NUMERICAL SIMULATIONS OF HYDROGEN / DISLOCATION INTERACTIONS AT A STRESS CORROSION CRACK TIP

J.-Ph. Chateau*, A. Paret*, D. Delafosse* and Th. Magnin*

We investigate the elementary mechanisms governing the stress corrosion cracking damage in f.c.c. alloys. Slow strain rate experiments show the important role of slip geometry on fracture crystallography and the relevant scale for modelling, i.e. the scale of the dislocations at the crack tip. A model is proposed that emphasizes the co-operative effects of corrosion and plasticity. A simulation of the hydrogen / dislocation interactions is carried out to assess key points of this model. Hydrogen diffuses in the hydrostatic stress fields of both dislocations and loaded crack. Its distribution around edge dislocations is shown to shield their elastic interactions, which leads to a softening effect. It allows higher dislocation densities in pile-ups, thus enhancing the fracture possibilities.

INTRODUCTION

The deleterious effect of a corrosive environment on the elongation to fracture of ductile f.c.c. metals and alloys is well established experimentally. The recent results obtained in stress corrosion cracking (SCC) last years [1] show that the main feature in fracture mechanisms is the interaction between deformation and corrosion (anodic dissolution and/or hydrogen). Based on the observations of the fracture surfaces, many SCC models have been proposed to describe and reproduce the fracture crystallography. They are based on elementary mechanisms in which local parameters play a key role (stress, diffusing species concentrations, material cohesion...). At the scale of these mechanisms, it is difficult to quantify experimentally the effects of these parameters and to validate the fracture mechanisms invoked. Thus, it is difficult to discriminate between the different models and to use them for fracture rate forecast. Numerical simulations can give us the evaluation of these local parameters. Simulations of the collective behaviour of dislocations at a crack tip have already shown to be successful in characterizing the brittle-to-ductile transition in b.c.c. metals [2]. We develop comparable simulations for f.c.c. materials, superposing diffusion of hydrogen from the crack tip. This could allow us to reach the characteristic space and time scales in the models and to test the influence of the macroscopic parameters on fracture rate.

* Ecole des Mines, centre SMS, 158 cours Fauriel, 42023 Saint-Etienne cedex 2, France

SCC OF AUSTENITIC STAINLESS STEELS

Slow strain rate tests ($\dot{\varepsilon} = 4.10^{-7} s^{-1}$) are performed on 316L type single crystals in $MgCl_2$ at 117°C and free potential. Using single crystals allows to control the slip conditions and to determine the fracture crystallography. Four orientations are tested: <100>, <110>, <111> and <149>. Detailed results, for both 316L and pure copper, are presented in [3]. In both cases, fracture initiates by localized dissolution in the slip planes. Two distinct situations appear:

(a) For easy glide orientations (Schmid factor > 0.4 and at least one slip plane containing the initial crack front: orientations <100>, <149> and <110> with a <001> propagation direction). Fracture is discontinuous at the scale of the micron and exhibits traces of a localized ductile rupture. For the <100> orientation, cracking is achieved by two symmetrical systems composed of three {111} facets. Two of them are disposed in a zigzag manner, making an average {110} type plane. The zigzag always involves a slip plane containing the macroscopic crack front. The third {111} facet lying next to the zigzag gives a local propagation direction close to <112>. This fracture pattern is also observed for <149> 'single slip' orientation and the <110> crystals when the macroscopic propagation direction is forced to be close to <100>.

(b) Other situations (low Schmid factor: <111> orientation - or no slip plane contains the macroscopic crack front: <110> orientation with a <110> propagation direction). Fracture exhibits a more brittle aspect, with smooth facets and sharp edges, even at the scale of the micron. Facets have an irregular size. Fracture seems to be due to decohesion along the low index planes (mainly {110}) subjected to the highest normal stress.

The first case corresponds to the type of fracture generally observed in polycrystals. Thus we will focus on the possibility of a zigzag fracture along {111} planes for modelling.

MODELLING OF THE CRYSTALLOGRAPHIC EFFECTS

The experimental results show the critical effect of both the tensile axis orientation and the propagation direction. Slip planes and their relative position with the crack impose both the type of fracture and the local propagation direction. This emphasizes the importance of the corrosion-plasticity interactions and shows that the relevant scale for modelling is the scale of the dislocations.

From previous experimental work, Magnin proposed a 'Corrosion Enhanced Plasticity Model' [4] which reproduces in two dimensions the 'easy glide' zigzag fracture. It is based on a localized softening effect due to the diffusion of vacancies and hydrogen produced by the electrochemical reactions at the crack tip. Slip planes activated at the very crack tip depassivate the metal and a localized anodic dissolution takes place. Figure 1 shows a sequence of a first simulation based on this model. The crack loaded in mode I emits edge dislocations on an inclined slip plane. During the first percents of a slow strain rate test, the sample is mechanically hardened. This hardening is modelled by the introduction of point obstacles in the slip plane, simulating to the forest.

Vacancies and hydrogen produced at the crack tip diffuse along the slip plane. In their diffusion zone, the resistance of the obstacles is neglected, thus simulating a

localized softening effect due to corrosion. Two zones co-exist: a softened zone (in grey), extending from the tip and a hardened zone (in black) further away. A mobile pile-up forms on the limit between the two zones, inducing stress concentration. The stress concentration applied at the crack tip (σ_a) is transferred at the head of the pile-up (σ_t). Considering that the presence of hydrogen segregating on the dislocations lowers the local k_{IC}, the stress concentration can be sufficient to form a crack's embryo by a kind of Stroh mechanism [5]. Hydrogen lowers the cohesion energy of the {111} planes and the normal stress can be sufficient to open the crack along the slip plane. Dislocations are emitted on the symmetrical plane to shield the new crack tip, leading to periodic changes of crack planes and a zigzag fracture along {111} micro-facets.

We want to simulate and quantify the mechanisms invoked in this model, particularly the softening effect. A possible mechanism is the diminution of the pair interactions between dislocations, generally attributed to hydrogen [6]. This can have a strong effect when we consider the formation of a pile-up in which the fracture possibility is related to the dislocation density. So, we first studied the effects of hydrogen alone.

NUMERICAL SIMULATION OF HYDROGEN / DISLOCATION INTERACTIONS

Simulation method.

Our simulations are based on the evolution of dislocations at a crack tip in two dimensions. At each time we can calculate the local total stress in the material and the forces exerted on each dislocation by using the expressions given in [7]. A simplified example of the forces on a dislocation in pure mode II can be written as:

$$f_i = \frac{K_{II} b_i}{\sqrt{2\pi x_i}} - \frac{\mu b_i^2}{4\pi(1-\nu)x_i} + \frac{\mu b_i}{2\pi(1-\nu)} \sum_{j \neq i} \frac{b_j}{(x_i - x_j)} \sqrt{\frac{x_j}{x_i}} \qquad (1)$$

where i and j are the indices of the dislocations, b their Burgers' vector and K_{II} the applied stress intensity factor. Three terms are considered: the force due to the loaded crack, the image force in the crack surfaces and the interactions with other dislocations. The dislocations are then moved in regards with the calculated force. Two contributions concerning hydrogen are added:

- the diffusion of hydrogen in the hydrostatic stress field of the crack and dislocations
- the evaluation of a 'hydrogen component' in the forces applied on the dislocations.

The diffusion of hydrogen is studied by superposing a grid adapted to the slip geometry at the crack tip (figure 2). At each time step ($\approx 10^{-6}$ s) the local hydrogen concentration in each box of the grid is computed by evaluating the fluxes through their external surfaces. Hydrogen diffuses in interstitial sites and induces lattice distortion. It thus interact with hydrostatic stress fields. This is taken into account in the evaluation of the local fluxes by adding a stress dependant term to the first Fick's law [8]:

$$\vec{J} = -D.\vec{\nabla}C + \frac{D.V^*}{RT} C.\vec{\nabla}\frac{\sigma_{kk}}{3} \qquad (2)$$

where C is the hydrogen volumic concentration, D its diffusion coefficient, V^* its partial molar volume and $\sigma_{kk}/3$ the hydrostatic stress. Hydrogen diffuses toward the zones in tension and relaxes the stresses. This stress relaxation is actualized at each time step and taken into account in the fluxes evaluation.

The hydrogen component in the forces on the dislocations is calculated by assuming that each diffusion box is equivalent to a dilatation line [6]. In other words, we consider that hydrogen is concentrated along lines placed at the centre of the boxes. The repartition of hydrogen assimilated to such a distribution of linear defects allows to calculate the force it induces on the dislocations, in the same way than for the dislocation interactions. The expressions we determined also take into account the presence of the free surfaces of the crack.

Hydrogen effect on pair interactions.

Using this method, figure 3a shows the distribution of hydrogen around two edge dislocations in 316L. The dislocations have the same Burgers' vector, the size of the boxes is 10b, the initial hydrogen content is H/M = 0.1. Hydrogen diffusion reaches a steady state. It segregates in the zones in tension 'below' the dislocations while the concentration in the zones in compression is decreased. This asymmetrical distribution of hydrogen from side to side of the slip plane of the dislocations and the resulting stress relaxation induces an additional shear stress in the slip plane. It is of opposite sign compared with the shear stress induced by the dislocations themselves: the segregation of hydrogen on the dislocations reduces the resolved shear stress. This causes a diminution of the interaction force between the two dislocations, as shown on figure 3b.

This mechanism leads to a softening effect due to hydrogen, that we are able to introduce in our simulations and to quantify. Note that there is a synergetic effect between diminution of the pair interactions and segregation. As dislocations have the same Burgers' vector, their stress fields add and the segregation of hydrogen increases when the distance between the dislocations decreases, which is facilitated by the diminution of their repulsion. This enhances the localization of hydrogen along the slip planes.

Stability of a pile-up in the presence of hydrogen.

A pile-up is formed at a crack tip by introducing a fixed dislocation in an activated slip plane. The fixed dislocation simulates the presence of a strong obstacle (such as a Lomer-Cottrell lock) at a distance of 0.5 μm from the crack. Under the load in mode I, the emitted dislocations pile-up on the obstacle and we determine their equilibrium positions. Placing this pile-up in a uniform hydrogen concentration, we determine the steady state repartition of hydrogen along the slip plane. Figure 4a shows the force exerted on each dislocation. The force on the source is negative: it is no more activated. The force on the first dislocation corresponds to the necessary resistance of the obstacle to form such a pile-up. In the absence of hydrogen, the pile-up would be in equilibrium: the force on the other dislocations is zero. In the presence of hydrogen, the force exerted on the dislocations at the head of the pile-up is positive and tends to draw them closer to the obstacle.

Taking these forces into account, the new equilibrium positions of the dislocations is determined step by step. The final state is presented in figure 4b. The pile-up is then in equilibrium in the presence of hydrogen. The dislocation density increased although the total force (dislocations + hydrogen) on the head dislocation is

the same. This shows that hydrogen promotes the formation of denser pile-ups for a given obstacle resistance. By relaxing the resolved shear stress, but not the total stress field of the dislocations, it enhances the formation of a crack's embryo at the head of the pile-up. This leads to a possible embrittlement mechanism due to hydrogen in ductile f.c.c. metals.

CONCLUSION

The experimental study of the fracture crystallography of austenitic stainless steels underlines the major role played by the interactions between corrosion and deformation and shows the good agreement with the Corrosion Enhanced Plasticity Model. Our first simulations allowed to obtain the first results concerning the modelling and the quantification of the hydrogen / dislocation interaction mechanisms.

The high hydrogen contents introduced for a significant softening effect can only be reached at the tip of a crack. This underlines the importance of anodic dissolution which creates a critical defect necessary to initiate the SCC mechanism. Our simulations show that hydrogen segregates on edge dislocations and decreases their pair interactions. The hydrogen / dislocation interactions localize the fracture mechanisms along the slip plane. The formation of denser pile-ups promotes local decohesion.

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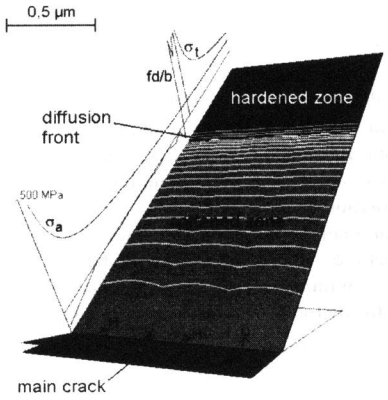

figure 1 : Numerical simulation of the mobile obstacle.

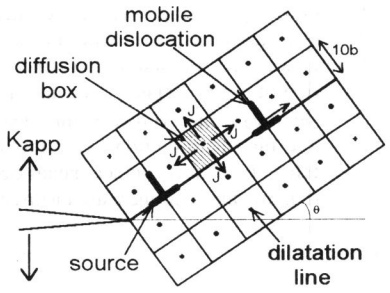

figure 2 : Scheme of the simulation method.

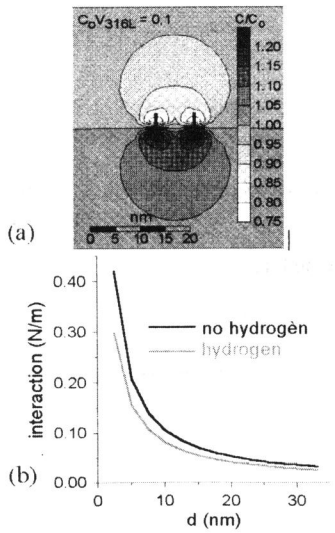

figure 3 : Segregation of hydrogen on two edge dislocations.

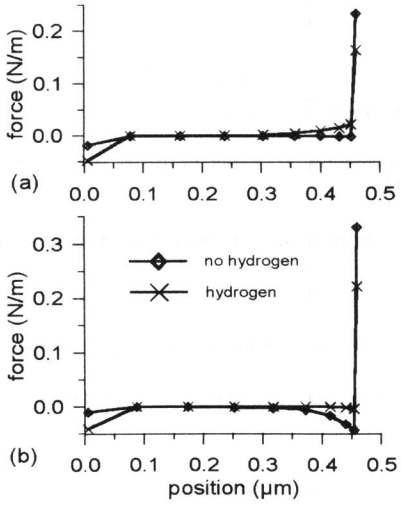

figure 4 : hydrogen effect on the stability of a pile-up.