THE ROLE OF NON-METALLIC INCLUSIONS IN HYDROGEN ASSISTED CRACKING OF STEEL

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ABSTRACT

The effect of inclusion volume fraction and orientation on hydrogen assisted cracking has been studied in a medium carbon hardenable steel. Crack growth rate and threshold stress intensity are found to be strongly dependent on orientation for a high strength condition tested in salt water and a low strength condition tested in H_2S saturated salt water. In the low strength condition crack growth is intermittent at low stress intensities and involves a combination of matrix embrittlement and internal pressurisation at the inclusions. The results are interpreted in terms of a critical stress and distance failure criterion, the orientation dependence arising from inclusion induced perturbations to the crack tip stress field.

KEYWORDS

Steel; hydrogen embrittlement; threshold stress intensity; inclusions; anisotropy; hydrogen sulphide.

INTRODUCTION

It is now well established that deformed non-metallic inclusions are mainly responsible for the pronounced orientation dependence in the fracture toughness of wrought steels. By comparison rather little is known about their effect on hydrogen assisted cracking behaviour. Traditionally hydrogen effects have been studied predominantly in high strength steels and early work by Davis (1963) and Hughes, Lamborn and Liebert (1965) demonstrated a strong orientation dependence in time to failure tests on smooth bar specimens. These results relate mainly to the effect of inclusions on crack initiation and provide little information on crack growth resistance. More recently, attention has been focussed on hydrogen induced cracking in lower strength steels following the failure of a number of pipelines handling sour oil and gas. In all of these failures it has been found that the fracture process involved the active participation of inclusions, usually highly elongated type II MnS (Moore and Warga, 1976). The work reported here is part of a systematic investigation of the effects of inclusion orientation and volume fraction on threshold stress intensity and crack growth behaviour for steels having a range of matrix strengths and exposed to environments of different hydrogen activity.

EXPERIMENTAL

Two medium carbon hardenable steels containing different sulphur contents but otherwise of the same nominal composition were selected for this investigation. One of the steels (En16C) contained a sulphur content of 0.038%, the other (En16M) was a resulphurised grade containing 0.15%S. The complete chemical analysis of the steels is given in Table 1. Both steels were forged at lower than normal hot working temperatures in order to increase the relative plasticity of the manganese sulphides. This produced a dispersion of highly elongated inclusions, the ratio of semi-axes being typically 50:4:1. The steels were quenched and tempered to produce a range of strengths, the results reported here being confined to the 1300 MPa and 850 MPa yield strength conditions.

TABLE 1 Chemical Composition of Steels (wt%)

	C	Si	Mn	Mo	P	S
En16C	0.37	0.26	1.35	0.24	0.009	0.038
En16M	0.32	0.21	1.32	0.23	0.009	0.15

For the measurement of K_{TH} and crack growth rates, 25mm thick modified T-type WOL specimens were employed. Specimens were machined with LT, TL, ST and SL orientations (Fig.1), side grooves being employed to inhibit crack deflection. After fatigue pre-cracking, each specimen was loaded to a high initial stress intensity and the crack allowed to grow under decreasing K conditions until eventually it arrested. A compliance technique was used to monitor crack length, the load and mouth opening being monitored continuously by an instrumented loading pin and a clip gauge respectively. The testing environment was 3½% NaCl solution for the high strength condition and H_2S saturated 3½% NaCl for the lower strength condition.

Crack Growth Behaviour

The crack growth behaviour of the lower sulphur steel at a yield strength of 1300 MPa is shown in Fig.2. All of the orientations exhibit the growth behaviour characteristic of hydrogen assisted cracking, namely a K independent stage II and a steeply falling stage I leading to an apparent threshold stress intensity, K_{TH}. A pronounced effect of testing orientation is apparent, resistance to crack initiation and growth being highest in the LT orientation and least in the SL.

The results from the higher sulphur steel, tested at the same strength and in the same environment, are shown in Fig.3. This reveals a wider spread in stage II growth rates and more pronounced anisotropy in threshold values. When the K_{TH} values are plotted against sulphur content as in Fig.4., it appears that there is a unique threshold value for the steel matrix that may be either increased or decreased by the presence of inclusions depending on their orientation and volume fraction. It is interesting to note that in the TL orientation an increase in sulphur content produces a decrease in the static fracture toughness (K_{Ic}) but an increase in K_{TH}.

When tempered to a yield strength of 850 MPa both steels were resistant to cracking in 3½% NaCl and it was necessary to employ H_2S saturated solutions. The resulting crack growth behaviour in the short transverse and TL orientations is illustrated in Fig.5. The longitudinal orientation is not included because the stress intensity necessary to induce cracking was beyond the range of plane strain validity. The cracking susceptibility shows a similar dependence on orientation and sulphur content to that observed in the higher strength condition.

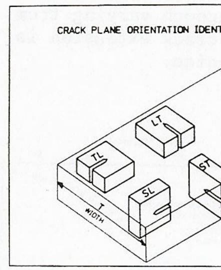

Fig.1. Orientation of test specimens

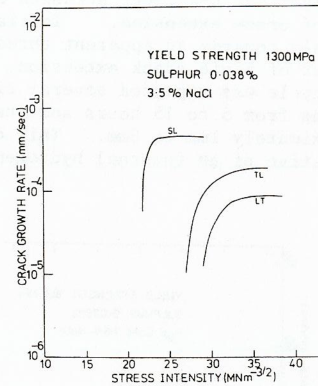

Fig.2. Crack growth behaviour of high strength steel containing 0.038%S

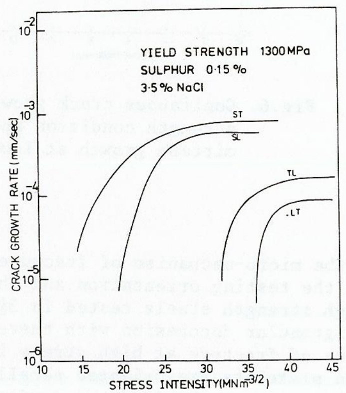

Fig. 3. Crack growth behaviour of high strength steel containing 0.15%S.

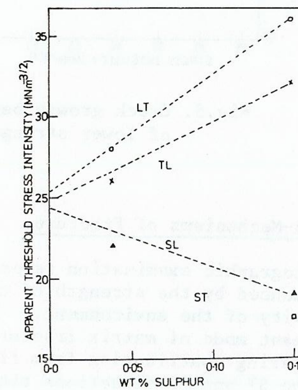

Fig. 4. Effect of sulphur content on threshold stress intensity

The significant difference is in the stage I growth behaviour. Whereas the higher strength steels tend to exhibit a near vertical stage I leading to a clearly defined K_{TH}, the lower strength steels reveal a progressive reduction in growth rate with decreasing stress intensity, there being no indication of any true threshold. The detailed nature of the crack growth behaviour is also quite different in the two strength conditions as illustrated in Fig.6. At the higher strength, cracking is continuous on the macroscopic scale and undergoes a smooth transition to zero growth as the stress intensity decreases with increasing time. By comparison the lower strength steel in H_2S saturated NaCl shows a discontinuous mode of crack extension. Initially the crack growth rate appears to decrease smoothly towards an apparent threshold condition but this is then interrupted by a burst of rapid crack extension. As shown in Fig.6, the deceleration/acceleration cycle was repeated several times, the time period between the crack bursts varying from 5 to 15 hours and the increment of crack growth varying from approximately 1mm to 3mm. This discontinuous mode of crack extension is indicative of an internal hydrogen pressurisation mechanism.

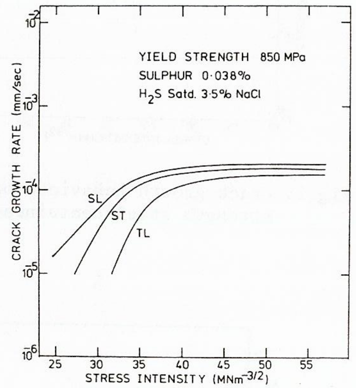

Fig.5. Crack growth behaviour of lower strength steel

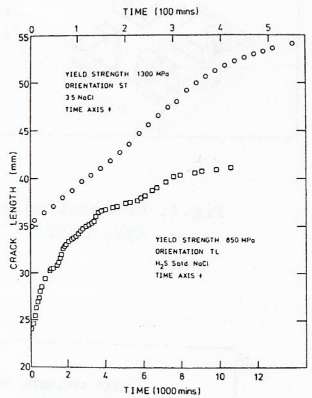

Fig.6. Continuous crack growth in high strength condition and intermittent growth at lower strength

Micro-Mechanisms of Fracture.

Fractographic examination revealed that the micro-mechanism of fracture was influenced by the strength of the steel, the testing orientation and the hydrogen activity of the environment. In the high strength steels tested in $3\frac{1}{2}$%NaCl, the dominant mode of matrix fracture is intergranular decohesion with there being an increasing contribution from fibrous modes of fracture at high stress intensities. In the ST and SL directions the inclusion platelets are oriented parallel to the fracture plane and their very high concentration on the fracture surface demonstrates that they provide preferential paths for crack propagation, (Figs.7 & 8).

In the TL orientation the inclusion platelets are oriented at right angles to the fracture plane with their major axes running in the direction of crack propagation. The fracture surfaces show evidence of decohesion of the inclusion matrix interfaces in the thickness direction, this resulting in the development of numerous parallel splits running in the direction of crack propagation (Fig.9). In the LT orientation, the inclusion platelets are at right angles to the fracture plane with their axes normal to the direction of crack propagation. Again decohesion of the inclusion matrix interface is observed and the presence of this weak interface at right angles to the macroscopic direction of crack propagation causes repeated local deflection of the crack into the short transverse plane (Fig.10).

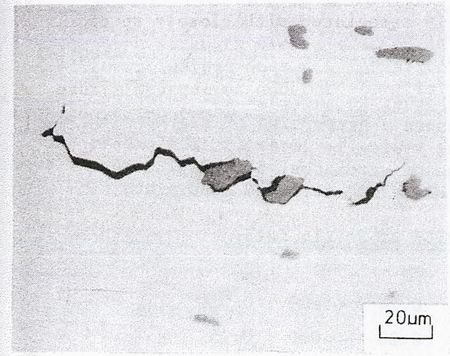

Fig.7. Nickel plated fracture from high strength steel in ST orientation.

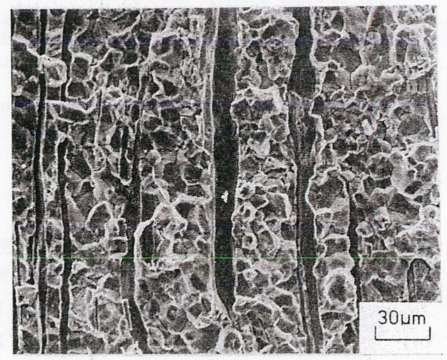

Fig.8. Fracture surface from high strength steel in SL orientation.

It should be noted that chemical cleaning of the fracture surfaces was necessary to permit examination in the scanning electron microscope and this resulted in the dissolution of the majority of the manganese sulphides. However, the inclusions were retained on the nickel plated sections (e.g. Fig.10) and it is significant that they show no evidence of chemical attack by the salt water solution.

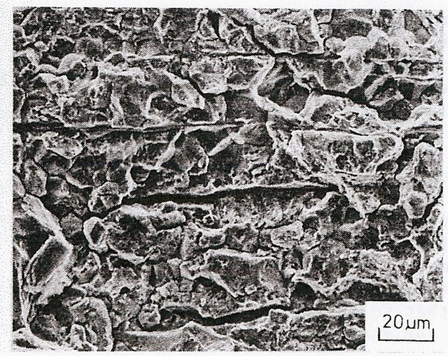

Fig.9. Fracture surface from high strength steel in TL orientation.

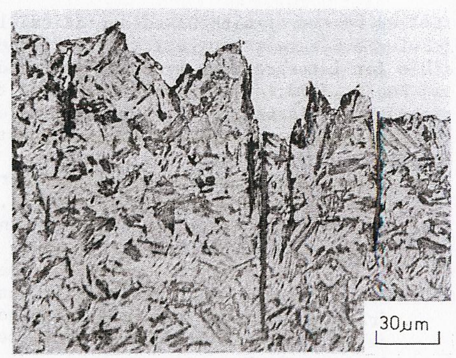

Fig.10. Nickel plated fracture from high strength steel in LT orientation.

The fracture surfaces from the steels in the lower strength condition reveal a much greater participation of inclusions (Fig.11). This reflects the increased size of the plastic zone associated with the lower yield strength. It is also of note that the mechanism of matrix separation has changed to a predominantly quasi-cleavage mode. The effect of orientation on the topography of the fracture surfaces was generally similar to that observed in the higher strength condition.

However, a significant difference was the observation of secondary cracks developing at quite large distances ahead of the main crack. A typical example is shown in Fig.12, this being located approximately 200μm ahead of the main crack. It is of note that the secondary cracks were invariably associated with closely spaced groups of inclusions.

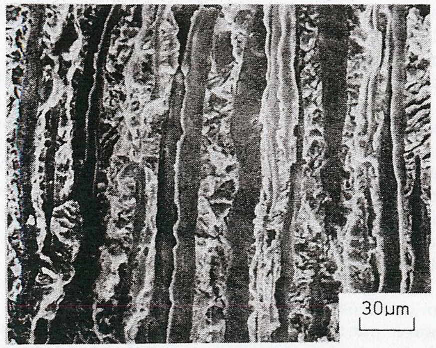

Fig.11. Fracture surface from lower strength steel in SL orientation.

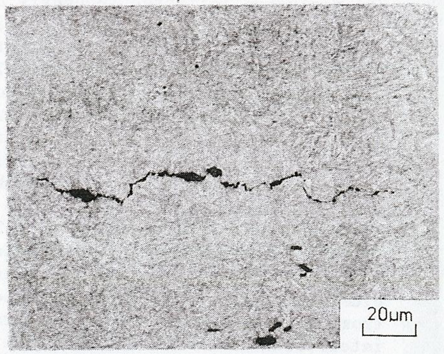

Fig.12. Secondary crack developing ahead of main crack in lower strength steel.

DISCUSSION

It is well established that transgranular cleavage fracture in mild steel is controlled by the attainment of a critical value of tensile stress and there is now growing evidence to suggest that a similar stress controlled mechanism is responsible for intergranular fracture induced by temper embrittlement (Ritchie, Geniets and Knott, 1973; Kameda and McMahon, 1980). Hydrogen assisted cracking in high strength steels bears many similarities to temper embrittlement and it is likely that this too is a critical stress controlled mechanism of fracture. Support for this proposition is provided by early work on hydrogen induced cracking from blunt notches by Troiano (1960) which showed that crack nucleation occurred below the surface at the point of maximum triaxial stress, and by a recent analysis of the effects of hydrogen pressure and matrix strength on K_{TH} values for cracking in gaseous hydrogen environments (Akhurst, 1979).

In specimens containing sharp cracks, the maximum tensile stress which can be attained ahead of the crack tip is the triaxially constrained yield stress, this being approximately 3 times the uniaxial yield stress. As the stress intensity increases there is no significant change in the magnitude of the maximum tensile stress but the region of plastic stress intensification moves progressively further away from the crack tip (Rice and Johnson, 1970). By analogy with the model for cleavage fracture toughness proposed by Richie, Knott & Rice (1973), it is suggested that K_{TH} represents the minimum value of stress intensity which will allow the attainment of a critical fracture stress σ_f, over some critical distance ahead of the crack tip. The latter is microstructurally controlled and in the case of an intergranular mode of fracture is likely to be some multiple of the prior austenite grain size.

Hydrogen is assumed to lower the cohesive strength of the steel, the grain boundaries being preferred fracture paths in the high strength condition due to the prior segregation of impurity elements.

In the high strength steels studied in this investigation the presence of non-metallic inclusions does not appear to produce any change in the basic mechanism of matrix separation and therefore it is reasonable to assume that they do not influence the critical distance criterion for hydrogen induced cracking. Their observed effect on cracking susceptibility must arise either by influencing the local concentration of dissolved hydrogen or by modifying the stress distribution in the crack tip process zone.

Manganese sulphide inclusions can dissolve in acidic solutions and therefore it is conceivable that they could interact with the crack tip environment and thereby increase the local hydrogen activity. The results of the present work indicate that this was not a significant factor affecting susceptibility to cracking. In the first place there is no evidence of chemical attack on the inclusions exposed on the fracture surface (Fig.10). Secondly, if there was a significant effect, the K_{TH} values would be expected to decrease and the stage II growth rates increase with increasing sulphur content. As shown in Fig.4, this was not observed for the LT and TL orientations. Finally, the opportunity for environmental attack is greatest when the inclusions have their major axes oriented at right angles to the exposed surface. In the present work this corresponds to the LT orientation which in fact exhibited the greatest resistance to cracking.

One other way in which the elongated inclusions might influence the concentration of hydrogen at the crack tip is by providing favourable diffusion paths and thus introducing anisotropy in the diffusivity of the steel. To check this, hydrogen permeation was measured as a function of orientation using the electrometric permeation technique (Devanthan and Stachurski, 1963), the input and exit surfaces being coated with palladium to avoid the possibility of the inclusions affecting the hydrogen activity. No significant effect of orientation was observed. This is supported by recent work by Pumphrey (1979) where no effect on permeability was observed for different orientations exposed to environments containing H_2S.

If the inclusions do not influence the supply of hydrogen then the major effect on cracking susceptibility must arise from their influence on the local stress field ahead of the crack tip. In the case of manganese sulphides, the inclusion matrix interface is weak and is observed to separate at very low tensile strains, probably within the elastic regime. In the SL and ST testing orientations, decohesion of the inclusion matrix interface leads to a concentration of strain in the adjacent steel matrix. Within the crack tip plastic zone, the maximum tensile stress is already about 3 times the uniaxial yield stress, this being a consequence of the triaxial state of stress induced by the surrounding elastic stress field. It follows that the strain concentration at the tips of the inclusions can produce no further increase in the maximum tensile stress. It should be noted, however, that the sizes of the plane strain plastic zones corresponding to the measured K_{TH} values are very small, ranging from about 15μm to 40μm. This is comparable to the inclusion centre to centre spacing. It follows that an elongated inclusion which has its centre in the general vicinity of the elastic-plastic boundary must have its far end in what is nominally the elastic region. Under these conditions there can and will be a stress concentration at the tip of the inclusion and a further local region of constrained plastic yielding can develop. The maximum stress which can be attained is still limited by the value of the constrained yield stress; however, the presence of the elongated inclusions increases the extent of the high stress region ahead of the crack tip. This allows the critical distance criterion to be satisfied at a lower applied stress intensity than would be possible in the absence of the inclusions. Also any increase in the inclusion volume fraction increases the spread of plasticity and hence further decreases K_{TH}. This is consistent with the observations.

In the TL orientation, decohesion of the interface occurs preferentially in the thickness direction. The most significant consequence is a local relaxation of lateral constraint which tends to produce a state of plane stress adjacent to the inclusions. This effect is clearly apparent in fibrous fractures produced by testing in air where 45° shear lips are observed adjacent to the inclusion induced splits. In the context of a critical stress controlled fracture process, any relaxation in plane strain constraint lowers the maximum stress which can be achieved in the plastic zone and thus makes the attainment of the critical stress and distance criterion more difficult. This mechanism has been exploited in the development of weakly bonded laminated steels which are resistant to cleavage fracture (Almond and co-workers, 1969) and is the suggested explanation for the observation of reduced fibrous to cleavage fracture transition temperatures in mild steels tested in the transverse and longitudinal orientations (Pickering, 1978). In the case of hydrogen cracking, a reduction in triaxiality has a further beneficial effect in that it reduces the equilibrium concentration of hydrogen in the crack tip process zone, thus producing an increase in σ_f. The combination of the decreased stresses in the plastic zone and the increased value of σ_f, means that the critical stress and distance criterion is more difficult to satisfy, this being reflected in an increase in K_{TH}. Unlike the SL and ST orientations, an increase in sulphur content should produce an increase in K_{TH} for the TL orientation. This is observed.

In the LT orientation, interface separation again takes place in the thickness direction and the resultant decrease in constraint would be expected to be reflected in a similar increase in K_{TH} to that observed in the TL orientation. However, an additional factor in this orientation is the observed multiple crack bifurcation caused by the weak inclusion interfaces running at right angles to the plane of fracture. This causes a further reduction in the effective stress intensity at the crack tip and may account for the observed increased K_{TH} values in this orientation.

When tempered to a yield strength of 850 MPa, both steels were resistant to cracking in the 3½%NaCl solution. This implies that the maximum stress in the plastic zone was insufficient to exceed the critical fracture stress. By increasing the hydrogen activity of the environment, the fracture stress is decreased and cracking again becomes possible.

The resistance to cracking in the LT orientation implies that the threshold value for hydrogen assisted cracking in the absence of inclusions is very high. This is supported by the crack growth behaviour in the TL and SL orientations which start to deviate from the K independent stage II at stress intensities in excess of 40 MPa $m^{\frac{1}{2}}$. Despite this, cracking continues to occur down to very low stress intensities with there being no indication of any true threshold. The discontinuous nature of the growth in this low K regime, the long time intervals between bursts of growth and the observation of disconnected cracks developing far ahead of the main crack, demonstrates that internal hydrogen pressurisation is playing a decisive part in allowing cracking to develop in the low strength condition. This is supported by the observation that thin samples of the steel develop surface blisters when exposed to the H_2S saturated NaCl solution.

It is well established that in environments containing hydrogen at high activity very high internal pressures can develop at non-metallic inclusions. However, the presence of an internal source of stress alone is not sufficient to account for the observed cracking. The quasi-cleavage mechanism of matrix separation in the intermittent growth regime is identical to that at high growth rates and

implies that a true hydrogen embrittlement mechanism is still operative. If the fracture process continues to be controlled by a critical stress and distance criterion, it follows that the internal pressurisation at the inclusions must not only fulfil the stress requirement but also allow the distance criterion to be satisfied at stress intensities lower than those necessary for cracking in the absence of inclusions.

The inclusion initiated secondary cracks were observed to develop at distances from the crack tip which were considerably in excess of the plane strain plastic zone size. In this situation, internal pressurisation results in a stress concentration at the tip of an inclusion which eventually can lead to the development of a local region of constrained yielding. The extent of the area of stress intensification is controlled by the degree of internal pressurisation and on the inclusion size and frontal radius of curvature. In the case of the lower strength steels, this local zone size is expected to be considerably smaller than that associated with the main crack. Also the maximum local stress arising from inclusion pressurisation can be no greater than that present in the plastic zone associated with the main crack. This implies that an isolated inclusion is unlikely to act as a site for secondary crack formation. This is consistent with the observations; secondary cracks were only observed to be associated with clusters of closely spaced inclusions. In the latter situation the stress fields from adjacent inclusions can interact to produce an extensive area of stress intensification. As the internal pressure continues to increase, the critical distance criterion is eventually satisfied and thus permits a burst of crack growth. The increment of crack growth in such bursts is of course limited by the extent of void pressurisation ahead of the crack tip.

In the present work, observations of hydrogen assisted cracking were confined to pre-cracked specimens in which the stress intensification arising from internal pressurisation supplemented that associated with the pre-existing crack. However, if there is a sufficient availability of hydrogen at high activity and if inclusions are sufficiently closely spaced, it should be possible to satisfy both the stress and distance requirements for hydrogen embrittlement by internal pressurisation alone. It is suggested that this is the explanation for the observed cracking of line pipe steels in sour oil environments where the highly elongated condition of the manganese sulphide inclusions satisfies the requirement of close proximity in spite of there being much lower inclusion volume fractions than those studied in this work.

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