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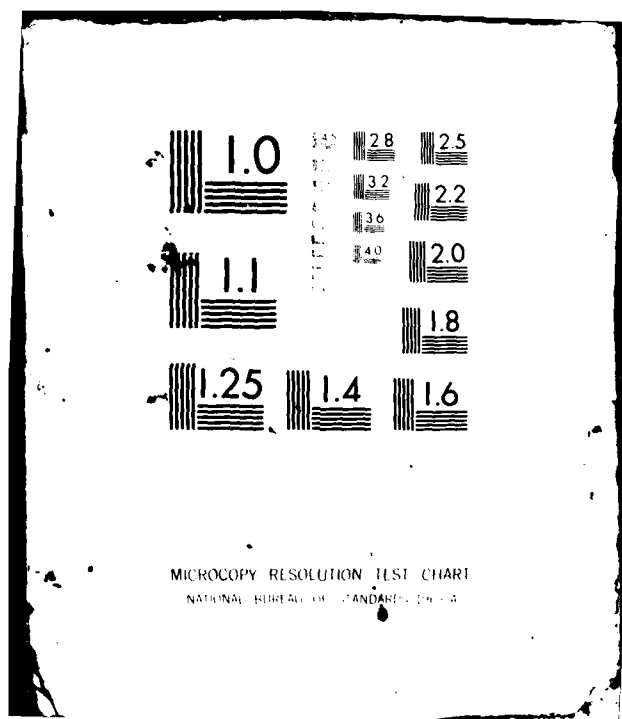
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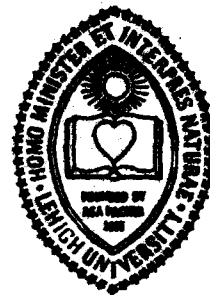
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FRACTURE MECHANICS AND CORROSION FATIGUE

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by

R. P. Wei and Gunchoo Shim

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FRACTURE MECHANICS AND CORROSION FATIGUE

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ABSTRACT

The role of linear fracture mechanics in providing a quantitative framework for corrosion fatigue research and application is reviewed. The importance of integrating chemistry, mechanics and materials science in the development of quantitative mechanistic understanding of corrosion fatigue is emphasized, and is illustrated by results of studies of environmentally assisted fatigue crack growth in gaseous and aqueous environments. Corrosion fatigue of steels in aqueous environments is considered to provide new perspectives for an integrated approach. The need for treating cyclic load frequency as an important variable and for electrochemical measurements at short times (<10s) is discussed.

Key Words: Corrosion fatigue, fatigue crack growth, fracture mechanics, metals, electrochemistry, surface chemistry.

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INTRODUCTION

Corrosion fatigue is a term that is used to describe the phenomenon of cracking (including both initiation and growth) in materials under the combined actions of a fluctuating (or cyclic) stress and a corrosive (deleterious) environment. Its importance in determining the durability and reliability of engineering structures is well recognized. Unfortunately, the term corrosion fatigue conjures up the notion of severe disintegration of the material through chemical attack, accompanied by fatigue cracking. In reality, however, relatively innocuous environments (such as atmospheric moisture) can greatly enhance fatigue cracking without producing visible corrosion in the commonly accepted sense (see [1,2] and references cited therein). To avoid this misconception, the term environmentally assisted fatigue cracking is now preferred, and the use of corrosion fatigue is to be understood within this context. Furthermore, with the development of fracture mechanics technology since the mid-1950's and the increased concern with fatigue crack growth in many applications, considerations of this problem have been subdivided naturally into two groups: initiation and growth. Only the aspects that deal with environmentally assisted fatigue crack growth are considered here.

Just over 10 years ago, a review of the then current state-of-the-art in fracture mechanics technology as it applied to environmentally assisted fatigue crack growth (or corrosion fatigue) was given by A. J. McEvily and R. P. Wei at another international conference on corrosion fatigue [1]. The assumptions and limitations

of this approach, and its engineering utility and usefulness in developing understanding of corrosion fatigue mechanisms and phenomenology were considered and discussed. In the intervening years, fracture mechanics technology has become more firmly established and has contributed significantly to the understanding of environmentally assisted fatigue crack growth. Understanding has come, however, from the recognition that mechanics (more specifically, fracture mechanics) is but one facet of the multi-faceted problem of corrosion fatigue, which involves also chemistry and metallurgy (or, more broadly, materials science).

In this paper, the role of linear fracture mechanics in providing a quantitative framework for corrosion fatigue research and application is reviewed and discussed. The importance of interfacing mechanics, chemistry and metallurgy in developing quantitative understanding of corrosion fatigue, and the relevance of this understanding to engineering are discussed. Environmentally assisted fatigue crack growth in steels exposed to aqueous environments is considered in the context of these discussions.

FRACTURE MECHANICS METHODOLOGY REVISITED

One of the significant developments in the understanding of fatigue crack growth and the utilization of crack growth data in design is associated with the introduction of fracture mechanics technology [1,3-5]. Through linear fracture mechanics, an appropriate crack driving force has been defined as a conjugate to the rate of fatigue crack growth, which is a measure of the material's response. The driving force is defined in terms of the crack-tip stress-intensity factor, K [6,7], or strain energy release

rate, G [6,7], or more generally in terms of the strain energy density factor, S [8,9], for mixed mode loading conditions. The range of these parameters (ΔK , ΔG or ΔS) representing the difference between the maximum and minimum values in one cycle of fatigue loading is more commonly used.

The use of these linear fracture mechanics parameters to characterize the mechanical driving force for crack growth is based on the recognition that crack growth is most likely to proceed from the highly stressed region at the crack tip. It is also predicated on the assumption that linear elasticity analysis results can be applied to an acceptable degree of approximation, and hence imposes the condition of limited plasticity in their use. The assumptions, utility, and restrictions of this approach have been discussed in detail elsewhere [1,3-5]. Specific guidelines have been incorporated now in ASTM Method of Test E647-81 [10].

INTERFACING CHEMISTRY, MECHANICS, AND METALLURGY

With respect to environmentally assisted fatigue crack growth, or corrosion fatigue, fracture mechanics technology contributes in two separable, but yet related ways. Firstly, it provides a formalized framework in which the crack driving force is quantitatively defined and the response to changes in loading, environmental and metallurgical variables can be measured, modeled and systematically examined. Through this formalism, measured crack response can be analyzed and used in estimating service performance. Secondly, because crack growth is the result of deleterious interactions of the environment with the microstructure in the

highly strained (stress) region at the crack tip, some form of fracture mechanics methodology must be incorporated into the quantitative analyses of these interactions.

To further illustrate these two aspects of corrosion fatigue, a schematic diagram and a flow diagram of the various processes that might be involved in environmentally assisted crack growth by hydrogenous gases is shown in Figures 1 and 2 [2]. Hydrogen embrittlement is assumed to be the mechanism for the enhancement of crack growth here. It is inferred that environmentally assisted crack growth is the result of a number of different processes operating in sequence. The rate of crack growth is controlled by the slowest process in this sequence. Modeling the influences of gas phase transport in terms of the effective crack opening and of stress enhanced diffusion in the crack tip region can be made in terms of linear fracture mechanics [11-13], and constitutes the interfacing of the three disciplines at one level. The second level involves a quantitative description of the embrittlement process that can lead to a prediction of the actual growth rates. Because this embrittlement takes place in the highly strained region immediately ahead of the crack tip, where the infinitesimal (small) strain assumption of linear elasticity no longer holds, linear fracture mechanics analysis is not expected to be directly applicable. Additional efforts are needed to link the continuum parameters with the processes (e.g., rupture of the metal-hydrogen-metal bonds) that occur at the microstructural level. Because of these difficulties, the application of linear fracture mechanics to corrosion fatigue is considered only at the first level, i.e.,

only in relation to the coupling between loading and environmental variables.

MODELING OF FATIGUE CRACK GROWTH IN GASEOUS ENVIRONMENTS

Modeling of environmentally assisted fatigue crack growth in pure gases and in binary gas mixtures, where one of the components acts as an inhibitor, has been made and verified [11,12,14,15]. Modeling was based on the proposition that the rate of crack growth in a deleterious environment, $(da/dN)_e$, is composed of the sum of three components [2,11,12].

$$(da/dN)_e = (da/dN)_r + (da/dN)_{cf} + (da/dN)_{scc} \quad (1)$$

$(da/dN)_r$ is the rate of fatigue crack growth in an inert, or reference, environment, and, therefore, represents the contribution of "pure" (mechanical) fatigue. This component is essentially independent of frequency at temperatures where creep is not important. $(da/dN)_{scc}$ is the contribution by sustained-load crack growth (i.e., by "stress corrosion cracking") at K levels above K_{Iscc} , and was first considered by Wei and Landes [16]. $(da/dN)_{cf}$ represents the cycle-dependent contribution which required the synergistic interaction of fatigue and environmental attack, and was considered by Weir et al. [11] and by Wei and Simmons [12].

In the model [11,12], environmental enhancement of fatigue crack growth is assumed to result from embrittlement by hydrogen that is produced by the reactions of hydrogenous gases (e.g., water vapor) with the freshly produced crack surfaces. More specifically, $(da/dN)_{cf}$ is assumed to be proportional to the amount of hydrogen produced by the surface reactions during each cycle,

which is proportional in turn to the "effective" crack area produced by fatigue during the prior loading cycles and to the extent of surface reactions. The time available for reaction is assumed to be equal to one-half of the fatigue cycle (or to $1/2f$, where f is the cyclic load frequency). Based on the assumptions of Knudsen (or molecular) flow and simple first-order reaction kinetics, the following relationships were obtained for transport-controlled and surface-reaction-controlled fatigue crack growth [11,12]:

Transport Control:

$$\begin{aligned} (da/dN)_{cf} &= (da/dN)_{cf,s} (p_o/2f)/(p_o/2f)_s \\ &= (da/dN)_{cf,s} (p_o/p_{o,s}) \quad \text{for } p_o \leq p_{o,s} \quad (2) \end{aligned}$$

Surface Reaction Control:

$$(da/dN)_{cf} = (da/dN)_{cf,s} [1 - \exp(-k_c p_o/2f)] \quad (3)$$

The terms p_o and k_c are the gas pressure in the external environment and the reaction rate constants respectively. $(da/dN)_{cf,s}$ represents the maximum enhancement in the rate of cycle-dependent fatigue crack growth, which recognizes that the extent of surface reaction is limited [11,12]. These models provide a quantitative procedure for assessing the influences of loading and environmental variables, and require the use of $(da/dN)_r$ and $(da/dN)_{cf,s}$ as experimentally measured limits for the material's response.

Two hypothetical cases from [12] are illustrated in Figure 3; one representing transport control and the other surface reaction control. Crack growth response curves are shown in terms of the ratio $(da/dN)_e/(da/dN)_r$ as functions of $p_o/2f$, and are for the case

where $(da/dN)_{e,s}/(da/dN)_r = 50$ and the ratio of reaction rate constants of 10^9 . For more complex reactions, such as those between hydrogen sulfide and steel, the form of the response would differ from the simple cases shown in Figure 3, and would reflect the different steps in the reactions [14]. For binary gas mixtures containing one inhibitor component, the response is modified to reflect the competition between the two gases for surface adsorption sites, and is dependent on the ratio of partial pressures and reaction rate constants for the two gases [12].

APPLICATION TO CORROSION FATIGUE OF STEELS IN AQUEOUS ENVIRONMENTS

It has been recognized that crack growth in steels, exposed to water and water vapor, is controlled by the rate of the water-steel surface reactions [2,17,18]. The reactions with water vapor have been shown to occur in at least two steps [19,20]. The initial, rapid step corresponds to the formation of a patchy $c(2 \times 2)$ layer on Fe(001) single crystal, and the second, slower step corresponds to the nucleation and growth of a two-dimensional FeO layer on Fe(001). It appears now that a further reaction takes place and leads to the formation and growth of Fe_3O_4 or $\gamma-Fe_2O_3$ [21]. Accordingly, the environmentally assisted fatigue crack growth response is expected to reflect these different reaction steps.

Recent work on high-strength steels showed that the concepts developed for the gaseous environments can be extended to fatigue crack growth in aqueous environments [22]. Data on HY130 and modified HY130 steels, tested in water vapor and in distilled water at room temperature, support the linkage of results between the

vapor and liquid phases and the expected correspondence between surface reactions and fatigue crack growth response (see Figure 4). The data in water vapor at low exposures (i.e., $p_0/2f$) correspond to the first step of the reactions with water, while those in distilled water correspond to the slower second step.

Based on these data, it is reasonable to suggest the following modification to eqn. (1) to reflect the existence of at least two steps in the reactions of steel with water:

High Frequencies

$$(da/dN)_e = (da/dN)_r + (da/dN)_{cf,1} \quad (4)$$

Low Frequencies

$$\begin{aligned} (da/dN)_e &= (da/dN)_r + (da/dN)_{cf,1,s} + (da/dN)_{cf,2} + (da/dN)_{scc} \\ &= (da/dN)_r^* + (da/dN)_{cf,2} + (da/dN)_{scc} \end{aligned} \quad (5)$$

At high frequencies, the time available for reaction is sufficiently short such that the first step of the water-steel reaction is incomplete, and that the contribution of sustained-load crack growth is negligible. At sufficiently low frequencies, the first step of the reaction is complete, and the contributions of the second step and from sustained-load growth become significant. It is now convenient to define a new "reference" rate, $(da/dN)_r^*$, which is the sum of the rate in an inert environment, $(da/dN)_r$, and the maximum contribution of the first step reactions, $(da/dN)_{cf,1,s}$.

Furthermore, it is useful to reinterpret eqn. (3), and rewrite it into the following form:

$$\begin{aligned} (da/dN)_{cf,i} &= (da/dN)_{cf,i,s} [1 - \exp(-v_i/2f)] \\ &= (da/dN)_{cf,i,s} [1 - \exp(-1/2f\tau_i)] \end{aligned} \quad (6)$$

Here, first-order reaction kinetics is again assumed, and the quantity $k_c p_o$ in the gaseous case is replaced by a characteristic frequency (v_i) or by the inverse of the reaction time constant (τ_i), where $\tau_i = 1/v_i$. The subscript i denotes parameters associated with the i^{th} step in the surface reactions, and s denotes the maximum or "saturation" value. The quantities v_i and τ_i provide measures of the reaction rate constants in the aqueous environments, and are related to the activation energies for the reactions.

$$v_i = 1/\tau_i \propto \exp(-\Delta H_i/RT) \quad (7)$$

Although the specific form of eqn. (6) must reflect the actual mechanisms of the reactions, it is useful as a first-order approximation for considering corrosion fatigue of high-strength steels at the lower frequencies; i.e., that which corresponds to the slow step of the reactions^{1/}. From considerations of the reactions of water vapor with iron and steel [17,19,20] and from experimental observations of fatigue crack growth response, the following assumptions and observations can be made with respect to crack growth in high-strength steels. $(da/dN)_r$ is observed to be independent of frequency and to be only mildly dependent on temperature (at least in the range of 10^{-8} to 10^{-6} m/c). Because of the limited extent of reactions [19-21], the "saturation" values $(da/dN)_{cf,1,s}$ and $(da/dN)_{cf,2,s}$ are assumed to be independent of temperature and frequency. The temperature and frequency dependent terms, therefore,

^{1/} It is unclear, at this time, that crack growth associated with the first reaction step would be surface reaction controlled. In water vapor, at pressures below that for capillary condensation, this portion is expected to be transport controlled [11,12].

are $(da/dN)_{cf,s}$ and $(da/dN)_{scc}$. For $(da/dN)_{cf,2}$, the temperature dependence is reflected through the temperature dependence of v_2 or τ_2 , and the frequency dependence is given explicitly by eqn. (6). $(da/dN)_{scc}$ is inversely proportional to frequency [16], and is directly related to the temperature dependence for sustained-load crack growth. Because the same reactions control both sustained-load and fatigue crack growth, the same activation energy will be associated with v_2 or $1/\tau_2$ (or simply v or $1/\tau$) and $(da/dN)_{scc}$. Clearly, elucidation of the connection between chemical reaction kinetics and corrosion fatigue cannot be obtained from examining the temperature dependence alone, and must now include cyclic load frequency as a significant parameter for fatigue crack growth.

To illustrate this point and the overall approach, fatigue crack growth data, similar to those shown in Figure 4, were obtained on a HY130 and a modified HY130 steel as a function of frequency at different temperatures in distilled water and in a buffered acetate solution (pH = 4.2). The data were analyzed in accordance with eqns. (5) and (6), and are shown in Figures 5 and 6 in terms of the difference, $(da/dN)_e - (da/dN)_r^*$, or of the environmental contribution $(da/dN)_{cf,2} + (da/dN)_{scc}$, versus the inverse of cyclic load frequency ($1/2f$). It is seen that the results are in good agreement with eqn. (6). The fact that environmental effects were beginning to be observed at higher frequencies at the higher temperatures is consistent with the increased rates of reactions with temperature. The characteristic frequency, v , or apparent reaction time constant, τ , at each temperature was determined from the fatigue data, and are shown in an Arrhenius plot in Figure 7. Based on a preliminary analysis, the apparent activation energy was found

to be equal to 50 ± 6 kJ/mol for distilled water and 39 ± 15 kJ/mol for the buffered acetate solution at the 95 pct confidence level. It is to be recognized that the values of v or τ are sensitive to the choice of the value of $(da/dN)_{cf,s}$. Hence, these values and the associated activation energy may change somewhat with additional data and with refinements in analysis. Nevertheless, these values of apparent activation energy are quite consistent with a value of 36 ± 28 kJ/mol (at the 95 pct confidence level) for the reaction of water vapor with AISI 4340 steel [17]. The results therefore tend to support the concept of surface reaction controlled crack growth.

DISCUSSION

The influence of frequency on fatigue crack growth in high-strength steels exposed to aqueous environments has been examined previously by several other investigators [18,23,24]. Data from these investigations are shown in conjunction with the data at room temperature for the HY130 steels in Figure 8 [22]. It is apparent that these data generally conform with eqn. (6), with apparent reaction time constants τ that differ for the different combinations of environments and steels.

Although the fatigue crack growth data strongly support the concept of surface reaction controlled crack growth for steels in aqueous environments, definitive support must await the development of supporting chemical data similar to those for the gaseous environments [17,25-27]. It is clear from the fatigue data, the relevant reactions would be those that occur during the very early stages of the reaction of the environment with the clean steel surfaces.

The time frame for these reactions is expected to be on the order of milliseconds to tens of seconds, and is extremely short as compared with that for traditional corrosion measurements. Chemical or electrochemical techniques, therefore, must be developed and used for making measurements at these short time intervals. These measurements must be made in environments that are representative of the solution chemistry at the crack tip. Analysis techniques must be developed also to provide unambiguous resolution of the various reaction steps from the experimental data. These efforts are in progress.

According to eqns. (5) and (6), the deviation in data from the "saturation" or plateau level at the lower frequencies (see Figures 5 and 6) should be attributed to the contribution by sustained-load crack growth, i.e., by the $(da/dN)_{scc}$ component. Unfortunately, however, the corresponding sustained-load crack growth rates that would be required to account for these contributions are too high by at least one order of magnitude. This discrepancy may be attributed to transient crack growth, or more likely, to the contributions from an additional step in the water-steel reactions. Further work is needed to clarify this issue.

SUMMARY

The role of linear fracture mechanics in the understanding of environmentally assisted fatigue crack growth (corrosion fatigue) and in the development and utilization of data for design are reconsidered. It provides the essential quantitative framework for corrosion fatigue research and application. It must be recognized, however, that linear fracture mechanics (or, more broadly,

mechanics) is but one facet of the multi-faceted problem of corrosion fatigue, which involves also chemistry and metallurgy (or materials science). Quantitative mechanistic understanding of corrosion fatigue can be expected only from investigations that integrate all of these disciplines.

As an illustration, modeling of fatigue crack growth in gaseous environments is reviewed. Its extension to crack growth in aqueous environments is considered. For high-strength steels, fatigue crack growth in aqueous environments appears to be controlled by the rate of reactions of the environment with the newly created crack surfaces, and the crack growth response tends to reflect the different steps in these reactions. The relevant reactions appear to be those that occur in 10 seconds or less. To better understand corrosion fatigue behavior, therefore, it is essential to recognize cyclic load frequency as a significant variable and to examine the frequency dependence for fatigue crack growth as a function of temperature. These data must be correlated to and supported by measurements of chemical reaction kinetics at very short times (<10 s) in environments that properly reflect the conditions at the crack tip.

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FIGURE CAPTIONS

- Figure 1: Schematic illustration of various sequential processes involved in environmentally assisted crack growth in alloys exposed to external gaseous environments. (Embrittlement by hydrogen is assumed and is schematically depicted by the iron-hydrogen-iron bond.) (After [2]).
- Figure 2: Flow chart illustrating the processes and parameters that affect environmentally assisted crack growth.
- Figure 3: Schematic illustration and comparison of gas transport and surface reaction controlled fatigue crack growth [12].
- Figure 4: The influence of frequency and exposure ($p_0/2f$) on fatigue crack growth for a modified HY130 steel in water vapor and in distilled water, at room temperature.
- Figure 5: The influence of frequency and temperature on fatigue crack growth for HY130 (solid symbols) and modified HY130 (open symbols) steels in distilled water.
- Figure 6: The influence of frequency and temperature on fatigue crack growth for HY130 (solid symbols) and modified HY130 (open symbols) steels in buffered acetate solution (pH = 4.2).
- Figure 7: The effect of temperature on the characteristic frequency (ν) for fatigue crack growth in aqueous environments.
- Figure 8: Room temperature fatigue crack growth response for high-strength steels in water vapor and in aqueous environments [18,22-24].

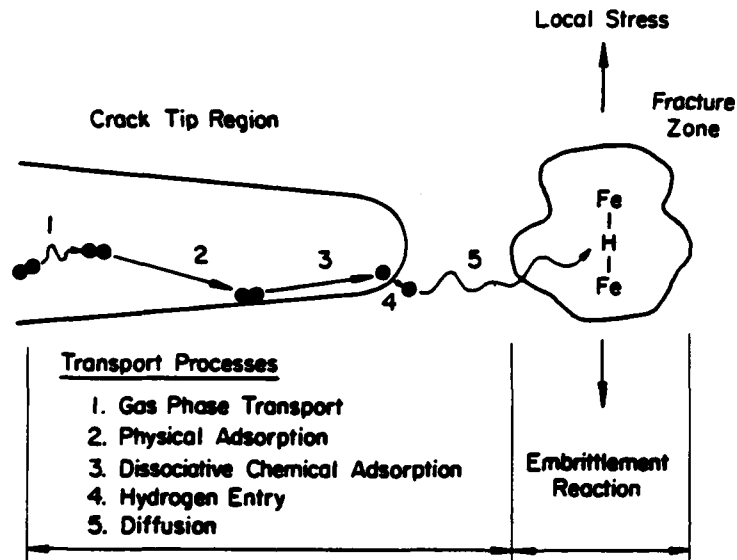


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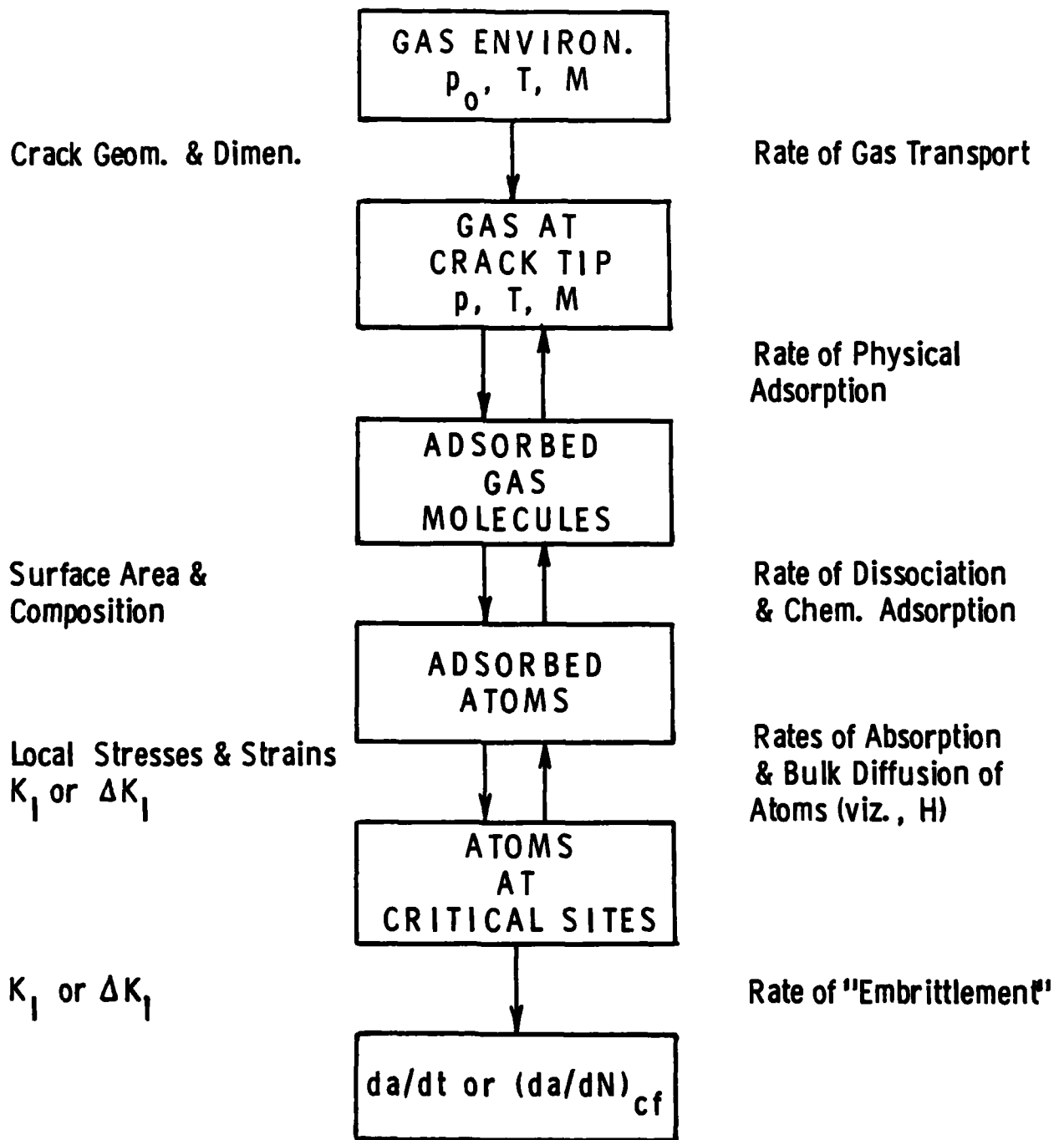


Figure 2: Flow chart illustrating the processes and parameters that affect environmentally assisted crack growth.

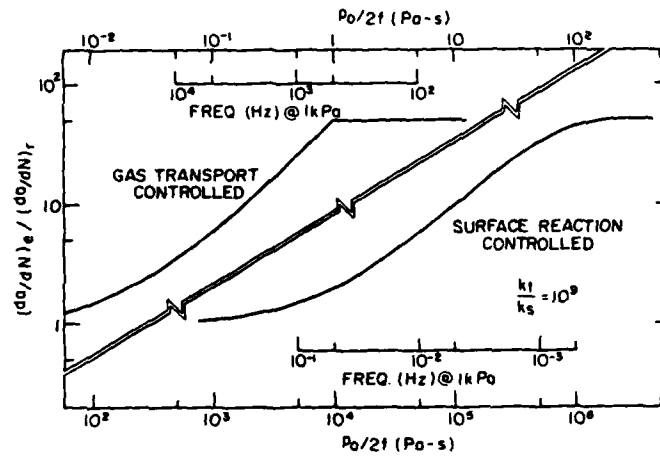


Figure 3: Schematic illustration and comparison of gas transport and surface reaction controlled fatigue crack growth [12].

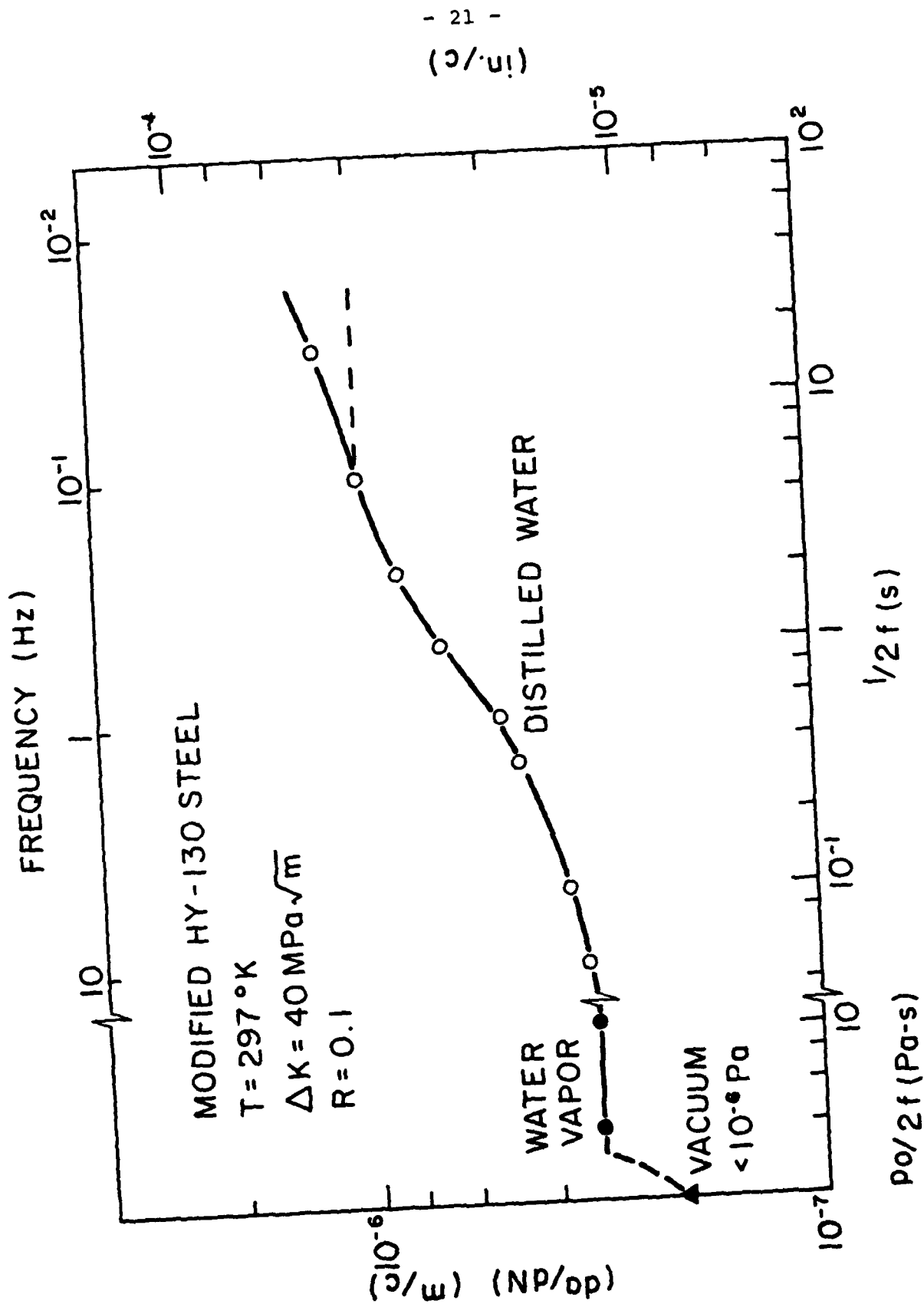


Figure 4: The influence of frequency and exposure ($P_o/2f$) on fatigue crack growth for a modified HY130 steel in water vapor and in distilled water, at room temperature.

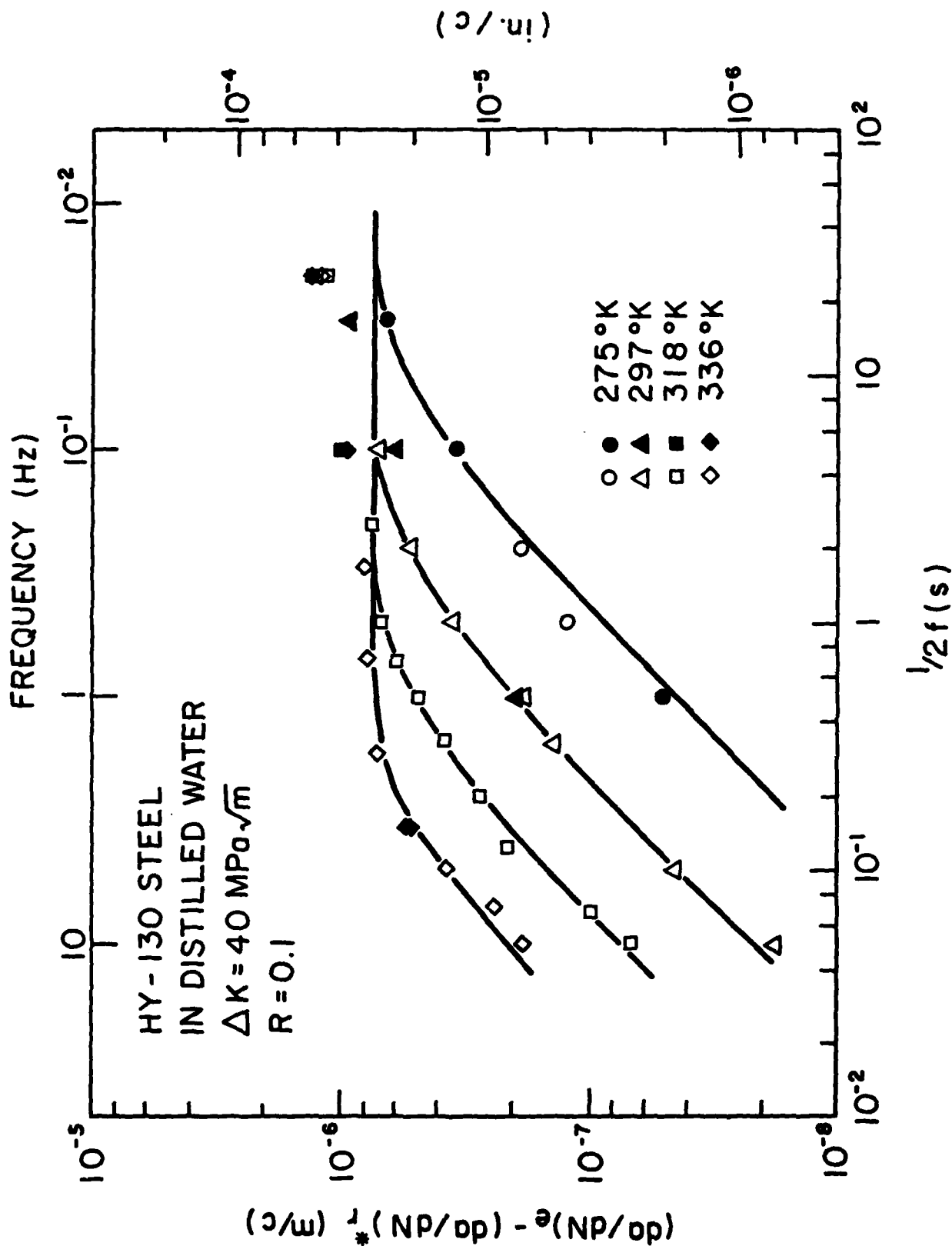


Figure 5: The influence of frequency and temperature on fatigue crack growth for HY130 (solid symbols) and modified HY130 (open symbols) steels

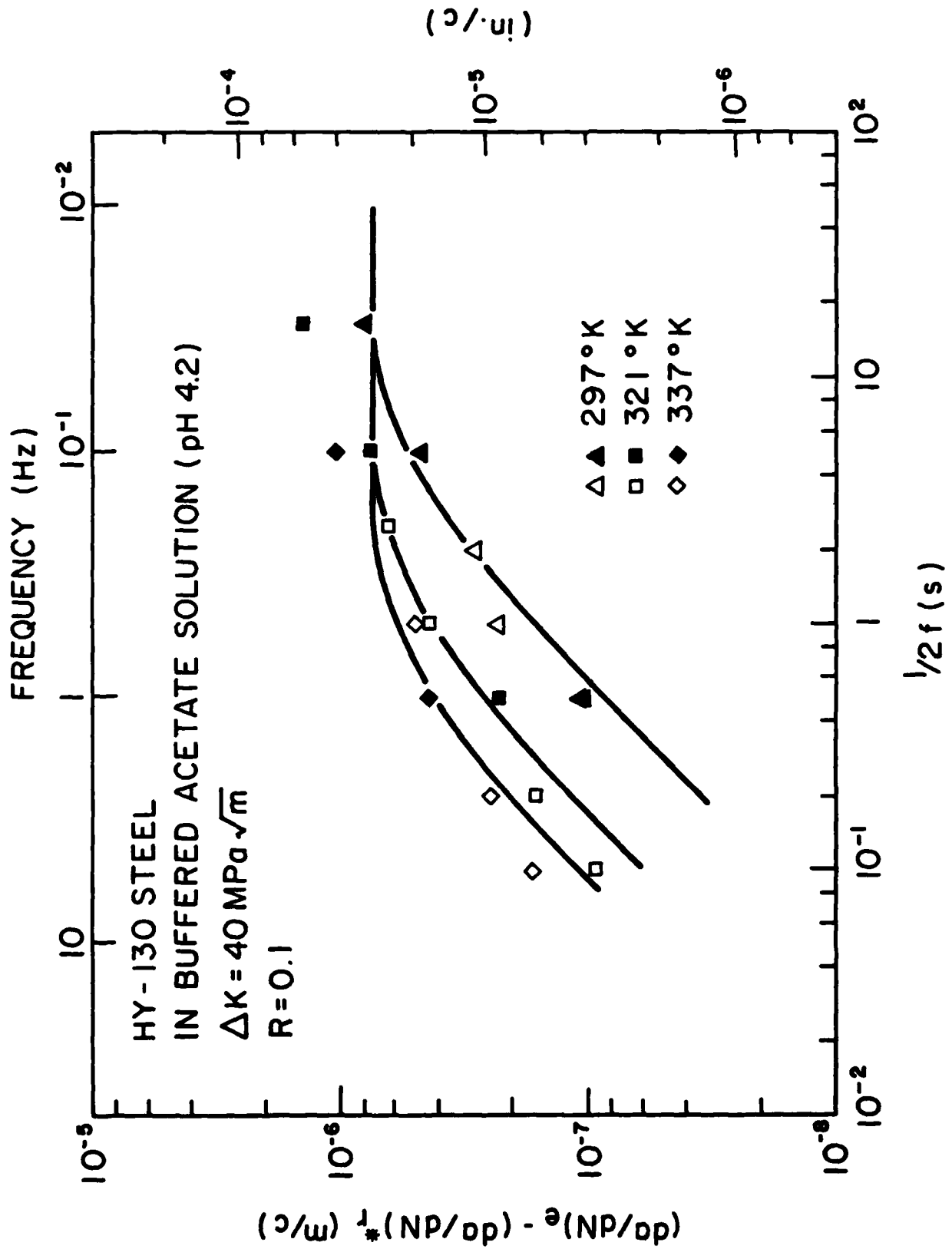


Figure 6: The influence of frequency and temperature on fatigue crack growth for HY130 (solid symbols) and modified HY130 (open symbols) steels in buffered acetate solution (pH = 4.2).

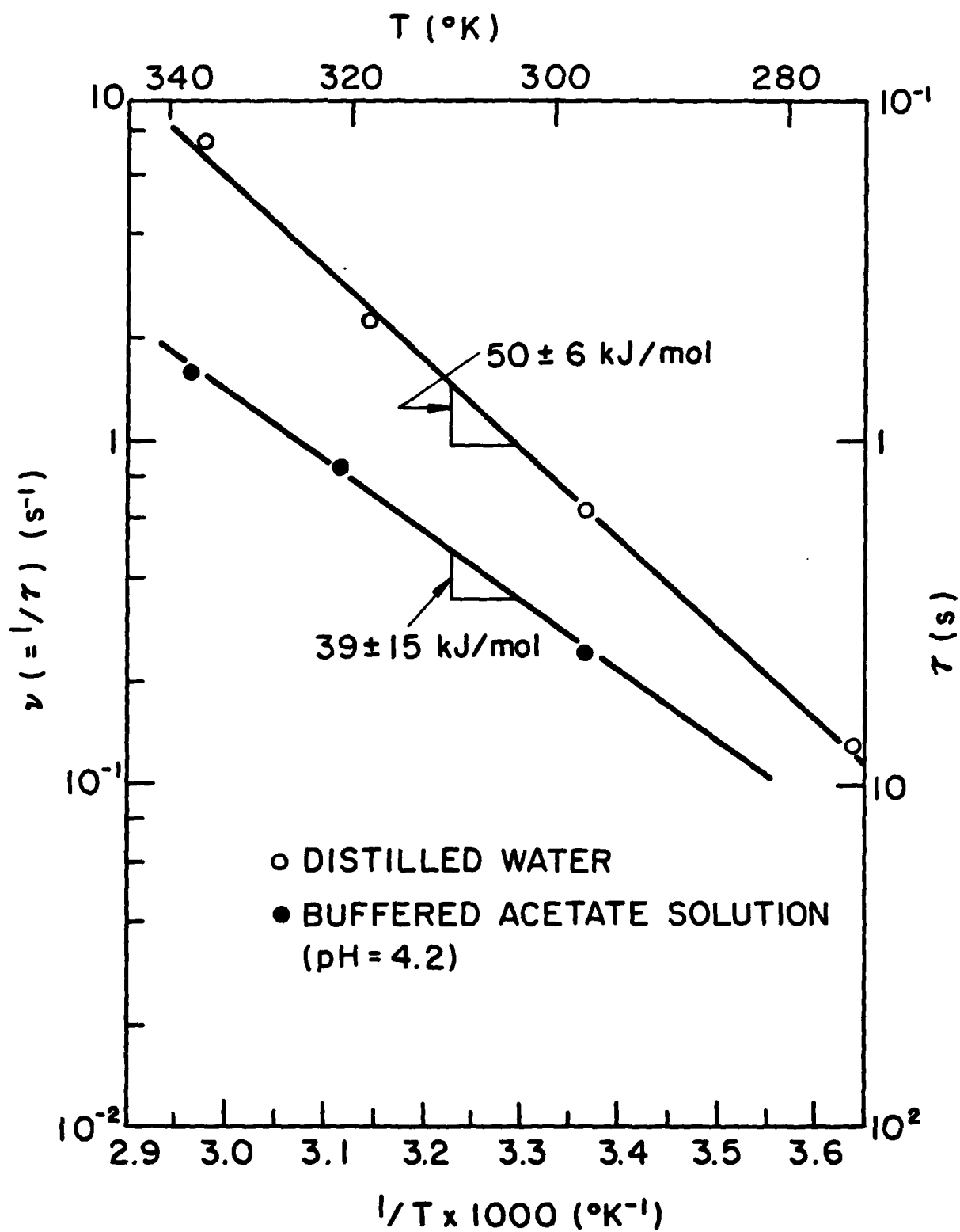


Figure 7: The effect of temperature on the characteristic frequency (ν) for fatigue crack growth in aqueous environments.

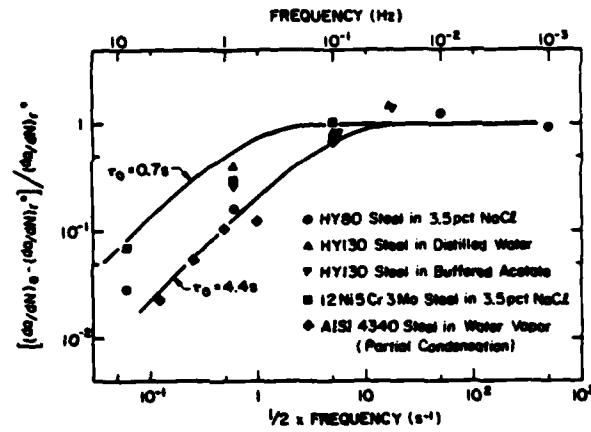


Figure 8: Room temperature fatigue crack growth response for high-strength steels in water vapor and in aqueous environments [18,22-24].

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