

Simulation of crack growth due to hydrogen-induced stress-corrosion cracking on a high-strength steel FeE690T

R. Falkenberg^{1,2}

¹ Christian Albrechts University Kiel, Materials Mechanics, Kaiserstr. 2,
D-24143 Kiel, Germany

² GKSS Research Centre, Institute of Materials Research, Max-Planck-Str. 1,
D-21502 Geesthacht, Germany

^{1,2}rainer.falkenberg@gkss.de

Keywords: Transient hydrogen diffusion, embrittlement, fracture, simulation, R-curves

Abstract. In this investigation the model of a pre-cracked C(T) specimen loaded in a hydrogen enriched environment is simulated. An elastic-plastic material model using von Mises plasticity with pure isotropic hardening describes the deformation behaviour. The crack extension is modelled by means of the cohesive zone model in order to investigate the crack growth behaviour. The influence of hydrogen on the mechanical behaviour is twofold: It affects the local cohesive strength and the local yield stress and thus accelerates material damage. The diffusion behaviour is particularly influenced by the hydrostatic stress and the plastic deformation as well as by the strain rate. In this investigation the crack growth behaviour of a ferritic steel is simulated by fully coupled diffusion and mechanical finite-element-analyses in Abaqus by using User Subroutines, which allow the simultaneous solution of the system equations of the mechanical as well as of the diffusion model. Experimental investigations had preceded which yielded the needed material data as well as experimentally determined fracture resistance curves at different deformation rates (CTOD-R-curves). These experimental results serve for comparison in order to be able to adapt crack growth parameters to the simulation.

Introduction

Hydrogen induced failure is observed mainly in steels with a high yield strength. The herewith-connected extensive elastic lattice expansion and the high mobility of hydrogen in the lattice enhance the accumulation of hydrogen in energetically convenient locations, which emerge in the proximity of e.g. notches or crack tips [18]. The reason for the embrittling effect is not completely understood yet. There are voids, which serve as traps and catch hydrogen atoms and therefore have an influence on the hydrogen transport. Especially in the case of HISSC the hydrogen is produced directly in the process zone due to the cathodic sub process. The induced degradation of the respective structural material due to hydrogen can be described by a reduction of the crack resistance [22]. The soluted hydrogen, accumulating in the process zone near the crack tip, affects crack initiation and crack growth. The increased crack growth rate is compared to a hydrogen-free environment. Since the hydrogen diffusion is a transient process, the crack growth rate depends on the load and deformation rate [4]. Hence the phenomenological description combines two essential aspects. The effect of the local hydrogen concentration on mechanical quantities, such as critical local strain and stress respectively, have to be considered as well as the violation of the hydrogen transport due to mechanical impact, such as traps caused by plastic deformation [18]. In spite of numerous studies, the understanding of the mechanism of hydrogen embrittlement is still vague. Many models have been developed and each one is going

along by sets of experimental observations and strong personal views. It appears that there are many ways to model hydrogen related failure and that all of the aspects cannot be accounted for by a single mechanism. Some research groups considered the influence of hydrogen as a reduction of atomic cohesive strength (H.E.D.E., Hydrogen enhanced decohesion) [19], [22], [7], [19] others described the damage due to hydrogen as a result of a modification of the yield stress (H.E.L.P., Hydrogen enhanced localised plasticity) [2], [3], [16]. In this investigation, the material behaviour, especially of crack growth, of low-alloyed high strength steel named FeE690T due to HISSC is described based on these models and simulated in Abaqus. The realisation of this description is done numerically by fully coupled diffusion and stress analyses, which are able to model the hydrogen transport as well as crack growth. For the latter a cohesive zone model is used, which had been in use for the description of crack growth for a long time already [5]. Cohesive zone models with implemented hydrogen concentration dependence are already in use [14], [15], [23]. On the other hand transport equations are used for diffusion modelling, which pay attention to the corresponding mechanical field quantities. A parameter study shows the influence of variable system parameters on the diffusion behaviour as well as on the crack resistance behaviour. Based on existing experimental data from fracture mechanic tests performed in a hydrogen enriched environment the quality of the simulation can be judged. Furthermore the experimental data represents the only guide to conclude indirectly on load assumption, boundary and initial conditions, as a measurement of the hydrogen concentration gives only insufficient results.

Hydrogen kinetic and transport phenomena

The hydrogen concentration per unit volume in trapping sites, C_T , is expressed as

$$C_T = \theta_T \alpha N_T, \quad (1)$$

where α denotes the number of sites per trap, θ_T the occupancy of the trap sites and N_T the trap density measured in number of traps per unit lattice volume. The hydrogen concentration in interstitial lattice sites, C_L , can be phrased as

$$C_L = \theta_L \beta N_L, \quad (2)$$

where β denotes the number of interstitial lattice sites per solvent atom, θ_L the occupancy of the lattice sites and N_L the number of solvent lattice atoms per unit lattice volume. If the available number of trapping sites per unit volume, αN_T is small compared with the available interstitial lattice sites per unit volume, βN_L , then

$$N_L = \frac{N_A}{V_M}, \quad (3)$$

where $N_A = 6.0232 \times 10^{23}$ atoms per mole is Avogadro's number and V_M is the molar volume of the host lattice measured in units of volume per lattice mole [12]. It is assumed in this model that traps are isolated, i.e. that they do not form an extended network. Hence, hydrogen transport between trap sites could only be executed by lattice diffusion. Only one kind of trap, which is saturable and reversible, is considered. Considering a body with volume V and surface S , conservation of mass requires that the rate of change of total hydrogen inside V is equal to the flux through S [10]:

$$\frac{\partial}{\partial t} \int_V \{C_L + C_T\} dV + \int_S \mathbf{J} \cdot \mathbf{n} dS = 0, \quad (4)$$

where $\partial/\partial t$ is the partial derivative with respect to time, C_L is the hydrogen concentration in lattice sites, C_T is the hydrogen concentration in trap sites, \mathbf{n} is the outward-pointing unit normal vector and \mathbf{J} the hydrogen flux, which depends on the stress state as follows

$$\mathbf{J} = -D_L \nabla C_L + \frac{D_L C_L \bar{V}_H}{RT} \nabla \sigma_h, \quad (5)$$

where D_L is the concentration independent lattice diffusivity, \bar{V}_H the partial molar volume of hydrogen, i.e. $2.0 \times 10^{-6} m^3$ for α -iron at 293K, R the universal gas constant, T the absolute temperature, and $\sigma_h = \frac{1}{3}(\sigma_{xx} + \sigma_{yy} + \sigma_{zz})$ the hydrostatic stress: Substitution of eq. (5) in eq. (4) gives

$$\int_V \left\{ \frac{\partial C_L}{\partial t} + \frac{\partial C_T}{\partial t} \right\} dV + \int_S \left\{ \left(-D_L \nabla C_L + \frac{D_L C_L \bar{V}_H}{RT} \nabla \sigma_h \right) \cdot \mathbf{n} \right\} dS = 0. \quad (6)$$

Using the divergence theorem we find for an arbitrary volume V

$$\frac{\partial C_L}{\partial t} + \frac{\partial C_T}{\partial t} - \nabla \cdot (D_L \nabla C_L) + \nabla \cdot \left(\frac{D_L C_L \bar{V}_H}{RT} \nabla \sigma_h \right) = 0. \quad (7)$$

In the case of equilibrium the hydrogen concentration in trap sites can be expressed as a function of the hydrogen concentration in lattice sites

$$C_T = \frac{N_T}{1 + \frac{1}{K_T \theta_L}}, \quad (8)$$

where K_T is the trap equilibrium constant ($K_T = e^{-\Delta E_T/RT}$) and $\Delta E_T = -60 kJ/mol$ [11] is the trap binding energy. Contrary to the results of Kumnick and Johnson the number of trap sites is modeled in this investigation as $N_T = N_{T0} + N_{T100} \varepsilon_p^{0.7}$ [8]. Since the number of lattice sites is constant and the temperature will be kept constant, the partial derivative of the hydrogen concentration in trap sites with respect to time becomes

$$\frac{\partial C_T}{\partial t} = \frac{C_T(1 - \theta_T)}{C_L} \frac{\partial C_L}{\partial t} + \theta_T \frac{dN_T}{d\varepsilon_p} \frac{\partial \varepsilon_p}{\partial t}. \quad (9)$$

Finally, using eq. (9), eq. (7) becomes

$$\frac{C_L + C_T(1 - \theta_T)}{C_L} \frac{\partial C_L}{\partial t} - \nabla \cdot (D_L \nabla C_L) + \nabla \cdot \left(\frac{D_L C_L \bar{V}_H}{RT} \nabla \sigma_h \right) + \theta_T \frac{dN_T}{d\varepsilon_p} \frac{\partial \varepsilon_p}{\partial t} = 0. \quad (10)$$

More details on the variational and finite element formulation of eq. (10) can be found in [9].

The mechanical behaviour in the presence of hydrogen

The influence of material values on the mechanical quantities is considered through two aspects in this investigation: The reduction of local cohesive forces between the atoms (H.E.D.E.) and the modification of the local yield stress (H.E.L.P.). In the preceded section only the mechanical quantities on hydrogen diffusion have been addressed. However hydrogen induced stress corrosion cracking describes the embrittlement of a structure, therefore it is necessary to take a complete coupling of mechanical and diffusion behaviour into account.

The cohesive law in the presence of hydrogen. Local failure due to material separation as a result of

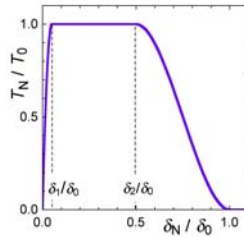


Figure 1: TSL, shape function

the influence of hydrogen is expressed in this investigation by a cohesive zone model. The separation law is described by [13]:

$$T_N(\delta_N) = T_0 f(\delta_N) \tag{11}$$

with the shape function, s. fig. 1

$$f(\delta_N) = \begin{cases} 2 \left(\frac{\delta_N}{\delta_1} \right) - \left(\frac{\delta_N}{\delta_1} \right)^2 & : \delta_N < \delta_1 \\ 1 & : \delta_1 < \delta_N < \delta_2 \\ 2 \left(\frac{\delta_N - \delta_2}{\delta_0 - \delta_2} \right)^3 - 3 \left(\frac{\delta_N - \delta_2}{\delta_0 - \delta_2} \right)^2 + 1 & : \delta_2 < \delta_N < \delta_0 \end{cases} \tag{12}$$

and the separation energy

$$\Gamma_0 = \int_0^{\delta_0} T(\delta_N) d\delta_N. \tag{13}$$

A reproduction of decohesion due to hydrogen influence is introduced by reducing the cohesive strength T_0 , which expresses the material damage [14]:

$$\frac{T_{0,hydr}}{T_0} = \left(1 - \alpha \frac{C_H}{C_0} \right) \tag{14}$$

Here α acts as a reduction factor, C_H is the local hydrogen concentration and C_0 corresponds to the concentration boundary condition. The material fails faster due to hydrogen influence, hereby one aspect of the hydrogen embrittlement could be expressed by a decreased separation energy Γ_0 .

The elastoplastic constitutive law in the presence of hydrogen. Besides the influence of hydrogen on the cohesive strength, which follows the H.E.D.E. theory, the influence on the local yield stress is considered. Some authors observed an increase [1] or a decrease respectively of the local yield stress due to the influence of hydrogen [21]. Regarding the effect of hydrogen on the dislocation behaviour in iron, Tabata & Birnbaum [20] determined the local flow stress of the material as a decreasing function of the amount of hydrogen in the material. By modifying the results of [17], the hydrogen effect on the local flow stress σ_{Y0} can be expressed by

$$\sigma_Y^H = \phi(c) \sigma_{Y0}(\epsilon_p), \tag{15}$$

where σ_Y^H is the local yield stress in the presence of hydrogen that decreases with increasing hydrogen concentration, $\phi(c)$ is a monotonically decreasing function of the local hydrogen concentration,

measured in atoms per solvent lattice atom, c is the sum of the occupancies θ_L and θ_T . σ_{Y0} is the local yield stress in the absence of hydrogen. A possible suggestion for $\phi(c)$ is a linear form:

$$\phi(c) = (\xi - 1)c + 1, \quad (16)$$

where $\xi < 1$ is a parameter. Hence, it is possible to model additionally to the material separation process the influence from hydrogen on the plasticity.

Results

A C(T) specimen with a width of $w = 40\text{mm}$ and a thickness of $t = 19\text{mm}$ and the material FeE690T (german designation 17MnCrMo3-3) was modelled. The mechanical properties of this material are $R_{p0.2} = 693\text{MPa}$ and $R_m = 820\text{MPa}$. The elastic-plastic material behaviour was modelled by von Mises plasticity with pure isotropic hardening. The cohesive parameters of the material tested in laboratory air are $T_0 = 2440\text{MPa}$ and $\delta_0 = 0.016\text{mm}$ with $\delta_1 = 0.05\delta_0$ and $\delta_2 = 0.65\delta_0$ which leads to a separation energy of $\Gamma_0 = 32\text{kJ/m}^2$ derived in [14]. Furthermore the diffusion coefficient of $D_L = 7.5 \cdot 10^{-9}\text{m}^2/\text{s}$, a trap density N_T with $N_{T0} = 0.61 \cdot 10^{20}\text{m}^{-3}$ and $N_{T100} = 3.31 \cdot 10^{21}\text{m}^{-3}$, was already determined in [8]. The hydrogen charge took place at the actual crack tip as a concentration boundary condition (Dirichlet boundary condition) in accordance with [18]

$$C_L = 1.989 \cdot 10^{26}\text{m}^{-3} \sqrt{p} \exp\left(-\frac{\Delta H_S}{RT}\right), \quad (17)$$

where p is the hydrogen pressure in atm and $\Delta H_S = 28.6\text{kJmol}^{-1}$ is the heat of solution [6]. An interchange of hydrogen across the model borders with the environment does not take place. Simultaneously a displacement boundary condition is applied which follows the respective deformation rates of $1\mu/h$, $10\mu/h$ and $100\mu/h$. A reduction factor of $\alpha = 0.67$ was chosen for the cohesive zone model and a softening parameter of $\xi = 0.8$ for the local yield stress. The last two values were determined by adjusting. The embedding of these numerous modifications, resulting from a particular diffusion equation, the cohesive element, the modification of the local yield stress and furthermore the calculation of additional values, was done in the finite element software Abaqus by User Subroutines. The crack extension Δa and the crack tip opening displacement δ_5 could be determined from these simulations, which had to be adjusted based on experimental results, see fig. 2. The experimental data has been derived in previous fracture mechanics SCC tests by subjecting these specimens at various constant load line displacement rates, while the specimens were in situ charged with hydrogen. The values of the CMOD (Crack Mouth Opening Displacement) were in each test directly measured by using specifically designed clip gauges which were attached to the specimens and converted in the δ_5 parameter [4]. Beyond this study the influence of the essential model parameters onto the behaviour of the system were investigated. In this case especially the diffusion coefficient D_L , the reduction factor α and the softening factor ξ were studied and showed a large impact on the crack growth behaviour. Furthermore it turned out that a time-dependent hydrogen application curve at the very first crack tip element had a significant influence on the crack initiation. The here fore investigated function was $C_L(t) = C_L(1 - \exp(-t/\tau))$, where τ acts as a time constant.

Closure

With this simulation a reproduction of the experimental results for different deformation rates is reached with minor deviation. The influence of the parameters D_L , α and ξ on the R-curves could be studied. This model works with only a few parameters: During adaption of the cohesive model

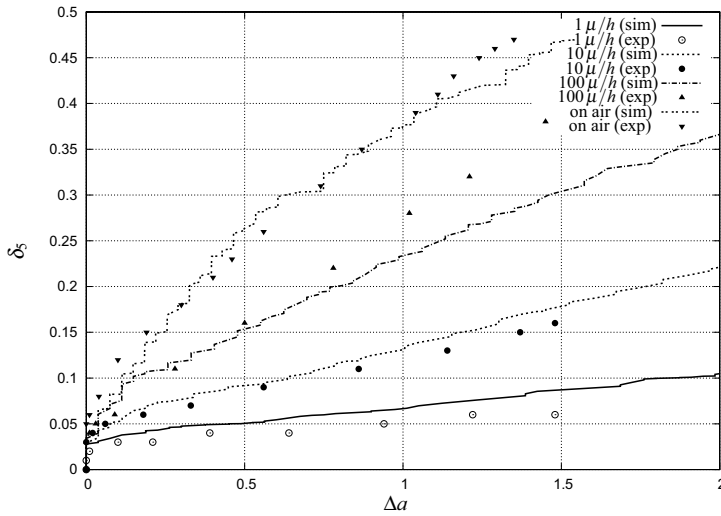


Figure 2: R-curves under different deformation rates

only two parameters need to be adjusted. The modelling of hydrogen embrittlement is done via two parameters, α and ξ , too. This first approach opens a convenient basis, which is further under development. For a future work it is imaginable to investigate the transferability of the results of the simulation to other specimen geometries in order to check if the results can already be used for a prediction of the crack growth behaviour. HISCC is a phenomenon, which is determined by many complex physical-chemical processes interfering with each other. Therefore the derivation of one general model for HISCC is difficult to obtain. The vast challenge of comprehensive data acquisition of these physical-chemical processes exceeds the time frame of one single investigation, as shown in [6]. It is rather results from decades of research. Hence there is still an immense fuzziness in describing processes in the context of absorption, adsorption and diffusion. A simulation, as done in this investigation, has therefore always the task of illuminating those scatter areas and determine physically possible intervals of hydrogen concentration as well as material parameters such as diffusivity and solubility. The coupling of different values complicates a clear statement. E.g. the immense range of variation of diffusion coefficients for one dedicated material could most likely result from an inadequate consideration of surface and solubility processes, which are crucial in permeation experiments. A particular challenge is moreover the quest for an eligible formulation of the coupling between the diffusion process and the mechanical behaviour. Further research on this topic is needed in order to establish a more appropriate modelling of HISCC.

Acknowledgements

The author would like to thank the DFG (Deutsche Forschungsgemeinschaft) for its support on the project Br 521/14-1.

References

- [1] A. M. Adair and R. E. Hook. The appearance and return of a hydrogen yield point in iron. *Acta Metallurgica*, 10(8):741–745, 1962.
- [2] H. K. Birnbaum, I. M. Robertson, P. Sofronis, and D. Teter. Mechanisms of hydrogen related fracture. a review. In *2nd International Conference on Corrosion Deformation Interaction*, pages 172–195. The Institute of Materials, London, 1997.
- [3] H. K. Birnbaum and P. Sofronis. Hydrogen-enhanced localized plasticity—a mechanism for hydrogen-related fracture. *Materials Science and Engineering A*, 176(1-2):191–202, 1994.
- [4] W. Dietzel, M. Pfuff, and G. G. Juilfs. Hydrogen permeation in plastically deformed steel membranes. *Materials Science*, 42(1):78–84, 2006.
- [5] M. Elices, G. V. Guinea, J. Gómez, and J. Planas. The cohesive zone model: advantages, limitations and challenges. *Engineering Fracture Mechanics*, 69(2):137–163, 2002.
- [6] J. P. Hirth. Effects of hydrogen on the properties of iron and steel. *Metallurgical Transactions A*, 11(6):861–890, 1980.
- [7] H. H. Johnson, J. G. Morlet, and A. R. Troiano. Hydrogen, crack initiation, and delayed failure in steel. *Trans. Met. Soc. AIME*, 212:528–536, 1958.
- [8] G. G. Juilfs. *Das Diffusionsverhalten von Wasserstoff in einem niedriglegierten Stahl unter Berücksichtigung des Verformungsgrades und der Deckschichtbildung in alkalischen Medien*. PhD thesis, 2001.
- [9] A. H. M. Krom, A. Bakker, and R. W. J. Koers. Modelling hydrogen-induced cracking in steel using a coupled diffusion stress finite element analysis. *International Journal of Pressure Vessels and Piping*, 72(2):139–147, 1997.
- [10] A. H. M. Krom, H. J. Maier, R. W. J. Koers, and A. Bakker. The effect of strain rate on hydrogen distribution in round tensile specimens. *Materials Science and Engineering A*, 271(1-2):22–30, 1999.
- [11] A. J. Kumnick and H. H. Johnson. Deep trapping states for hydrogen in deformed iron. *Acta Metallurgica*, 28(1):33–39, 1980.
- [12] J. Lufano, P. Sofronis, and D. Symons. Hydrogen transport and large strain elastoplasticity near a notch in alloy x-750. *Engineering Fracture Mechanics*, 59(6):827–845, 1998.
- [13] I. Scheider and W. Brocks. Simulation of cup-cone fracture using the cohesive model. *Engineering Fracture Mechanics*, 70(14):1943–1961, 2003.
- [14] I. Scheider, M. Pfuff, and W. Dietzel. Simulation of hydrogen assisted stress corrosion cracking using the cohesive model. *Engineering Fracture Mechanics*, In Press, Corrected Proof.
- [15] S. Serebrinsky, E. A. Carter, and M. Ortiz. A quantum-mechanically informed continuum model of hydrogen embrittlement. *Journal of the Mechanics and Physics of Solids*, 52(10):2403–2430, 2004.
- [16] P. Sofronis and H. K. Birnbaum. Mechanics of the hydrogen-dislocation-impurity interactions—i. increasing shear modulus. *Journal of the Mechanics and Physics of Solids*, 43(1):49–90, 1995.

- [17] P. Sofronis, Y. Liang, and N. Aravas. Hydrogen induced shear localization of the plastic flow in metals and alloys. *European Journal of Mechanics - A/Solids*, 20(6):857–872, 2001.
- [18] P. Sofronis and R. M. Mcmeeking. Numerical analysis of hydrogen transport near a blunting crack tip. *Journal of the Mechanics and Physics of Solids*, 37(3):317–350, 1989.
- [19] E. A. Steigerwald, F. W. Schaller, and A. R. Troiano. The role of stress in hydrogen induced delayed failure. *Trans. Met. Soc. AIME*, 218:832–841, 1960.
- [20] T. Tabata and H. K. Birnbaum. Direct observations of the effect of hydrogen on the behavior of dislocations in iron. *Scripta Metallurgica*, 17(7):947–950, 1983.
- [21] J. Tien, A. W. Thompson, I. M. Bernstein, and R. J. Richards. Hydrogen transport by dislocations. *Metallurgical and Materials Transactions A*, 7(5):821–829, 1976.
- [22] A. R. Troiano. The role of hydrogen and other interstitials in the mechanical behaviour of metals. *Trans. ASM.*, 52:54–80, 1960.
- [23] Y. Xu and J. L. Bassani. A steady-state model for diffusion-controlled fracture. *Materials Science and Engineering: A*, 260(1-2):48–54, 1999.