JOURNAL OF MATERIALS SCIENCE **28** (1993) 3322–3328

Formation and analysis of stack cracks in a pipeline steel

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Extensive cracking of the type known as stack cracking was demonstrated in a cathodically charged X65 microalloyed pipeline steel containing a weldment. It is shown that the formation and propagation of rolling-plane cracks, which constitute the primary stages of the stack cracking, is due to local concentration of hydrogen gas pressure and a lowering of the cohesive strength of a number of interfaces by hydrogen. The characteristic S-shape of individual cracks which occurred during the linking up of cracks was attributed to stress interactions at crack tips and cleavage cracking normal to the rolling plane. An explanation of hydrogen embrittlement fracture is given in terms of electronic state modifications of the steel, including charge polarization.

1. Introduction

In an investigation to study the formation of stack cracks and their relation to stress and metallurgical state it was found that in cathodic hydrogen charging tests simulating sour service pipeline conditions, stack cracking occurred in an X65 microalloyed pipeline steel containing a weldment [1, 2]. The steel was silicon and aluminium deoxidized and calcium treated and thus contained very low sulphur (0.001 wt %). Extensive cracking was observed near the weldment and only minor cracking was seen in the parent metal. Longer exposure times were required for cracks to develop in the parent metal than in a region near the weldment [1, 2].

This paper is concerned with the mechanism of initiation and formation of stack cracks. A model is proposed of the fracture process associated with the stack cracking based on experimental observations. Details of the experimental procedure have been published previously [2].

2. Initiation and formation of stack cracks

It has been found experimentally that the presence of a stress, internal or external, was not an essential prerequisite to cause stack cracking [1]. It was also found that most of the stack cracks initiated at inclusions; the majority of these inclusions did not contain sulphur. Thus, it was possible that some of these inclusions produced tessellated stresses (tension) in the matrix due to the difference in thermal contractions between inclusions and the matrix, as suggested by Brooksbank and Andrews [3–6]. In the presence of tessellated stresses, hydrogen will diffuse or be transported by dislocations and accumulate around these inclusions. The subsequent build-up of hydrogen pressure allied with low adhesive forces between the inclusions and the surrounding matrix will promote the formation of cracks.

Additionally, during rolling of the steel plate the initial (as-cast) inclusions would be fragmented and produce small fragments of inclusions as strings of small incoherent inclusions in the rolling plane; voids were seen to form at the end of these inclusions. Hydrogen will accumulate at these individual inclusions and the associated voids; subsequent hydrogen pressure build-up will enlarge the voids formed and initiate/propagate cracks in the weakly bonded (rolling plane) interface in which the inclusions are located. These cracks, from individual inclusions, subsequently join up to form long macro-cracks in the rolling plane (Fig. 1a and b). This coexistence of weak interfaces, voids at inclusions (fragmented oxides, silicates or carbides) in the rolling plane, and internal pressure from the accumulated molecular hydrogen pressure, causes the formation of the arrays of cracks parallel to the rolling direction.

It has been demonstrated experimentally [2] that hydrogen gas pressure alone will produce this cracking, i.e. in the absence of an externally applied stress. It was also found that rolling plane cracks occurred most frequently in a region close to the weld, identified as a "soft" region, i.e. a low yield-strength region [2]. This type of rolling plane crack also occurred in the parent metal. However, a greater number of cracks were observed in the soft region than in the parent metal (Fig. 2a and b) and a much shorter charging

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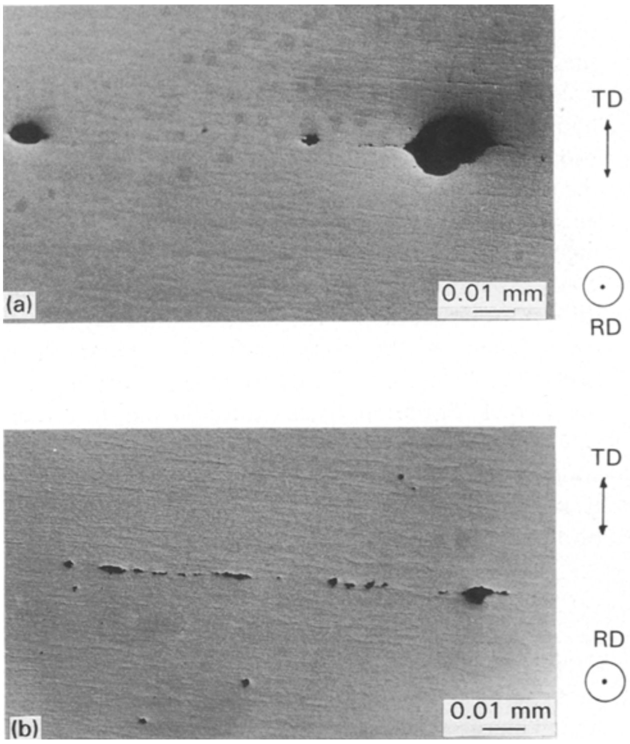

Figure 1 Inclusion-related voids which have enlarged due to hydrogen pressure, showing incipient cracking from the voids in the rolling plane: (a) after a short charging time; (b) after a slightly longer charging time. Specimens hydrogen charged at zero applied stress. TD, thickness direction; RD, rolling direction.

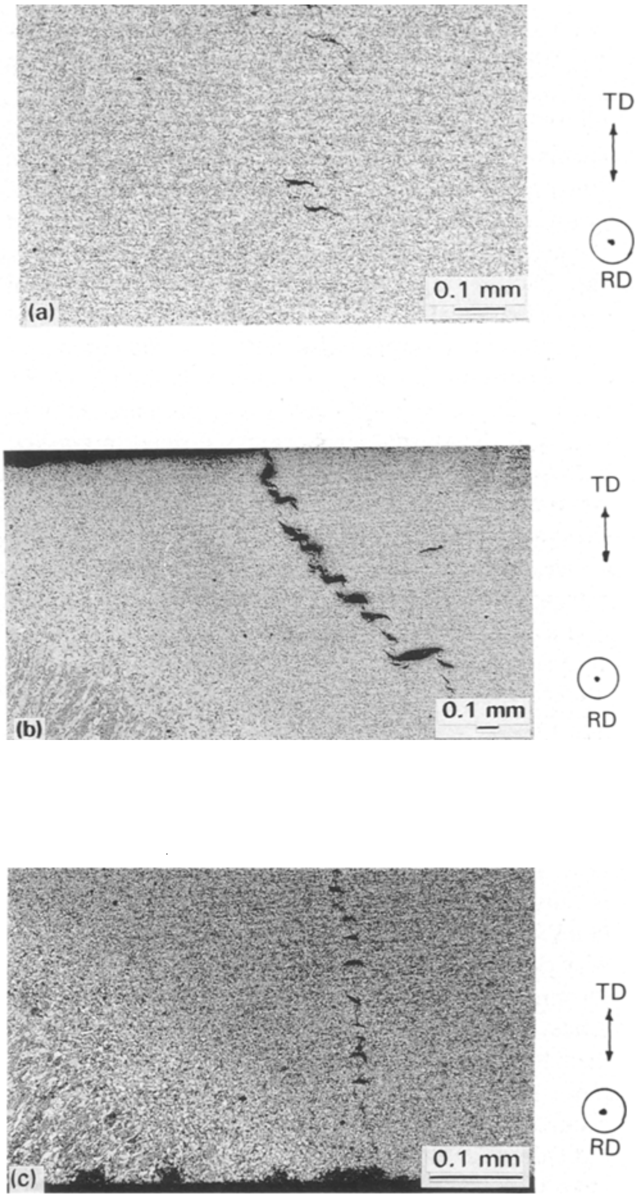

Figure 2 Examples of stack cracks. (a) Cracks in parent metal, charged for 1500 h at 0.2 yield stress. (b) A larger fraction of linked stack cracks in the soft zone near the weldment, charged for 1500 h at 0.2 yield stress. (c) Stack cracks formed in the soft zone of a specimen charged for 167 h at zero applied stress.

time was required for cracks to develop in the soft region compared to the parent metal. A higher incidence of linking of cracks, i.e. transverse or vertical cracks, was also observed in the soft region. Both effects are probably due to the lower yield strength of the soft region [1, 2] which allows a lower molecular hydrogen pressure to expand rolling-plane cracks in the region and similarly, the linking of cracks will occur at a lower internal hydrogen pressure.

The applied stress affects only the time to final fracture. The initial branching or turning of the rolling plane cracks to form the characteristic "S" shape was probably due entirely to the hydrogen pressure [1]. The frequent observation by SEM (energy-dispersive analysis of X-rays, EDAX) of titanium on the fracture surfaces suggests that hydrogen probably accumulated at TiC or TiCN precipitates (strong hydrogen traps); under the influence of stress these hydrogen atoms will be released and diffuse or be transported by dislocations to weak interfaces such as inclusion/matrix, cementite/matrix and/or carbide/matrix interfaces where recombination occurs to form molecular hydrogen with a consequent pressure build-up and eventual cracking. The role of dislocations in transporting hydrogen atoms was anticipated and was demonstrated in an experiment [1] where cathodically charged specimens, which had been previously coated on the flat faces with a hydrogen impervious coating, fractured in the soft region under the protective coating; in these specimens the soft region was at 3 mm depth from the edge of the coating. The time to fracture was about 2 h more than in the same type of specimen without the protective coating. It was calculated that by lattice diffusion mechanisms, the hydrogen concentration front could have moved only a few micrometres in 1 h [7, 8] while by dislocation transport a depth of a few centimetres would have been achieved [9].

The extension of rolling-plane hydrogen-induced cracks has been considered by Iino [10] and also Ikeda *et al.* [11]. In his investigation with X65 grade steels, prepared by laboratory-scale melting, and containing sulphur ranging from 0.003 %–0.007 %, Iino found that the formation of these cracks (referred to as hydrogen blister-crack array) was caused by hydrogen precipitation at an inclusion/matrix interface and that the manner of the subsequent hydrogen-induced fracture is considerably influenced by external stresses. In the absence of an external stress, fracture was predicted to develop by step-wise linking of the cracks (blisters), whereas in the presence of an external stress (sufficiently high compared to zero and sufficiently low compared to the yield strength of the material) fracture developed by linking of the blisters formed in stacked arrays in a plane approximately perpendicular to the external stress axis, with blister size and spacing of the order of 100 μm. The specimens used in Iino's experiments did not contain a weldment and the grain size of the steels was not reported. Iino discussed the extension and propagation of the crack leading to the fracture in relation to the stress distribution around the hydrogen-induced cracks and the role of the external stress was also related to the characteristic extension, stacking and step-wise linking of the cracks. Iino concluded that in the presence of sufficiently high external stress cracks formed in stacked arrays in a plane approximately perpendicular to the stress axis, while in the absence of an external stress, the cracks link in a stepwise manner. Contrary to the above, in the present experiment it was found that external stress did not significantly effect the manner of the formation and extension of the cracks, i.e. in the absence of external stress the cracks still form in a stacked array in the soft region (near the weldment [1], Fig. 2c.

Ikeda *et al.* [11] analysed the stress and strain distributions to explain crack extensions and linking around two small cracks parallel to the plate thickness. The linking of the primary (rolling plane) cracks is caused by interactions in the stress fields of the two crack tips. When the tips of two parallel cracks are very close, the two cracks will join-up directly without forming small secondary cracks. However, if the distance between the two cracks is quite large, intermediate (secondary) crack(s) could develop in-between these primary cracks, and the crack extension will occur by linking of these small cracks. Additionally, individual small cracks also develop (due to weak interfaces such as inclusions) in front of a primary crack and this also promotes linking. In developing their analysis, Ikeda *et al.* assumed that (i) separation between matrix and inclusion occurs at an early stage and plastic deformation assists it, (ii) hydrogen embrittlement occurs in the plastic region, and (iii) cracks propagate normal to the applied tensile stress. They concluded that an external applied stress might assist the crack development through the superposition of resolved stresses on the internal hydrogen pressure at potential cracking sites. Whereas Ikeda *et al.* found that cracks generally initiated at MnS inclusions, in the present work cracking and cleavage fracture were initiated from incoherent non-metallic inclusions of the type oxide, silicate, aluminate or spinel but not MnS, probably because the low sulphur content (0.001 %) of the steel precluded the formation of MnS. Ikedo *et al.* identified hydrogen embrittlement of plastically deformed regions, while the experience of the current work [1, 2] is that plastic deformation, by virtue of removing the soft region (low yield-strength region) from the microstructure, precludes hydrogen embrittlement rather than promoting it. Ikeda *et al.* observed only secondary cracks parallel to the primary crack, while secondary cracks transverse to the rolling plane were observed in the current experiments. This latter observation was ascribed to the presence of cementite films at grain boundaries transverse to the rolling plane. Despite these differences, the analysis of the development of stack cracking due to hydrogen embrittlement by Iino and Ikeda *et al.* is broadly in accord with the interpretation proposed in the current work, i.e. there is agreement on the mechanism of stack cracking due to hydrogen embrittlement. This mechanism consists of a sequential cracking and crack linking process which may have different time characteristics for individual steps in the development of the stack cracks, i.e. characteristic "delay" or "incubation" times may exist, e.g. for primary crack formation, crack extension and the formation of the "S" tail at the end of primary cracks, similar consideration may apply for formation and fracture of transverse cleavage crack, also for the link-up of the primary rolling plane and transverse cracks.

3. Mechanism and fracture model

In a hydrogen-charged steel containing particulate imperfections such as oxide and or silicate particles or inclusions, hydrogen atoms can precipitate as molecular hydrogen in a microcrack or inclusion matrix interfaces and exert an internal pressure, P, which will operate on the crack in a hydrostatic manner. It was concluded [1] that hydrogen embrittlement by a pressure mechanism may not be the only mechanism involved and that a decohesion mechanism also may have contributed to crack propagation. According to Oriani's decohesion theory [2], hydrogen will reduce the cohesive force between atoms and cause cleavage cracks to be formed at a lower stress than in the absence of hydrogen.

As the crack length increases from a nominal a to $a + \Delta a$ the hydrogen pressure inside the crack will decrease instantaneously and the extension of this crack will either cease or slow down depending on local energy criteria and the local hydrogen concentration. After time Δt, under continuous hydrogen charging or conditions of supersaturation of lattice hydrogen, more hydrogen will precipitate and the hydrogen pressure will increase to a level P_{crit}, which promotes crack extension. This is illustrated in Fig. 3. This may not, however, occur uniquely in the largest crack because the required pressure increment, ΔP, in some of the smaller cracks will be reached earlier than the large crack because less hydrogen is required in smaller cracks to generate ΔP than in larger cracks, and will thus selectively cause these to propagate, possibly by cleavage. However, some of the cracks may be stopped, deflected or blunted, e.g. by coherent or incoherent particles, grain boundaries or a weak interface.

If the cracks are sufficiently near to one another then, with time, the effect of a hydrogen concentration gradient between an instantaneously low pressure region (a large crack) and high pressure region (small cracks) may operate. The concentration gradient will provide a driving force for diffusion of hydrogen to the large crack and create conditions for crack extension

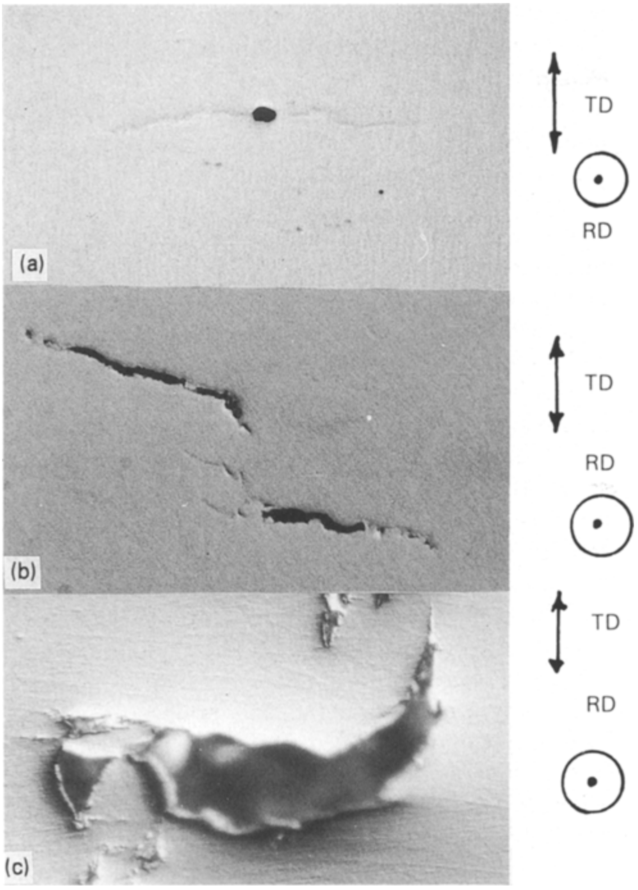

Figure 3 Showing the development of a small inclusion-initiated crack into a characteristic S-shaped stack crack. (a) Charging time, t; hydrogen pressure P_0; crack length a_0. (b) Charging time $t + \Delta t$; hydrogen pressure, P_{crit}; crack length, $a_0 + \Delta a$; (c) $P(< P_0)$, linking.

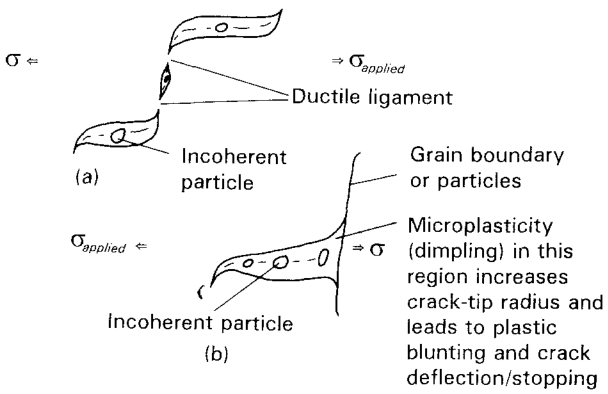

Figure 4 Diagramatic representation of a selection of some of the types of (a) crack linking and (b) crack blunting/stopping observed experimentally.

(increase in hydrogen pressure), if not hindered or stopped as described above. This sequence will be repeated in step-wise events with cracking, stopping, hydrogen diffusion, pressure drop and pressure build-up being sub-steps in the sequence. The sequential nature of the development of the characteristic S-shape is shown in Fig. 3.

This time-dependent sequence can be expressed in terms of "delay" times which involves a large number of cracks, some of which will not contribute to the final fracture. This sequence will continue to cause cracking and linking in the observed step-crack manner so as to increase the cross-sectional area of "cracked" material, such that the ligaments of uncleaved material between the array of cracks in any one preferred region or plane provides insufficient area to sustain the applied load and final failure will occur in a ductile mode unaffected by hydrogen. Fig. 4 illustrates some of the possible crack-linking and stopping processes.

Observations of fracture surfaces of the hydrogen-embrittled specimens in this current work revealed cleavage regions to account for more than 80 % of the total fracture surface. The cause of the predominance of cleavage must be related in some way to interactions between hydrogen, dislocations and the restriction of slip processes. It is a generally held view that hydrogen restricts dislocation movement and thereby inhibits general plasticity, minimizes plastic strain, and causes non-ductile fracture such as cleavage [13, 14]. Additionally, as mentioned earlier, hydrogen could reduce the cohesive force between atoms. Typical rosettes of cleaved regions are shown in Fig. 5. Cleavage originated at the centre of the rosettes, which usually contained incoherent inclusions, and fanned outwards. These cleavage rosettes were always surrounded by ductile areas (ligaments) which had failed by microvoid coalescence with the characteristics of a typical high strain-rate fracture process. Thus, it is deduced that hydrogen directly affected the formation and extension of the cleaved regions in this aspect of the embrittlement, and had little or no effect on the fracture of the remaining cross-sectional area by microvoid coalescence.

Fig. 6 shows the fracture model (Fig. 6b) developed to explain and visualize the formation of cracks and the subsequent fracture. The macrograph in Fig. 6a illustrates the appearance of the crack on the surface of a specimen (plan view), while the micrograph in Fig. 6c shows the cracks on the side surface of a specimen after polishing and etching with Nital, and the micrograph in Fig. 6d shows the appearance of a typical fracture surface.

The fundamental mechanisms which cause the absence or loss of ductility in metals and alloys has been the subject of a recent study of transition metal-based intermetallic compounds [15] in which the basic approach has been to correlate the state of embrittlement with the electronic state and hence to describe general mechanical behaviour in these compounds. This recent work has been directed to show that the number of unfilled d-state electrons contributes substantially to the mechanical properties of transition metals/alloys. The overlap of atomic d-orbitals creates a directional chemical bond (in contrast to s-orbitals which are isotropic); the relative amounts of the possible mixtures of s, p and d-orbital overlaps will determine the resultant bond directionality in alloys. The relevance of this approach to hydrogen embrittlement of steels is now explored.

In the hydrogen-charged steels examined here, there is direct experimental evidence for three distinct types of brittle fracture modes and one mode of ductile fracture; brittle fracture exists intergranularly, by

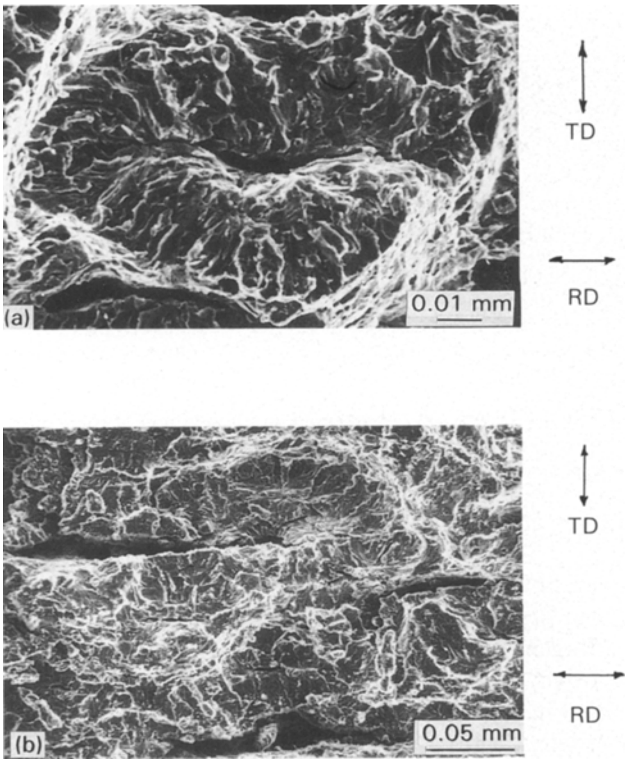

Figure 5 Cleavage on the fracture surface in the soft region; specimen charged for 140 h at 0.6 yield stress: (a) cleavage rosettes with a rim of ductile failure region (overload, high strain-rate fracture); (b) cleavage on the fracture surface associated with stack cracks (in the rolling plane); this provides the characteristic S-shape of the stack-cracking phenomenon which leads to linking of rolling-plane cracks.

cleavage and by incoherent particle/matrix separation, while the ductility is limited to final, overload, high strain-rate fracture.

With the exception of the cleavage fracture on planes approximately normal to the stressing direction, the other brittle fractures (either intergranular, interfacial or mixed interfacial/cleavage) can be considered to emanate from atomically sharp cracks where a tensile stress concentration ahead of the crack can increase to cause failure by atomic decohesion in a tensile mode, or increase by a lesser amount to cause failure by shear. (The source of stress, whether from internal molecular hydrogen pressure or from applied tensile loading, is unimportant.) For the case where brittle cracks are formed in the rolling plane and parallel to the tensile loading direction, the presence of the incoherent particles and the weak particle/matrix interfaces constitutes a crack and permits the accumulation of high-pressure molecular hydrogen gas which generates a tensile stress at the crack tip normal to and exceeding the applied tensile load. Rapid local crack extension from this source results in a lowering of the molecular gas pressure; after this primary crack extension the shear mode of cracking comes into operation leading to the characteristic S-shape shown in Fig. 3.

The units of transgranular cohesive failure, normal to the rolling direction and tensile loading direction, identified as cleavage rosettes (Fig. 5), invariably contain an inclusion, which is a molecular hydrogen-gas nucleus, at the centre of each rosette and the cleavage "petals" fan out from this central source. The force required to cause this decohesion must be related to the charge polarizability [16] on a cleavage plane so that the electron density between these two specific atomic planes will be lowered by "filling-in" adjacent undisturbed bonds that lie normal to the stress acting across the (cleavage) plane from either of the two sources.

The influence of hydrogen on the electronic structure of iron has been modelled by Eberhart *et al.* [16] and by Messmer and Briant [17] who showed that in a dilute hydrogen solution a hydrogenated iron lattice had a reduced directionality which led to an increase in iron–iron atom separation of 10 %. Thus reduction in directionality was attributed to the incorporation of more isotropic s-orbitals of hydrogen which would reduce the instantaneous shear strength and promote ductility. However, at high levels of hydrogen concentration (as in cathodic charging in this study) the overlap between the hydrogen antibonding orbital and the symmetrical orbital in the iron lattice causes intense charge polarization. In the regions of high hydrogen concentrations, i.e. in the core of rosettes, which are subject to radial and hoop stresses, the hydrogen atoms parallel to the strain directions will tend to separate and the situation prevails where the electron density is reduced by flow into iron–iron bonds perpendicular to the (cleavage) plane.

This study has identified the potential for local high concentrations of hydrogen in nominally dilute solutions of hydrogen in iron to cause brittle cleavage fracture. The threshold stress that has been identified and assigned a value of about 60 % of the (uncharged) yield strength of this steel [2] is explained by considering the elastic extension of atomic bonds in parts of the microstructure where microcracks exist; this allows locally high hydrogen concentrations to develop and create conditions for the formation of cleavage rosettes, which are the visual evidence of cleavage fracture.

4. Conclusions

1. The formation and propagation of rolling-plane cracks, the accumulation of which led to a feature known as stack cracking, was due to a combination of hydrogen pressure and lowering of the cohesive forces of various interfaces by hydrogen.
2. Lattice diffusion was not the only mode of hydrogen transport. Dislocation transport of hydrogen was also inferred.
3. The deflection of the initial planar (rolling plane) cracks to form characteristic S-shape cracks was attributed to stress distributions and interactions at crack tips and cleavage normal to the rolling plane.
4. Final failure was caused by the formation of discreet embrittled regions, consisting of cleavage fractures; the linking of these cleavage "rosettes" by microvoid coalescence (MVC) at high strain rates occurred at final fracture. Hydrogen embrittlement was the cause of the cleavage rosettes in the fracture plane while the MVC fracture of the uncleaved ligaments between rosettes was due to the applied stress.

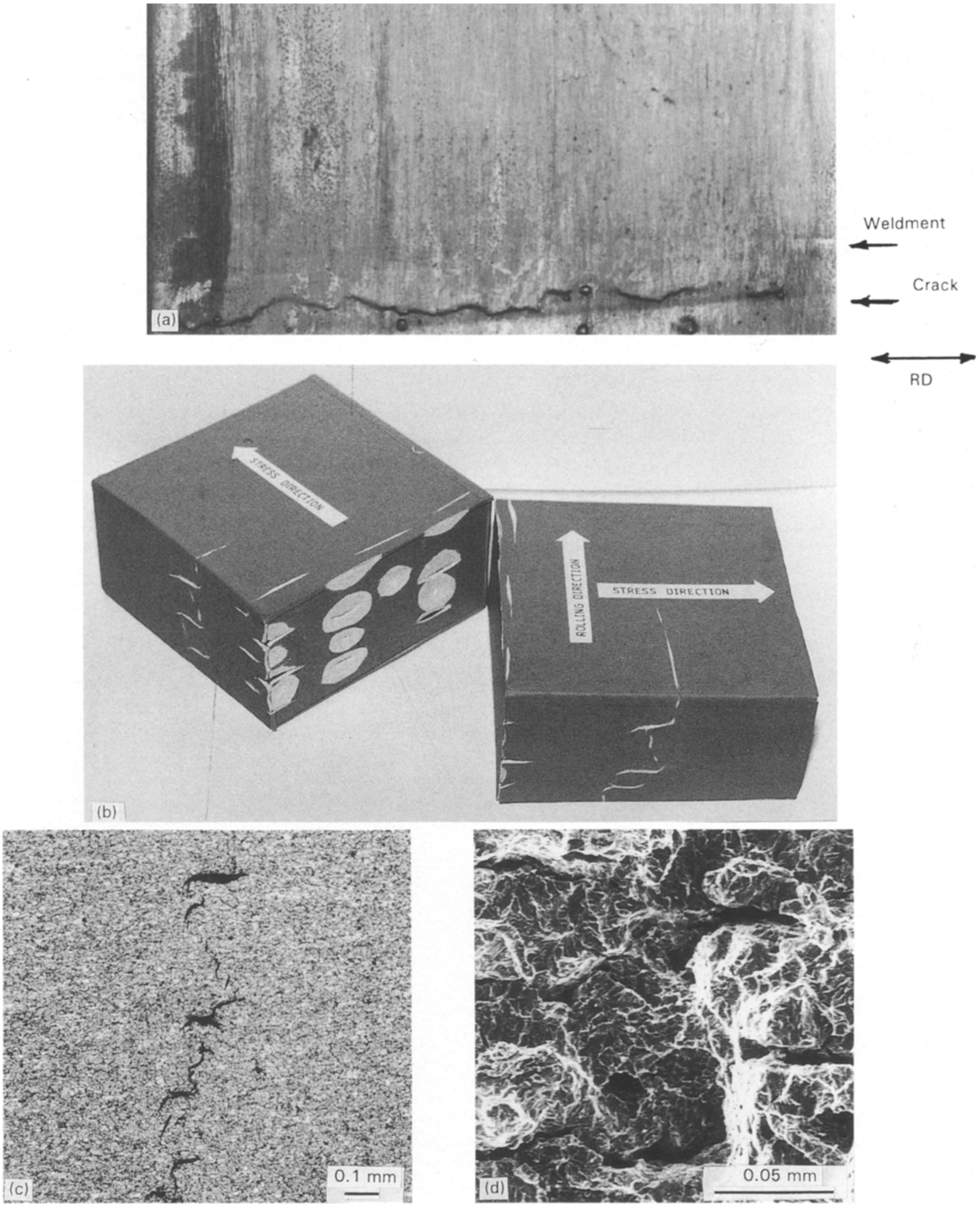

Figure 6 The model of stack cracking leading to fracture. (a) Plan view of a sheet specimen with an extended crack near a weldment. (b) Block model of the orientation relationships between the rolling direction, stressing direction, stack cracks and cleavage rosettes. (c) Stack cracks in the soft zone near a weldment (as Fig. 2b). (d) Cleavage rosettes and rolling-plane cracks (as in Fig. 5b).

5. An explanation of the fracture mode which occurs in the presence of locally high concentrations of hydrogen is given in terms of electronic state modifications to the steel.

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Received 22 June
and accepted 24 July 1992