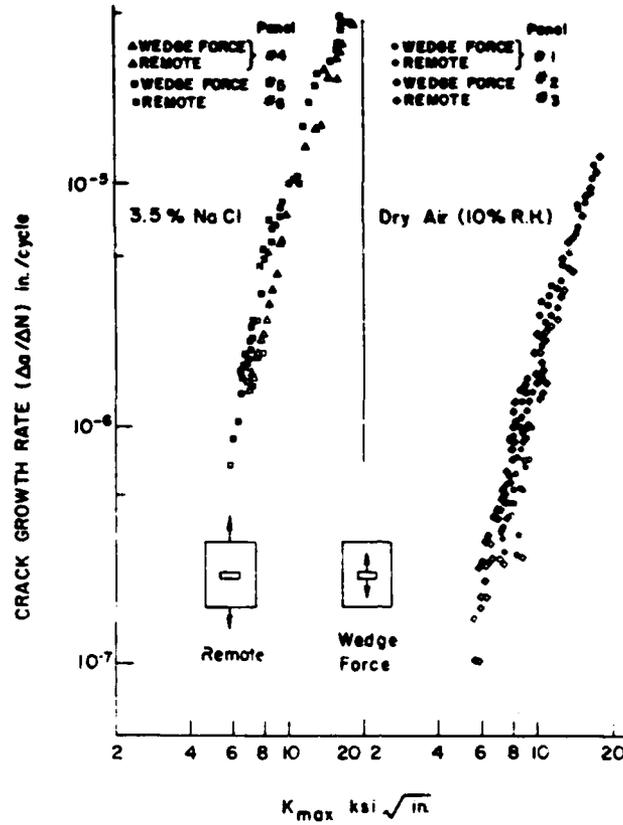
Fig. 2 - Compatibility data for aluminum alloy 7075-T6 upon intensity factor.
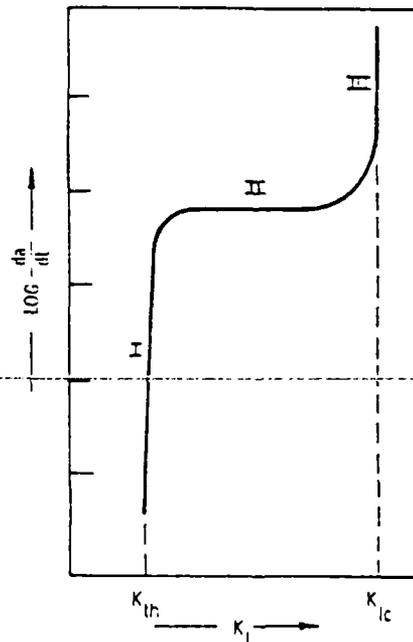
Fig. 3 - Schematic of monotonic load environment enhanced crack growth response.
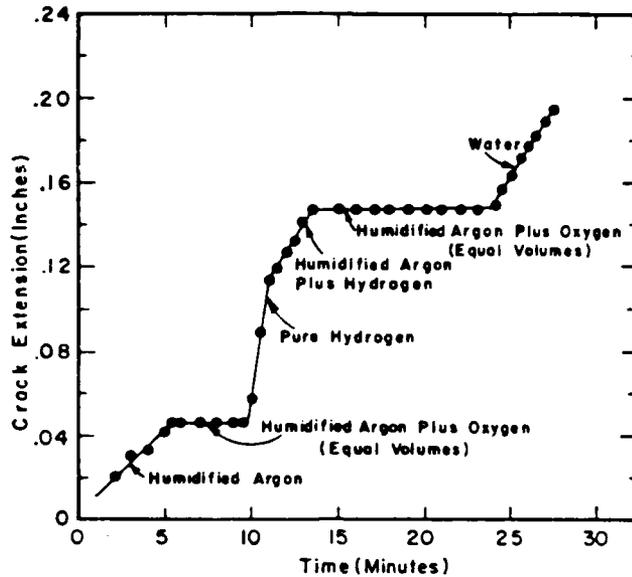
Fig. 4 - Subcritical crack growth in different water, water
vapor, hydrogen, and oxygen environments, H-11 steel,
230 ksi yield strength [34].
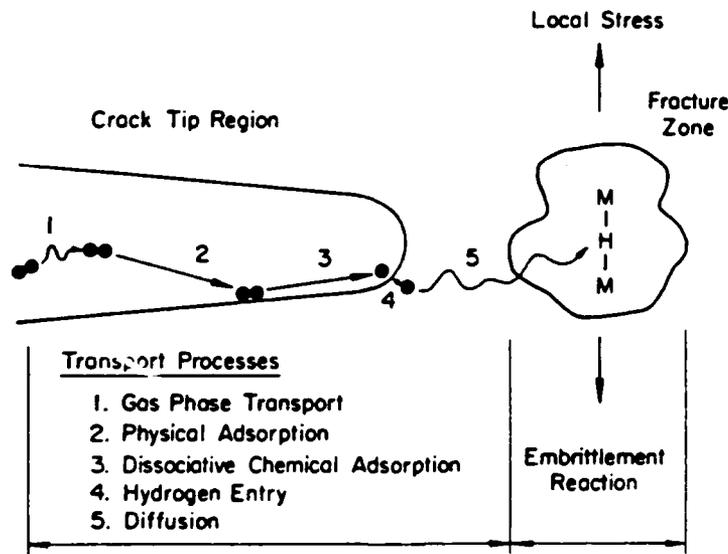
Fig. 5 - Schematic illustration of sequential processes in
environmental enhancement of crack growth by hydro-
genous gases. Embrittlement by hydrogen is assumed
and is schematically depicted by the metal-hydrogen-
metal bond.
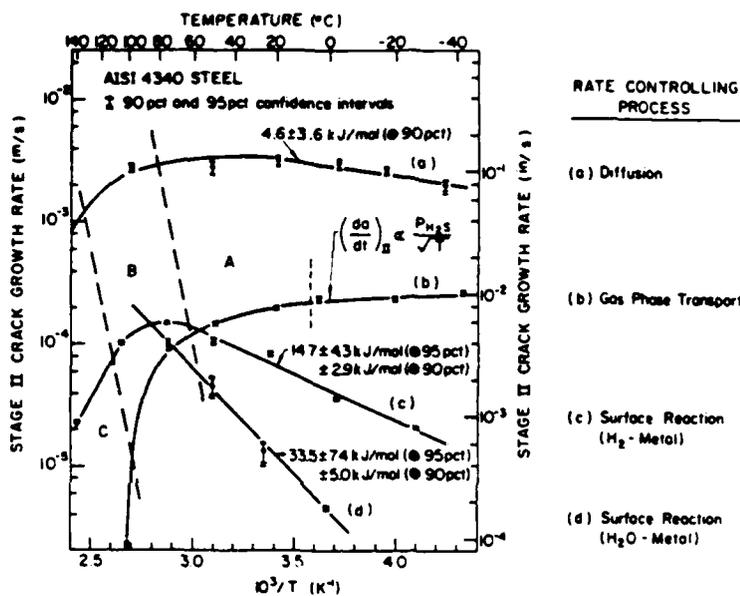
Fig. 6 - Temperature dependence for Stage II crack growth and
the corresponding rate controlling processes at low
temperatures for AISI 4340 steel in various hydro-
genous environments: (a) H_2S at 2.66 kPa, (b) H_2S at
0.13 kPa, (c) H_2 at 133 kPa, and (d) H_2O (liquid)
[51,60,61].
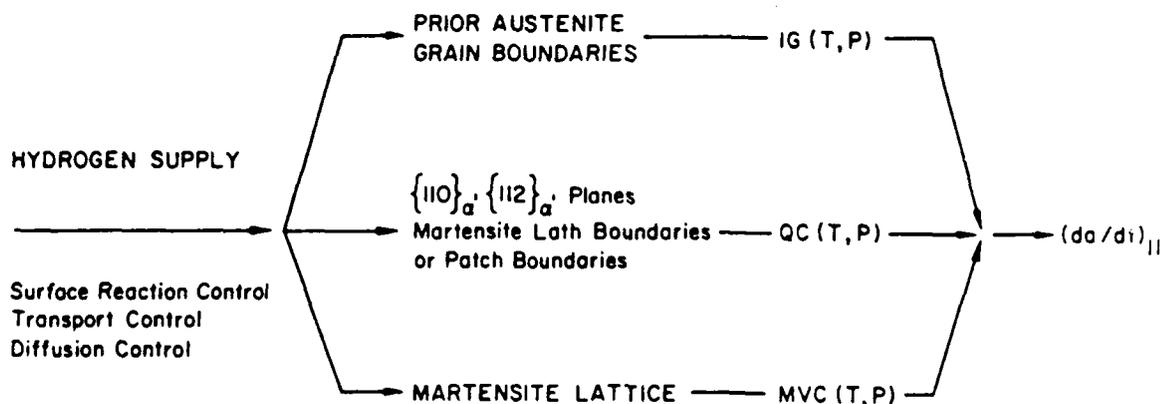
Fig. 7 - Schematic illustration of the partitioning of hydrogen
and its relationship to hydrogen supply and the re-
sulting Stage II crack growth rate [49].
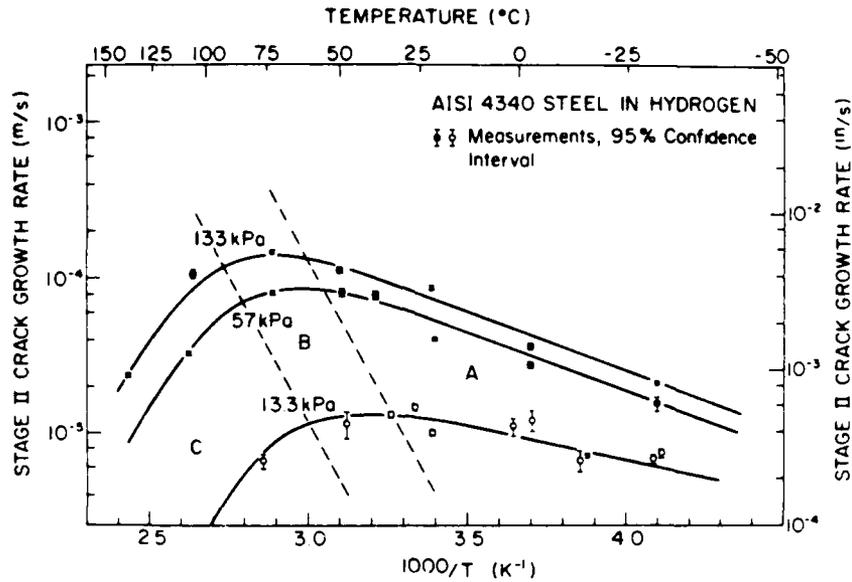
TEMPERATURE (°C)

Fig. 8 - Comparison between model predictions sustained load and crack growth data on AISI 4340 steel tested in hydrogen at different pressures and temperatures [49,61].
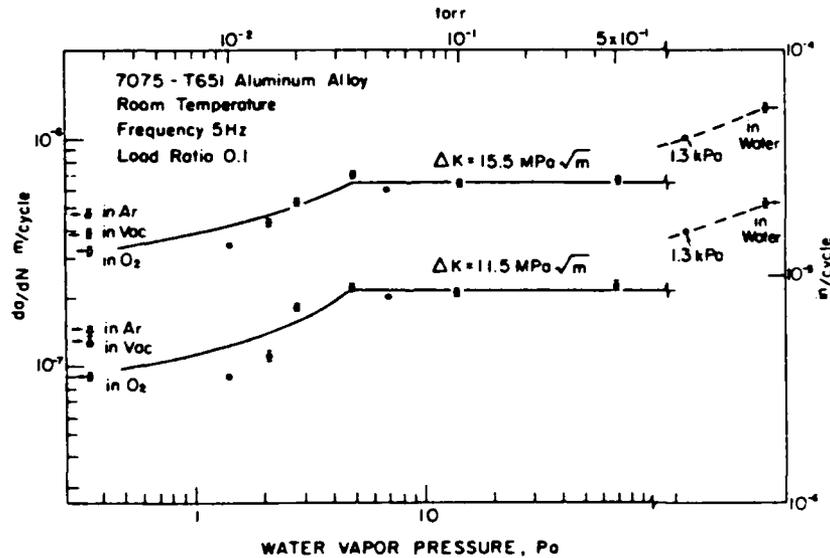
Fig. 9 - The influence of water vapor pressure and of other environments on fatigue crack growth in 7075-T651 (AlMgZn) alloy at room temperature. The solid lines represent predictions of a model for transport-controlled crack growth. The dashed lines indicate surface-reaction-controlled growth and reflect the influence of segregated magnesium [85].

PRESSURE / FREQUENCY (Pa-s)

HYDROGEN SULFIDE PRESSURE (Pa)
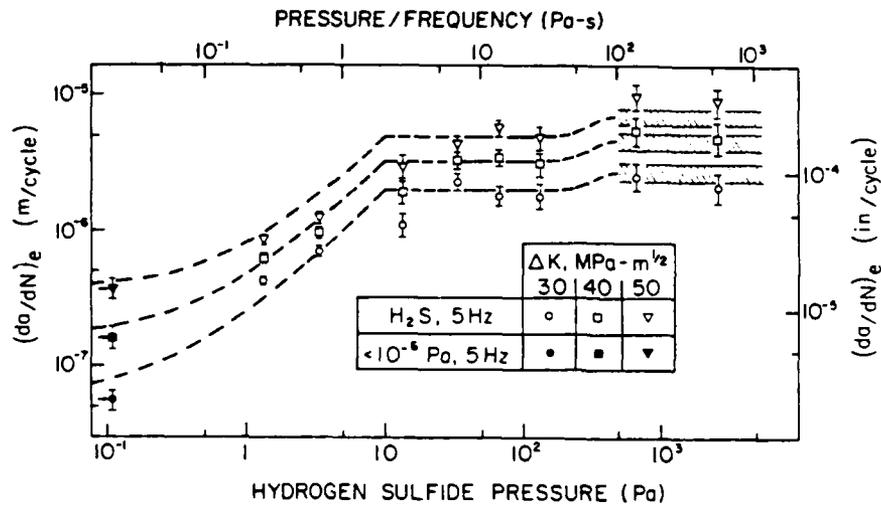
Fig. 10 - The influence of hydrogen sulfide pressure on fatigue crack growth in a 2-1/4Cr-1Mo (A542 Class 2) steel at room temperature. The dashed lines represent predictions of a model for transport-controlled crack growth. The solid lines indicate surface-reaction-controlled growth and reflect the second step of hydrogen sulfide-iron surface reactions.

FREQUENCY f (Hz)

EFFECTIVE TIME 1/f (sec)
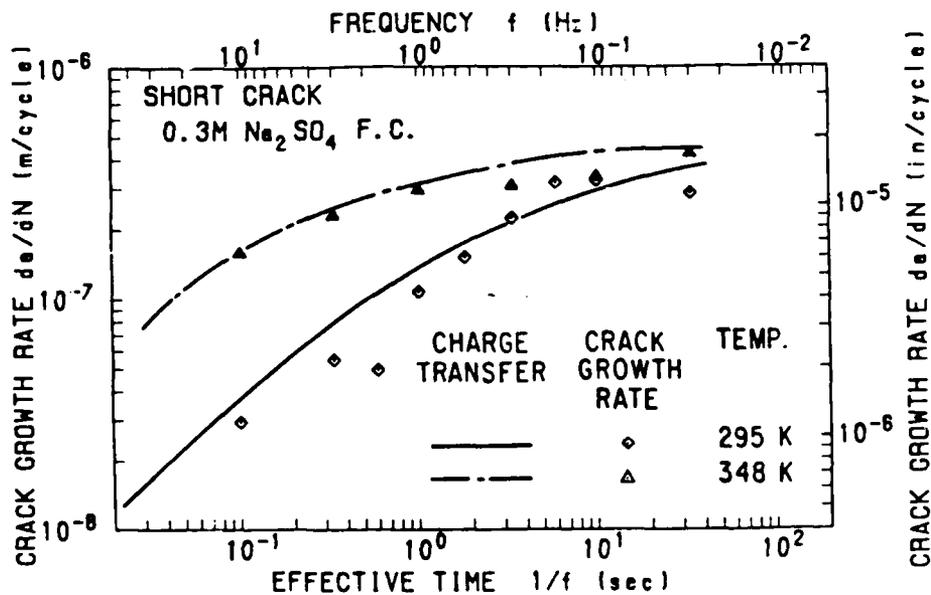
Fig. 11 - The influence of frequency and temperature on fatigue crack growth for NiCrMoV steel in 0.3N Na_2SO_4 solution [80].
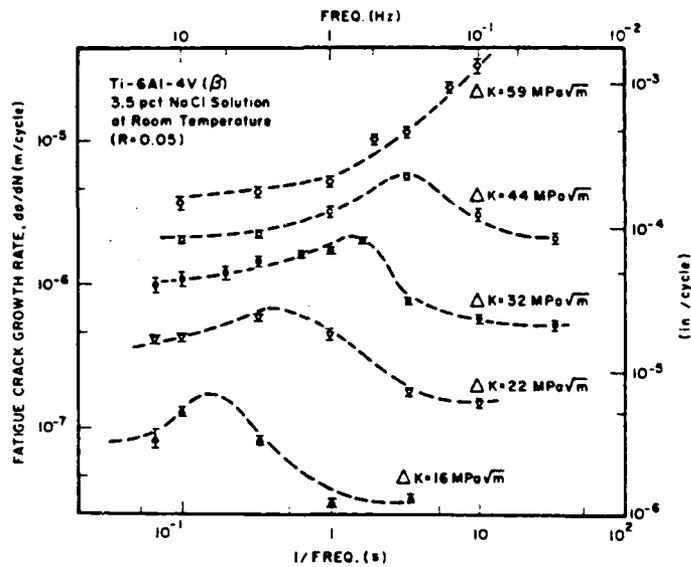
Fig. 12 -   The influence of frequency on fatigue crack growth in
a Ti-6Al-4V alloy exposed to 0.6M NaCl solution at
room temperature and R = 0.1 [83].
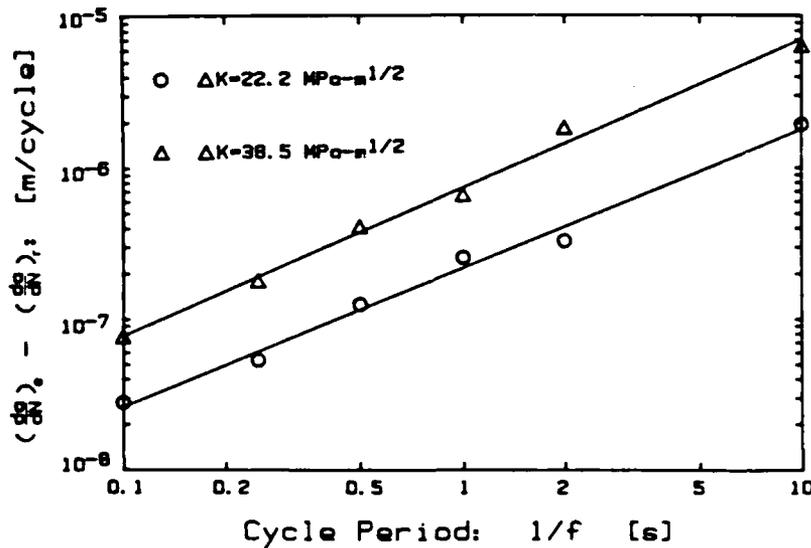
Fig. 13 -   Influence of frequency on fatigue crack growth rate
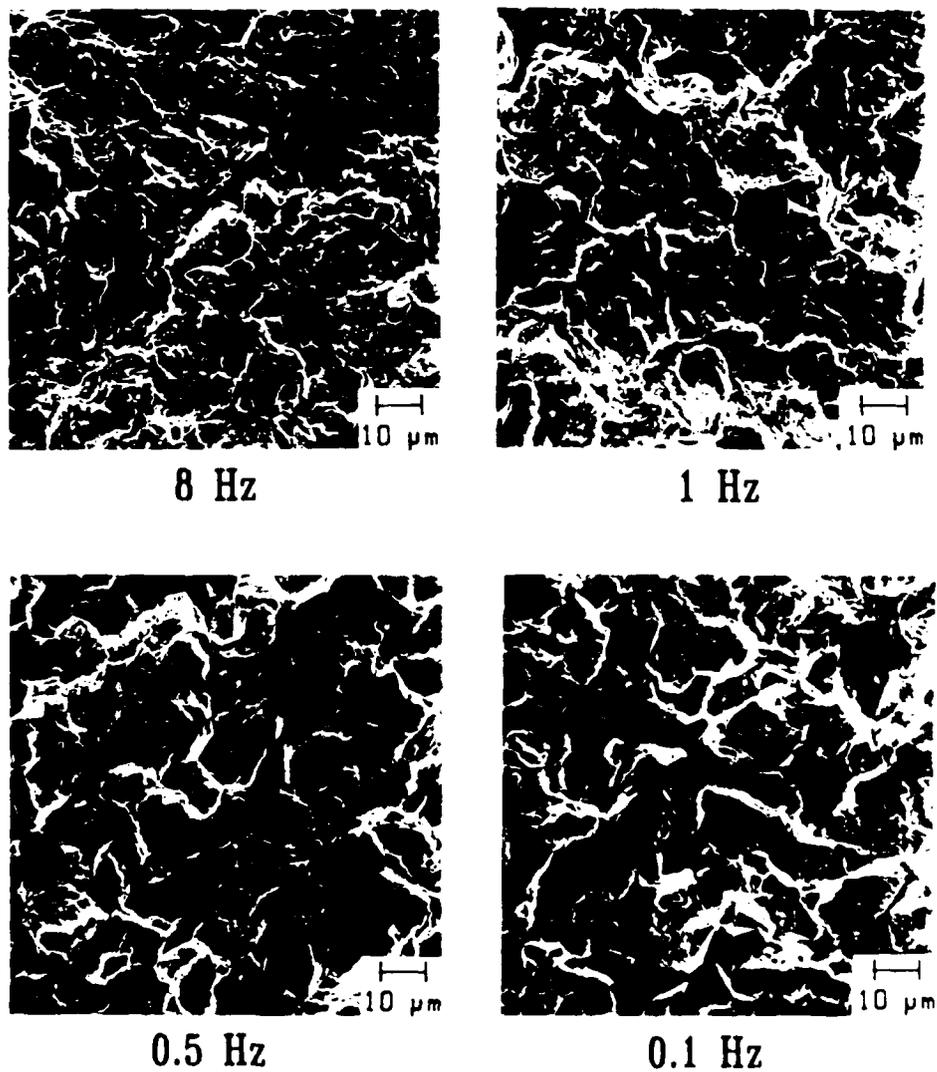for an AISI 4340 steel in water vapor at 585 Pa at
room temperature [84].

8 Hz   1 Hz

0.5 Hz   0.1 Hz

Fig. 14 -   Microfractographs of AISI 4340 steel stressed in water
            vapor, showing changes in fracture surface morphology
            with frequency [84].
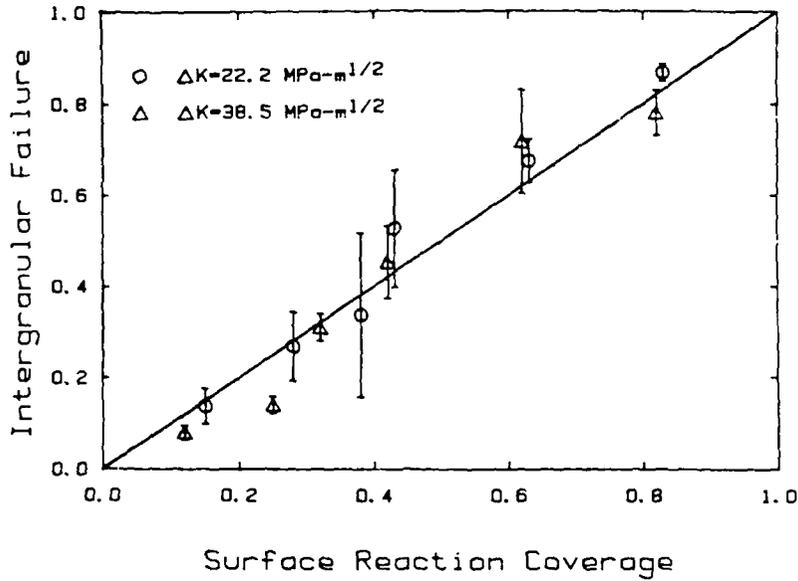
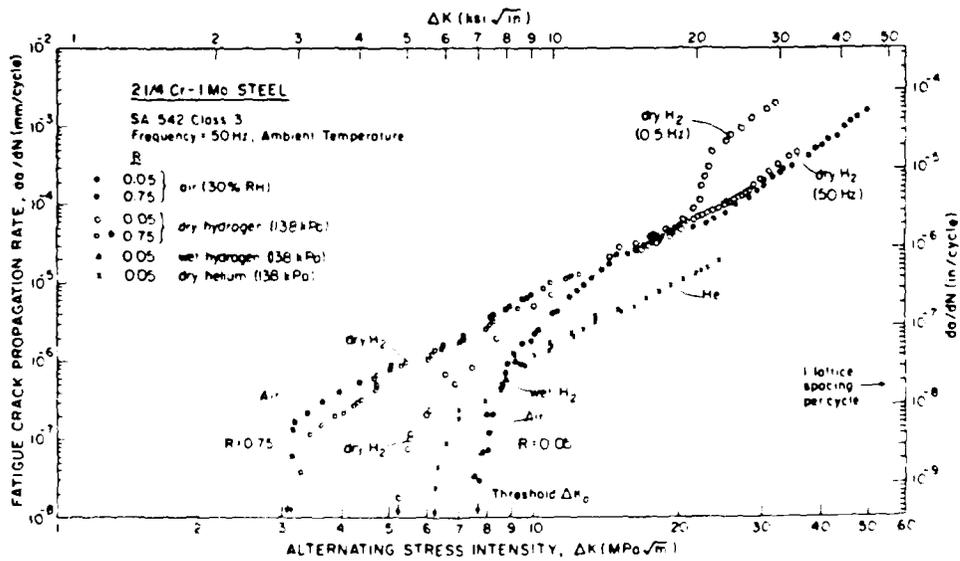Fig. 15 - Correlation between surface coverage and fraction of intergranular failure (or φ) for AISI 4340 steel.

Fig. 16 - Oxide induced closure and hydrogen embrittlement effects on corrosion fatigue crack propagation in a low strength bainitic steel exposed to moist, oxidizing and dry gaseous environments. (after Ritchie et al. [91])
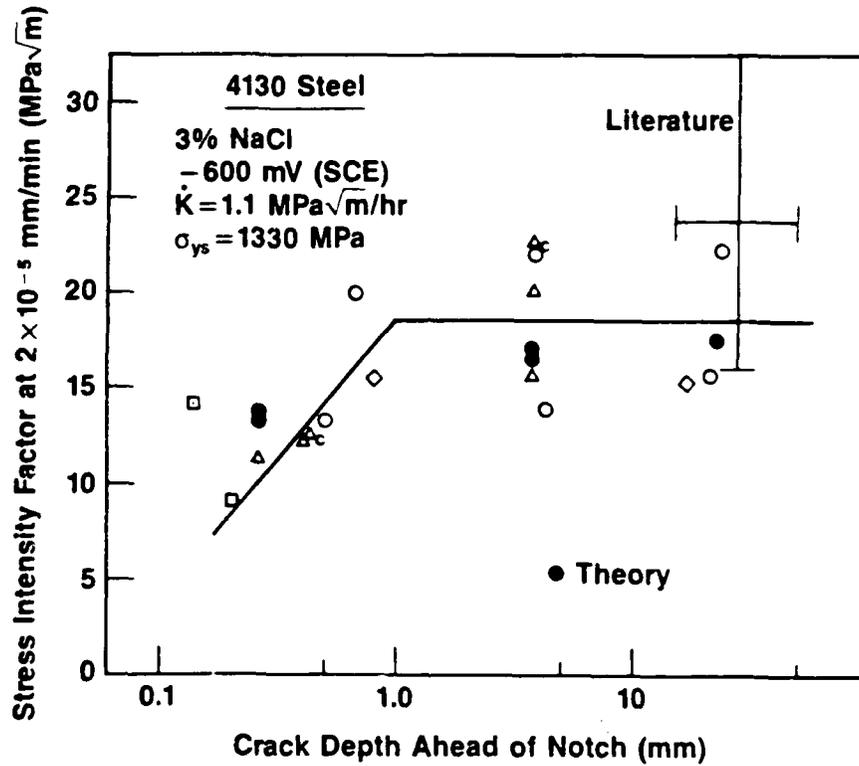
Fig. 17 - The measured and predicted effect of precrack depth on the threshold stress intensity for stress corrosion cracking of high strength steel exposed to 3% NaCl. (after Gangloff and Turnbull [100])
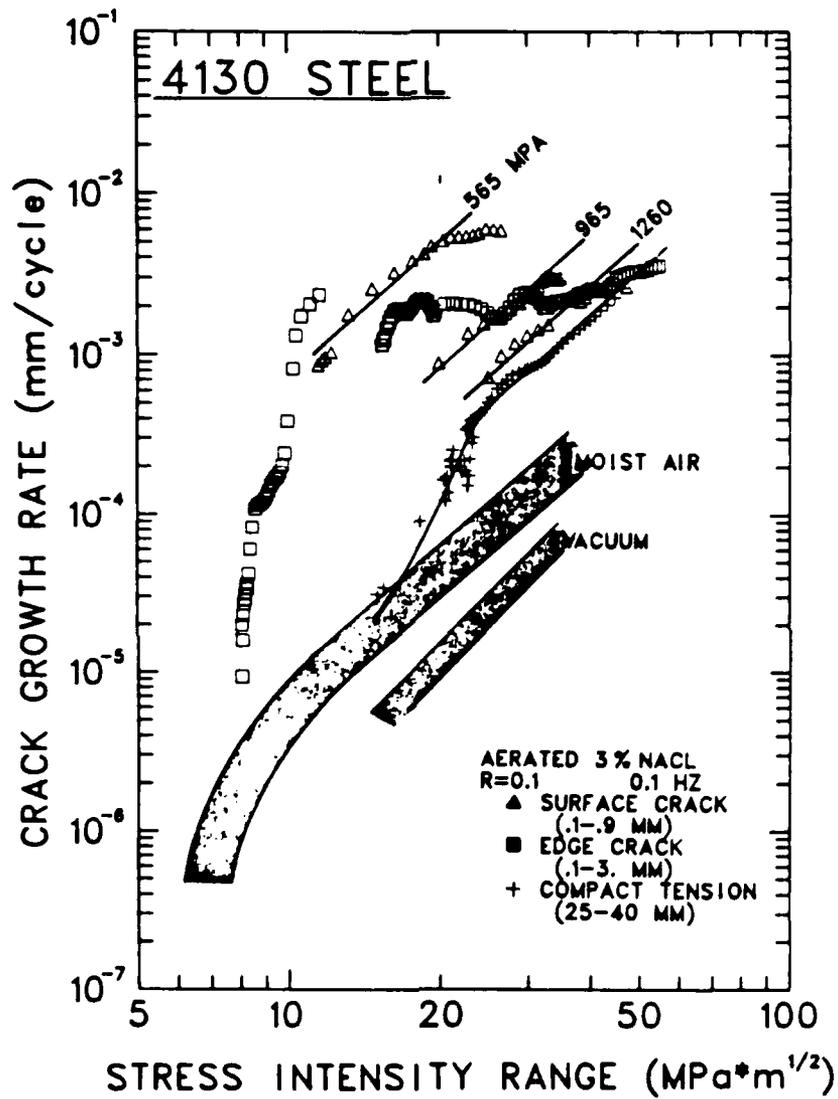
Fig. 18 - The effect of short crack size on corrosion fatigue propagation rates in high strength steel in 3% Nacl. Applied stress ranges are shown for surface cracks. (after Gangloff [104])
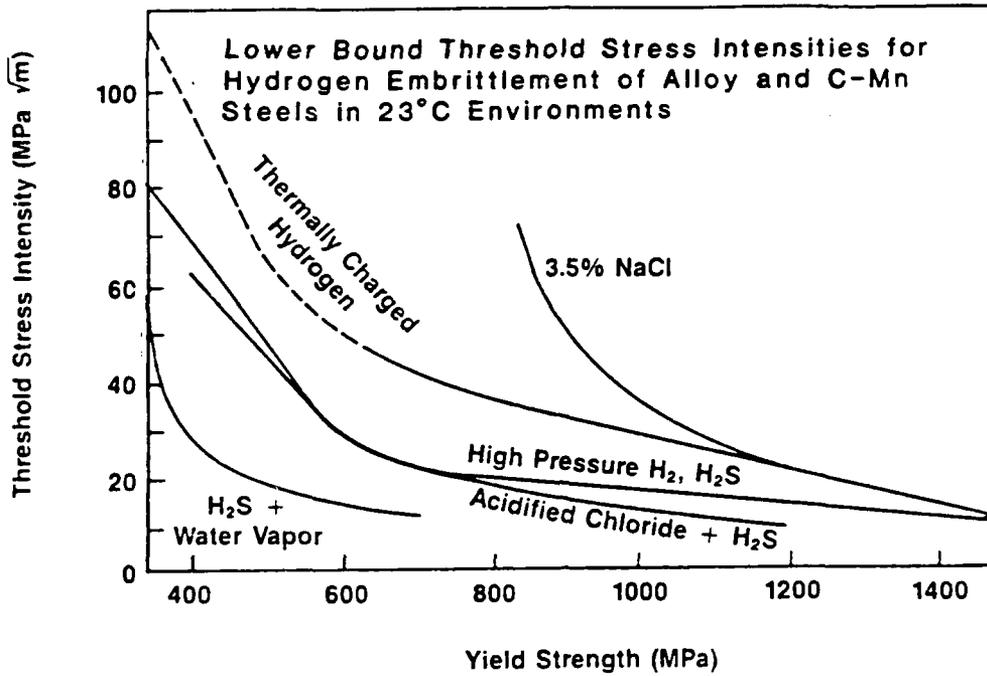
Fig. 19 - Lower bounds on K_{ISCC}-σ_{ys} for ferrite-pearlite, bainitic and tempered martensitic steels stressed in five hydrogen producing environments. Over 500 measurements are represented. (after Gangloff [111])
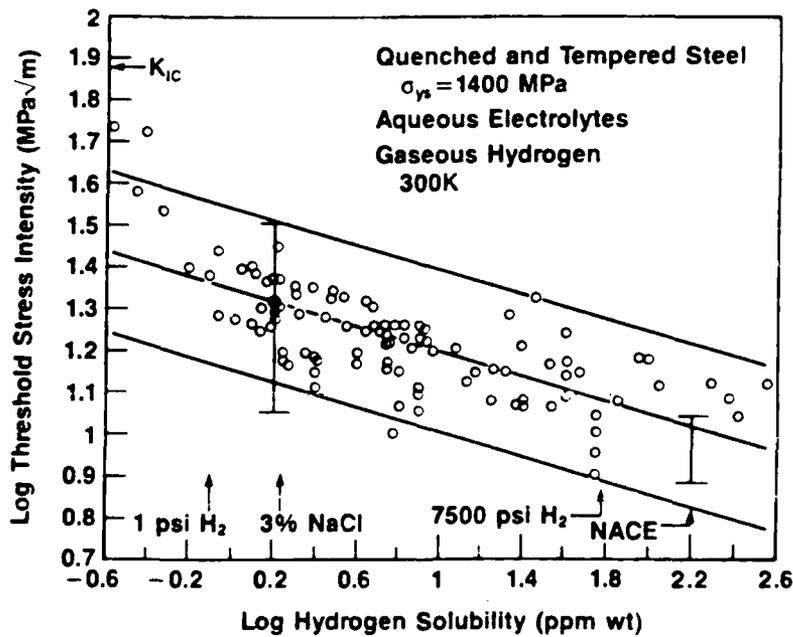

Fig. 20 - Correlation between K_{ISCC} and hydrogen uptake for high strength steels fractured in gaseous hydrogen and electrolytes. (after Gangloff [111])
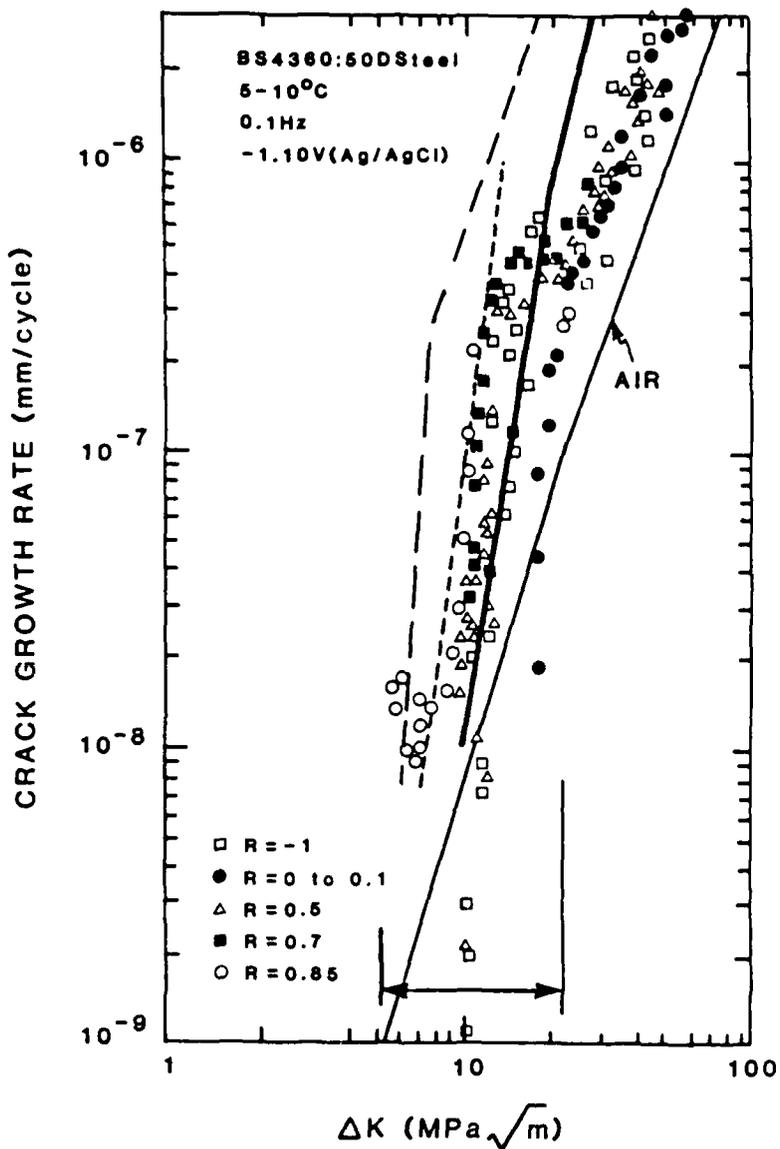
Fig. 21 - Corrosion fatigue crack growth rate versus ΔK for low
strength BS4360:50D C-Mn steel in seawater after Scott
et al. [93]; seawater + H_2S ———— [115], oil + H_2S
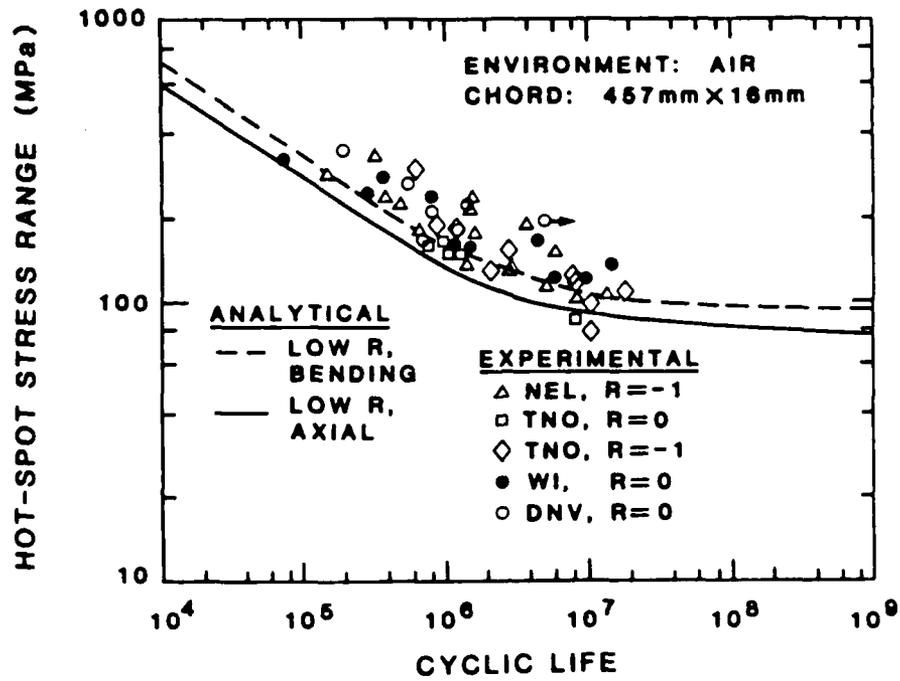———— [117], and high pressure H_2 --------[116].

Fig. 22 -   Fracture mechanics life prediction for fatigue crack
propagation in welded tubular joints exposed to moist
air and compared to full scale component measurements
[121].

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