Effect of Crack Closure in Nb–H Alloys

C. C. LIN and K. SALAMA
Department of Mechanical Engineering, University of Houston,
University Park, Houston, Texas 77004, USA

ABSTRACT

The effect of crack closure on near-threshold fatigue crack propagation rate has been investigated in niobium-hydrogen alloys. The study was undertaken with the ultimate goal of determining the role of hydrogen in conjunction with effects of crack closure on the fatigue and embrittlement processes of niobium as a representative of group VA metals. Fatigue tests were performed at room temperature on a hydrogen-free specimen as well as specimens containing hydrogen in solid solution and in the form of hydride. Compact specimens were tested using a tension-tension loading cycle at a frequency of 4 Hz and load ratios of 0.05, 0.4 and 0.75.

KEYWORDS

Fatigue crack propagation, Threshold, Crack closure, Hydrogen embrittlement, Niobium.

INTRODUCTION

Numerous investigations in the literature have concentrated on the effect of hydrogen on monotonic mechanical behavior of group VA refractory metals : vanadium, niobium and tantalum (Farahani et al., 1981, Ghar et al., 1977). Nevertheless, little attention has been directed to the influence of hydrogen on the fatigue crack propagation behavior in this group of metals. Among the early investigators in this field, Beevers (1970) found that the restriction of twin formation by the hydride precipitates can increase the life of α-titanium. Wilcox (1964), however, reported that hydrogen in solid solid solution and hydride forms significantly reduce the fatigue life of tantalum.

In contrast, Lee et al., (1976) reported that vanadium containing hydrogen in the form of hydride significantly improved fatigue life in the high cycle tension-compression operation. Chung et al., (1978) found that vanadium alloys containing hydride are more resistant to crack initiation than alloys with hydrogen in solid solution. Owen et al., (1987) suggested that hydrogen in solution is the predominant cause for both the strengthening and embrittlement observed in V and V-Cr alloys. Hydrides, however, either existing or stress induced, appear to make little or no contribution.

Fariabi et al., (1983) investigated the effects of hydrogen on the near-threshold fatigue crack growth rate of niobium at room temperature. The results indicate that the threshold stress intensity range, ΔKth, decreases with the addition of hydrogen and reaches a minimum, (ΔKth)min, at a hydrogen concentration approximately equal to the solubility limit of hydrogen in niobium. As the hydrogen concentration exceeds the solubility limit, ΔKth increases with the increase of the amount of hydrogen dissolved in the specimen. From these results, it was suggested that dislocation-hydrogen interaction plays an important role in the embrittlement process. Polvanich et al., (1987) have investigated the effect of temperature on near-threshold fatigue crack growth rate in the niobium-hydrogen alloys at 273 and 400 K. At both temperatures, the results show that the behavior of ΔKth as a function of hydrogen concentration is similar to that obtained by Fariabi et al., (1983) at room temperature. It was also found that (ΔKth)min increases linearly with the increase in temperature.

The effect of crack closure of fatigue crack growth has been studied by Halliday et al., (1981). They showed that the difference in crack growth rates can be substantially accounted for by an effective stress intensity factor range parameter in Ti alloys. Gray et al., (1984) observed that the influences of grain size and mean stress on fatigue crack growth in Ti alloys are attributed to mechanisms of roughness-induced closure and the degree of crack path deviation. Shih et al., (1974) showed that crack closure occurs during fatigue for the Ti alloys. However, crack closure cannot be regarded as the sole cause for the various observed phenomena for fatigue. It cannot completely account for the influence of stress ratio on fatigue crack growth; neither can it fully explain the delay phenomenon under variable-amplitude loading.

Pendse et al., (1985) investigated the role of hydrogen attack in influencing fatigue crack growth in pressure vessel steels. They found that the microstructural damage from grain boundary void formation following prolonged exposure to high temperature, high pressure hydrogen environment, which causes severe degradations in strength, ductility and toughness properties, has only a marginal effect on the propagation of fatigue crack at near-threshold levels since any acceleration in growth rates due to hydrogen attack damage is offset by a local reduction in crack driving force from the concomitant increase in crack closure.

Todd et al., (1988) recently proposed a new mechanism of crack closure resulting from hydrogen embrittlement. As they investigated the cathodically protected ASTM A710 steel tested in seawater, they observed that the increased threshold stress intensity range and slow crack growth rates could not be attributed to calcareous deposit formation, but appeared to be a function of hydrogen embrittlement resulting in metal wedges developing in the crack wake and contributing to crack closure.

EXPERIMENTAL PROCEDURES

The specimens used in this investigation were made of pure niobium (99.85 %) furnished by Teledyne Wah Chang Corporation in the form of a sheet 1 mm in thickness. Compact tension specimens were machined out of this sheet in thickness. The dimensions chosen such that a plane stress condition was satisfied. The specimen preparation as well as the method used to introduce hydrogen are described in detail elsewhere (Fariabi et al., 1983).

Fatigue tests were conducted in a tension-tension loading with a closed-loop electro-servohydraulic material testing machine in a haver-sinusoidal load cycle operating at a frequency of 4 Hz. The tests were performed at load ratios of 0.05, 0.4 and 0.75. The stress intensity range, ΔK, was determined using the expression

$$\Delta K = \frac{\Delta P}{BW^{1/2}} f(a/w)$$

where ΔP is the load range, B and W are the thickness and the width of the specimen, respectively, and f(a/w) is the compliance function, where a is the crack length.

The change in crack length was determined by measuring changes in the electrical potential at two points across the crack where a constant alternate current was applied. The system used in measuring the crack extension as well as the method used in measurements are described in detail elsewhere (Polvanich et al., 1987). The tests usually were terminated when the crack length was about 60 % of the specimen width.

Measurements of crack closure to determine crack opening load on near-threshold stress intensity level were made by means of electrical potential and clip-on gage technique. The applied load versus the output (electrical potential and displacement) curve were recorded on a x-y recorder at a frequency of 0.04 Hz. Closure load was determined from the point where the curve deviated from linearity during the unloading cycle. To detect the slight variations of linearity in load-output curves, a subtraction circuit system was used to facilitate the specifying of the closure load (Kikukawa et al.,1976).

RESULTS

The fatigue tests were performed at 296 K on hydrogen-free as well as hydrogenated niobium specimens. All specimens were fatigued using a tension-tension loading cycle at a frequency of 4 Hz and load ratios of 0.05, 0.4 and 0.75. Figures 1(a) through 1(c) show the crack growth rate, da/dN, as a function of stress intensity range, ΔK, for all the hydrogen concentrations used in this study. The majority of the curves in the figures represent the crack growth rate in Regions I and II. In some specimens, however, the tests were extended to cover Region III or part of it. From these figures, it can been seen that at the low crack growth rate (near-threshold), there is a strong dependence of ΔKth on the concentration of hydrogen in the specimen. This region is then followed by Region II where da/dN gradually increases with ΔK.

The threshold stress intensity range, ΔKth, as a function of hydrogen concentration is plotted in Fig. 2. From Fig. 2, one can see that at R=0.75, ΔKth of hydrogen-free niobium exhibits the highest value and this value decreases continuously as the hydrogen concentration is further increased. The results at R=0.05 and 0.4 show that ΔKth of hydrogen-free niobium is still the highest and decreases as hydrogen is introduced but

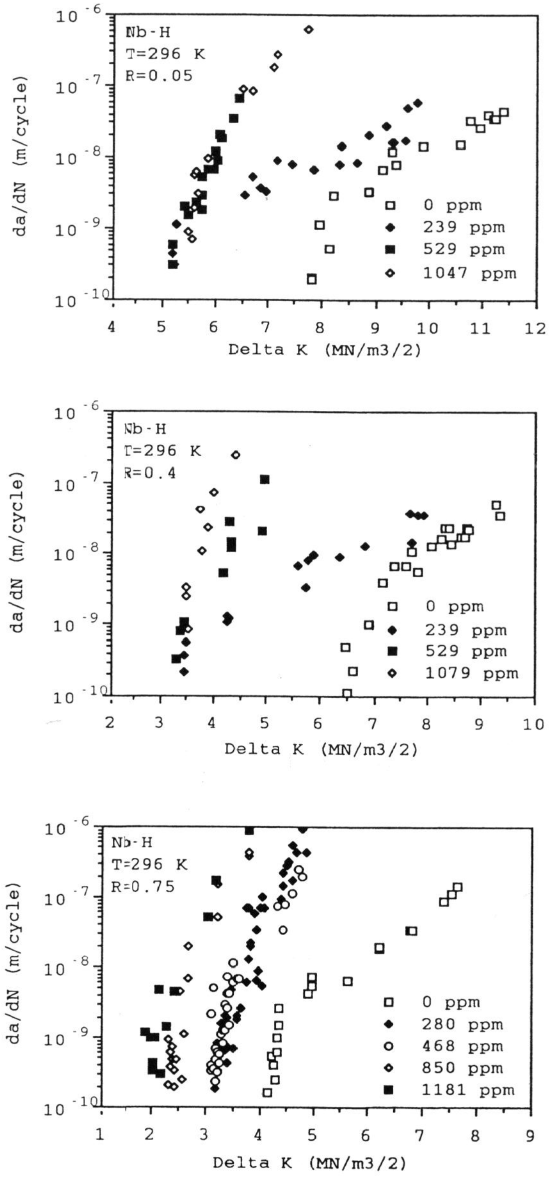

Fig. 1. Fatigue crack propagation rate, da/dN, as a function of stress intensity range, ΔK for different hydrogen concentrations in Nb-H alloys at (a) R=0.05, (b) R=0.4 and (c) R=0.75.

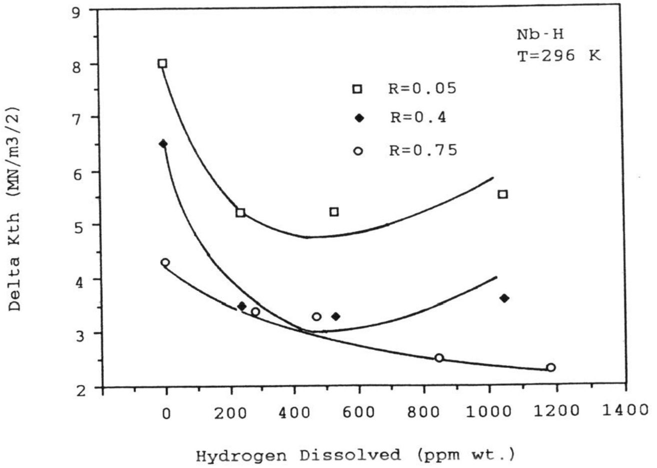

Fig. 2. Dependence of threshold stress intensity range, ΔK_{th}, on hydrogen concentration in Nb-H alloys at R=0.05, 0.4 and 0.75.

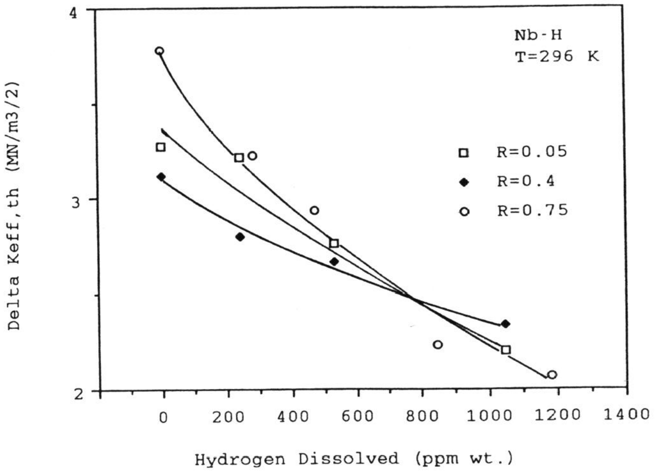

Fig. 3. Effective threshold stress intensity range, $\Delta K_{eff,th}$, as a function of dissolved hydrogen in Nb-H alloys at R=0.05, 0.4 and 0.75.

reaches a minimum at a critical concentration where the maximum embrittlement occurs. As the hydrogen concentration exceeds this critical concentration, ΔKth increases with the increase of hydrogen content in the specimen. Also observed in Fig. 2 is that the values of ΔKth at R=0.4 are lower than those obtained when the load ratio equals 0.05. Moreover, the variation in ΔKth with hydrogen concentration is much reduced at R=0.75.

Crack closure measurements were made on near-threshold stress intensity level at frequency of 0.04 Hz. Figure 3 represents the effective threshold stress intensity range, ΔKeff,th, as a function of hydrogen concentration. From this Fig., one can observe that at R=0.75 the relationship between effective threshold stress intensity range and hydrogen concentration shows the similar behavior as threshold stress intensity range vs. hydrogen concentration. The effective threshold stress intensity range decreases continuously with the addition of hydrogen. This Fig. also shows that at R=0.05 and 0.4, the ΔKeff,th decreases continuously with the addition of hydrogen in solid solution and in the form of hydride and does not exhibit the same trend as ΔKth vs. hydrogen concentration.

In order to determine the mode of fracture which occurred during crack propagation, the fracture paths of fatigued specimens were examined using an optical microscope. Figure 4 displays the fractograph of fracture paths of niobium-hydrogen alloy. Crack branches can be observed at the specimen containing 850 ppm wt. H tested at R=0.75.

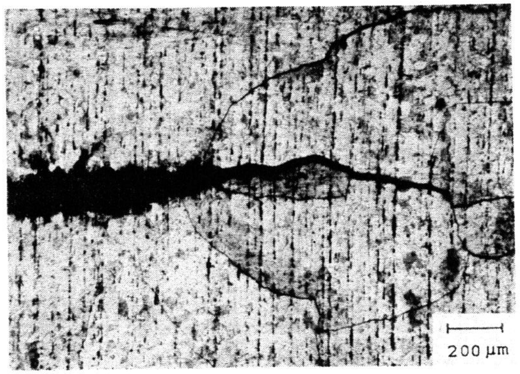

Fig. 4. Fractograph of fracture paths of niobium specimen containing 850 ppm wt. H at R=0.75.

DISCUSSION

From Fig. 2, one can see that at R=0.75, ΔKth decreases continuously even though the amount of hydrogen exceeds 402 ppm wt. which is the stress-free solubility limit of hydrogen in niobium at room temperature (Table I). The critical hydrogen concentration where the maximum embrittlement occurs is not observed as it was reported earlier (Fariabi et al., 1983). This behavior indicates the presence of two mechanisms which are responsible for the embrittlement of niobium with hydrogen. The first mechanism is responsible for the hydrogen embrittlement in the solid solution phase, and the second mechanism becomes operative when the hydride phase is present.

Table I Variations of the critical concentration at maximum embrittlement, Ccr, and the solubility limit, C, as a function of temperature.

T K	Ccr ppm wt.	C ppm wt.
400	260	1271
350	350	775
296	450	402
273	650	253
220	990	75

From their study of ion microprobe analysis for niobium hydride in hydrogen-embrittled niobium, Grossbeck et al., (1976) reported that the thickness of the Nb-H layer is about 1 µm. The SEM evidence also indicated that hydride-like layers occur at fracture surfaces with dimensions in the order of microns (Grossbeck,1975). From Table II, one can see that the magnitude of CTODmax is several order of microns and is comparable with the thickness of the Nb-H layer. This result indicates that there is a possibility that the hydride occurred at fracture surfaces in niobium may contribute to crack closure which is similar to the oxide-induced crack closure proposed by Suresh et al., (1981). However, it has been observed that the thickness of the oxide is in the order of 6 nm below 400 K (Grundner, 1984) and hence no indication of oxide-induced crack closure is to be expected in niobium. Thus, in addition to plasticity-induced crack closure operated at all Nb-H alloys, the formation of hydride may lead to an increase in the magnitude of the crack closure at low load ratios. The hydride-induced crack closure becomes more effective at low temperature since the solubility limit of the specimen is much lower at these temperatures.

Figure 3 shows the variation in ΔKeff,th with the hydrogen concentration. The values of ΔKth and ΔKeff,th are also listed in Table II. These values indicate that except for the test at R=0.75, the intrinsic fatigue crack growth thresholds, ΔKeff,th, do not follow the same trend as the apparent ΔKth values for hydrogen concentration exceeding Ccr. The effect of hydrogen and load ratio in niobium can be seen after separating the influence of crack closure from ΔKth, a significant decrease in ΔKeff,th can easily be observed. Consequently, the crack closure behavior appears to influence the ΔKth markedly. Moreover, ΔKeff,th is essentially independent of load ratio for the same hydrogen concentration and hence crack closure can account for the variation of ΔKth.

The role of hydrogen in influencing the near-threshold fatigue behavior becomes complicated due to the effects of two competing mechanisms between the promoting hydrogen embrittling and the retarding crack closure effect. Table I lists the values of critical concentration, Ccr, where maximum embrittlement occurs and the stress-free solubility limit, C, of hydrogen in niobium. As reported earlier (Polvanich et al., 1987), the value of the activation energy, Q, which represents the slope of the linear relationship between the natural logarithmic of hydrogen concentration for maximum embrittlement, Ccr, and the inverse absolute temperature, 1/T, is equal to 1370 cal/mole or 0.059 eV. This activation energy can be considered as the energy barrier for crack closure to overcome the hydrogen embrittling effect in niobium. Thus, Ccr represents the balance point between hydrogen embrittling and crack closure effect.

Table II Values of threshold stress intensity range, ΔKth, effective threshold stress intensity range, ΔKeff,th, maximum cyclic crack tip opening displacement, CTODmax, and load ratio, R, as a function of hydrogen concentration, Co, for temperature of 296 K.

Co ppm wt.	R	ΔKth MN/m3/2	ΔKeff,th MN/m3/2	CTODmax μm
0	0.75	4.3	3.78	13.12
280	0.75	3.4	3.23	7.04
468	0.75	3.2	2.94	6.08
850	0.75	2.5	2.23	3.52
1181	0.75	2.3	2.07	2.56
0	0.05	8.0	3.28	3.12
	0.4	6.5	3.12	5.19
239	0.05	5.2	3.22	1.13
	0.4	3.5	2.80	1.28
529	0.05	5.2	2.76	1.11
	0.4	3.3	2.67	1.12
1047	0.05	5.5	2.20	0.99
	0.4	3.6	2.34	1.06

The effect of hydrogen and load ratio on fatigue crack growth properties are interrelated. The effect of hydrogen on ΔKth is controlled by the concentration of hydrogen at the crack tip and its influence on crack closure. The hydrogen may induce hardening and softening of material thus affecting plasticity-induced crack closure. Values of maximum cyclic crack tip opening displacement, CTODmax, calculated from experimental results are given in Table II. From this table, one can see that CTODmax increases as load ratio is increased and hence the crack remains open for most portion of the fatigue cycle. The increasing CTODmax associated with a high load ratio leads to reduction in plasticity-induced crack closure under plane stress condition (Budiansky et al., 1978). Table III lists the cyclic plastic zone size, r, from previous microhardness measurements (Fariabi et al., 1984). The cyclic plastic zone size have been observed to show a similar behavior as ΔKth vs. hydrogen dissolved. Furthermore, the values of cyclic plastic zone size at R=0.4 are lower compare to those at R=0.05 and hence, the lower plasticity results in less crack closure. Consequently, the amount of plasticity-induced crack closure increases with decreasing load ratio. The ΔKth values have been observed to be considerably higher where larger plastic zone is formed and hence more plasticity-induced closure at low load ratio. Conversely, ΔKth values are lower resulting from the lower formation of plastic zone at high load ratio.

At a low load ratio R=0.05, the amount of available hydrogen around the crack tip is much less due to the low hydrostatic stress. The effect of closure can be significantly enhanced at low load ratio due to small crack opening displacement. As the hydrogen concentration is above the critical concentration where maximum embrittlement occurs, the hydrogen embrittlement of niobium is offset by overcoming the energy barrier of crack closure. At load ratio R=0.4, the closure effect on specimen reduces significantly and thus the ΔKth does not increase remarkably as the hydrogen concentration exceeds the critical concentration compared to result obtained at R=0.05. Therefore, with the greater crack opening displacement and less cyclic plastic zone size at higher load ratio, ΔKth values would decrease. At a high load ratio of R=0.75, the effect of crack closure is negligible and cannot compensate for the intrinsic embrittlement of hydrogen in solid solution or in the form of hydride. Accordingly, the ΔKth decreases as the hydrogen concentration is increased for all niobium-hydrogen alloys.

Table III Values of plastic zone size, r, and load ratio, R, as a function of hydrogen concentration, Co, for temperature of 296 K.

Co ppm wt.	R	r mm
0	0.05	1.60
	0.4	1.50
70	0.05	6.00
116	0.05	0.42
	0.4	0.44
250	0.05	0.20
	0.4	1.00
360	0.05	0.34
	0.4	0.16
535	0.05	0.50
	0.4	0.14
660	0.05	0.30
	0.4	0.18
1025	0.05	1.80
	0.4	1.45

For specimens containing high hydrogen concentration tested at high load ratio of 0.75, the crack branches can be observed at the region around the crack tip. Nakasa et al., (1986) showed that there exist certain planes where the combination of triaxial tensile stress and concentration of hydrogen atoms trapped by dislocation is optimum for crack nucleation. At high load ratio of 0.75, the specimen is subjected to high external stress and hydrogen will be promoted to move toward the crack tip. Due to the intrinsic embrittlement effect of hydrogen, crack branches start to form at the crack tip region and propagate along the main crack. The distributions of crack branches which are related to the availability of hydrogen at the crack tip can be either continuous or discontinuous.

CONCLUSIONS

From the results obtained in the present research, the following conclusions can be drawn :

(1) The threshold stress intensity range, ΔKth, of niobium is very much influenced by the presence of hydrogen. At lower load ratio R=0.05 and 0.4, the ΔKth increases as the hydrogen concentration exceeds the critical concentration where the maximum embrittlement occurs since the effect of crack closure can compensate for the intrinsic embrittlement of hydrogen in solid solution or in the form of hydride. In contrast, at higher load ratio R=0.75, the ΔKth decreases as the hydrogen concentration increases while the critical concentration where the maximum embrittlement occurs does not exist.

(2) The behavior of the threshold stress intensity range as a function of hydrogen concentration suggests the presence of two mechanisms to be

responsible for the embrittlement of niobium with hydrogen. The first mechanism is responsible for the hydrogen embrittlement in the solid solution phase, and the second mechanism becomes operative when the hydride phase is present.

REFERENCES

Beevers, C.J. (1970). In: the Science, Technology and Application of Titanium, R.I. Jaffe and N.E. Promisel, eds., Pergamon Press, 535-553.

Budiansky, B. and Hutchinson, J.W. (1978). Analysis of Closure in Fatigue Crack Growth. J. of Appl. Mech., 45, 267-276.

Chung, D.W. and Stoloff, N.S. (1978). Fatigue Behavior of Niobium Hydrogen Alloys. Metall. Trans., 9, 1387-1399.

Farahani, M.M., Attia, F. and Salama, K. (1981). Hydrogen Embrittlement in Single and Poly-Crystal. Metall. Trans., 12, 631-638.

Fariabi, S., Collins, A.L.W. and Salama, K. (1983). Effect of Hydrogen on Near-Threshold Crack Propagation in Niobium. Metall. Trans., 14, 701-707.

Fariabi, S., Collins, A.L.W. and Salama, K. (1984). Effects of Plastic Zone in Near Threshold Fatigue Crack Propagation in Niobium. In: ICF 6, S.R. Valluri, D.M.R. Taplin, P. Rama Rao, and J.F. Knott, eds.

Gahr, S. Grossbeck, M.L. and Birnbaum, H.K. (1977). Hydrogen Embrittlement of Nb. Acta Metall., 25, 125-134.

Gray III, G.T. and Luetjering, G. (1984). The Influence of Crack Closure on Fatigue Crack Propagation of Ti alloys.In: Fatigue 84, C.J. Beevers, ed.

Grossbeck, M.L. (1975). Ph. D. Thesis, University of Illinois, Urbana.

Grossbeck, M.L., Birnbaum, H.K., Willams, P. and Evans, C.A. (1976). Ion Microprobe Analysis for Niobium Hydride in Hydrogen-Embrittled Niobium. Anal. Chem, 48, 964-968.

Grundner, M. and Halbritter, J. (1984). On the Natural Nb_2O_5 Growth on Nb at Room Temperature. J. Surface Science, 136, 144-154.

Halliday, M.D. and Beevers, C.J., (1981). Some Aspects of Fatigue Crack Closure in Ti Alloys. J. of Testing and Evaluation, 9, 195-201.

Kikukawa, M., Jono, M. and Tanaka, K. (1976). Fatigue Crack Closure Behavior at Low Stress Intensity Level. In: Proc. of 2nd Int. Conf. on the Mechanical Behavior of Materials, Boston. 716-720.

Lee, K.S. and Stoloff, N.S. (1976). Fatigue of Vanadium-Hydrogen Alloys. Proc. of Int. Conf. on Effect of Hydrogen on Behavior of Materials, A.W. Thompson, and I.M. Bernstein, eds., 404-414.

Nakasa, K. and Takei, H. (1986). Crack Branching Angel of High Strength Steels in Delayed Failure. Engng. Fracture Mech., 24, 513-522.

Owen, C.V., Spitzig, W.A. and Buck, O. (1987). Effects of Hydrogen on Low Temperature Hardening and Embrittlement of V-Cr Alloys. Metall. Trans. 18.

Pendse, R.D. and Ritchie, R.O. (1985). A Study of Crack Propagation in Prior Hydrogen Attacked Pressure Vessel Steels. Metall. Trans., 16, 1491-1501.

Polvanich, N. and Salama, K. (1987). Temperature Dependence of Fatigue Crack Propagation in Niobium-Hydrogen Alloys. In: Hydrogen Embrittlement : Prevention and Control, ASTM STP 962, L. Raymond, ed., 417-427.

Shih, T.T. and Wei, R.P. (1974). A Study of Crack Closure in Fatigue. Engng. Fracture Mech., 6, 19-32.

Suresh, S., Zamiski, G.F. and Ritchie, R.O. (1981). Oxide-Induced Crack Closure: An Explanation for Near-Threshold Corrosion Fatigue Crack Growth Behavior. Metall. Trans. 12, 1435-1444.

Todd, J.A., Li, P., Liu, G. and Raman, V. (1988). A New Mechanism of Crack Closure in Cathodically Protected ASTM A710 Steel. Scripta Metall., 22.

Wilcox, B.A. (1964). Fatigue Behavior of Hydrogen Charged Tantalum. Trans. of AIME, 230, 1199-1200.