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Hydrogen Enhanced Fatigue Crack Growth Rates in a Ferritic Fe-3wt%Si Alloy

Antonio Alvaro[a], Di Wan[b], Vigdis Olden[a], Afrooz Barnoush[b]

[a]SINTEF Industry, 7456 Trondheim, Norway
[b]Department of Mechanical and Industrial Engineering, Norwegian University of Science and Technology, 7491 Trondheim, Norway

Abstract

It is well known that the presence of hydrogen in ferrous materials promotes both static fracture and affect the material fatigue crack growth rates. The latter is often referred to as Hydrogen Enhanced Fatigue Crack Growth Rate (HE-FCGR) which defines the reduction of crack growth resistance of the material under cyclic stresses when hydrogen is present. When it comes to the determination of the life of components exposed to hydrogen it is therefore of paramount importance to establish such hydrogen induced variation in crack speed in the material in order to avoid unexpected catastrophic failures. In this study the fatigue crack growth rate was determined for a Fe-3wt%Si alloy. Compact tension specimens were used to determine the Paris regime of the fatigue crack growth rate curve of the material. Two environmental conditions were investigated: laboratory air and in-situ electrochemically charged hydrogen. Different mechanical conditions, in terms of load ratio (R=0.1 and R=0.5) and test frequency (f=0.1 Hz, 1 Hz and 10 Hz), were used under electrochemically charged hydrogen conditions. The results show that compared to the specimens tested in air, there is a clear detrimental effect of H for the specimens tested in hydrogen, in terms of accelerated crack growth. The strength of the impact of hydrogen in enhancing the fatigue crack growth rates of the Fe-3wt%Si alloy clearly depends on the test conditions. Fractographic investigations were also used to unveil the mechanisms involved in the process leading to accelerate crack growth in presence of hydrogen.

Keywords: Fatigue Crack Growth, Hydrgen Embrittlement, Steel

Introduction

It is well established that the presence of atomic H in ferrous materials under cyclic stresses affects the fatigue behavior of metallic materials and steels in particular. This is an important issue both for the new and long-standing applications in energy fields which operate in aggressive working environments, to avoid catastrophic consequences for environment, industrial economy and personnel health. Therefore the development of the knowledge of the mechanical behavior of metallic materials when directly in contact with hydrogen is strongly connected to a quantitative evaluation of the hydrogen influence on mechanical properties of steels which is necessary for a suitable proper design of hydrogen-contact prone components and structure. On the other hand, such information is of paramount importance when it comes to mainteinance strategies and decision about inspection periods: platforms, umbilicals, risers, flowlines and subsea pipelines are continuously subjected to oscillatory environmental loads and, at the same time, to hydrogen uptake from cathodic protection or H_2S containing fluids (Colombo et al. (2016), Suresh et al. (1983)). Degradation by H under such conditions would manifest as reduced resistance to fatigue crack growth Failures during operation in which hydrogen has played an important role, however, are difficult to be discerned from the hydrogen-free failures, due to the volatile nature of the H atom. Worldwide, it is generally considered that over 80% of all service failures can be related, to different degree, to mechanical fatigue (Ritchie (1999)). By considering that offshore structures are often designed by use of defect-tolerant principles, where knowledge of defect size and fatigue crack growth rate is used to determine the remaining life of a component, it is therefore evident as the knowledge of the H effect on the latter becomes vital for both reliable design and life extension of existing oil and gas fields.

In structural components containing micro cracks and small defects, the main portion of the total cycles to failure is associated with crack growth which happens within three stages: stage I (short cracks), stage II (long cracks) and stage III (final fracture). While Stage III is related to unstable crack growth and can be considered of the least importance for the fatigue life, both stage I and II can be more or less affected by the presence of H. The strength of the impact of hydrogen on the crack growth properties of a material depends on several factors which are often not mutually independent: the material system, the frequency, the load ratio, temperature, pressure and or potential level, just to mention the most influential. It is therefore important to understand the nature and quantify the strength of hydrogen induced acceleration of fatigue crack growth rate in order to assess the eligibility of a material in applications where hydrogen uptake is happening under fatigue life design.

Experimental procedures

Material

The material used in this study is Fe-3wt%Si with simple ferritic structure. The detailed chemical composition is shown in Table 1:

Table 1: Chemical composition of the Fe3%Si alloy used for the test

Element	**C**	**Si**	**Mn**	**P**	**S**	**Cr**	**Ni**	**Mo**
wt.%	0.018	3.000	0.055	0.008	0.003	0.010	0.006	0.003
Element	**Cu**	**Al**	**Ti**	**Nb**	**V**	**B**	**Zr**	**Fe**
wt.%	0.013	0.015	0.001	0.002	0.001	0.0002	0.0010	balance

In order to obtain the most equiaxed grain as possible, the raw plates were obtained by rolling follow by 10% cold rolling plus annealing at 800 ℃ for three times with the last annealing temperature of 1050 ℃ before the final straightening. The average grain size was of about 300 µm, as shown in Figure 1a).
The yield strength, the ultimate tensile strength and the HV10 and were 508 MPa ,555 MPa and 175 respectively

Fatigue crack growth tests

Fatigue crack growth rate testing was performed on Compact Tension (CT) specimens (in conformity to the dimensions recommended in ASTM E647 standard, see Figure 1b), which were cut by electron discharge machining (EDM) from the raw material.

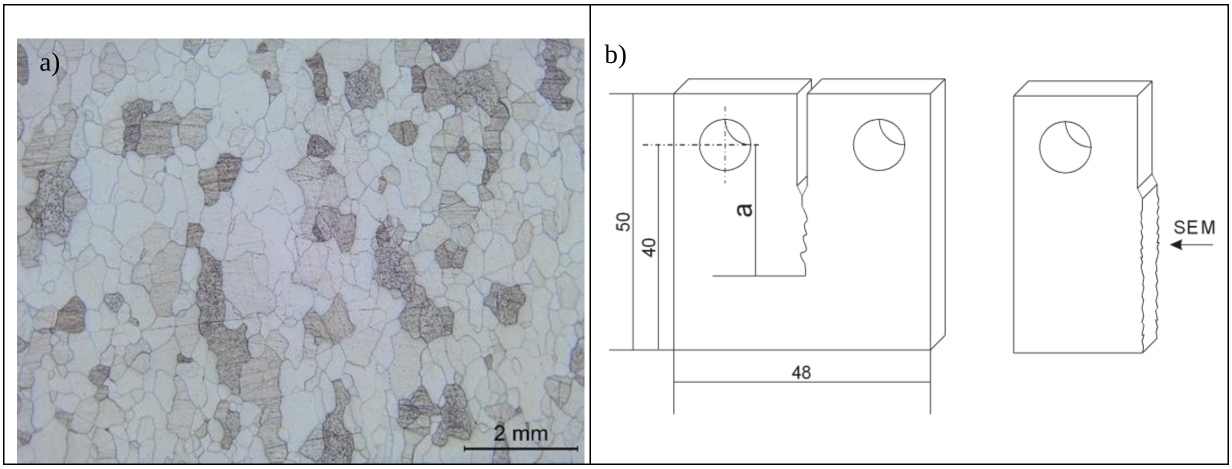


Figure 1: a) Microstructure of the investigated material; b) Geometry and dimensions of the CT specimens tested.

The FCGR tests were carried out both in air and in-situ electrochemical charging conditions at room temperature. The specimens were in-situ cathodically charged in a 0.1M Na_2SO_4 electrolyte with a constant potential of -1400 mV_{SCE}. Multimeters were used during the whole test in order to check the values and keep the circuit run as designed.

Before tests, a pre-crack was produced by fatigue according to the procedure described in XXXXX.Alvaro et al. (2015). First, a lower bound threshold stress intensity factor, i.e. ΔK_{th}, ranging from 10-12 MPa $m^{0.5}$ was used to initiate the crack from the notch. Then a reduction in the ΔK value of 5% was adopted stepwise until the target *K*-value was reached and the crack growth stabilized. Typically, these pre-cracks were in the range of 2~4 mm after approximately 200 000 cycles for this material. It should be noted that when the pre-crack procedure stops, the crack front is not necessarily a straight line: the start/stop reference crack length are estimated though an average of nine points equally distributed across the specimen width. To get the crack further growing, a 5% increase in the ΔK from the last step of pre-cracking procedure was used in order to minimize the effect of the plastic zone from the pre-cracking procedure present ahead of the crack. An alternate current-potential drop (AC-PD) crack growth rate measure box was used during the test to record the FCGR behavior. At the end of the test the specimens were cracked in liquid nitrogen and the the *da/dN* vs ΔK curves were obtained from start/stop surface measurements.

Test in air were performed at 10 Hz and at two load ratios, i.e. 0.1 and 0.5. In-situ cathodically (14.2 gr/l Na_2SO_4) charged FCG test were carried out in a built-on-purpose test rig (see Figure 2) at three different frequencies, 0.1 Hz, 1 Hz and 10 Hz and two load ratios, R=0.1 and R=0.5.

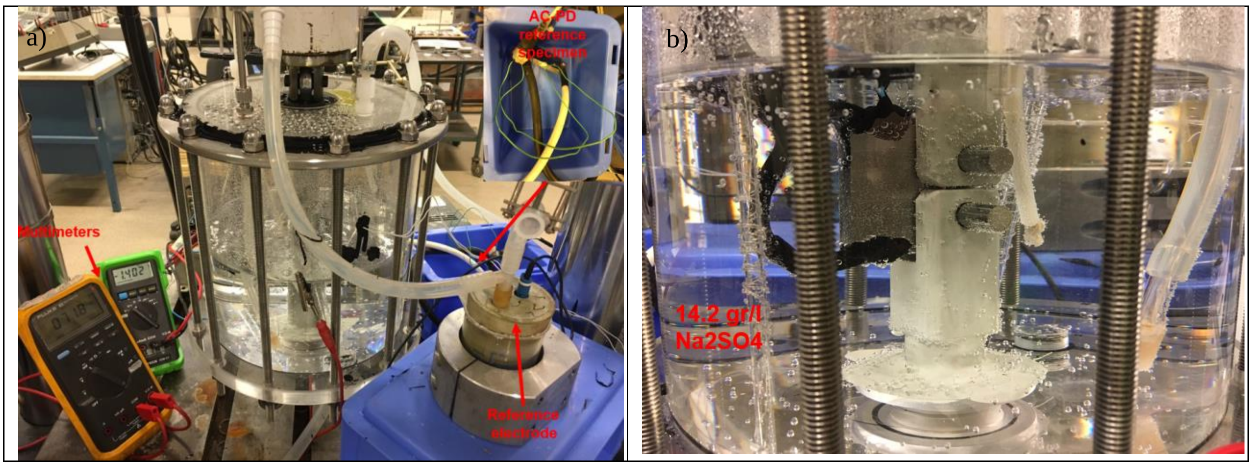


Figure 2: Set-up of the test rig for in-situ electrochemically charged fatigue crack growth rate test.; b) Close up of the chamber with the specimen.

Characterization

SEM fractographic characterization was performed on all the post-mortem specimens. The FEG Quanta 650 environmental SEM (Thermo Fisher Scientific Inc., USA) was operated at 20 kV acceleration voltage with an aperture size of 50 µm. Measurements on the fracture surfaces were done by using the in-built software on the cross section of the specimen as shown in Figure 1b).

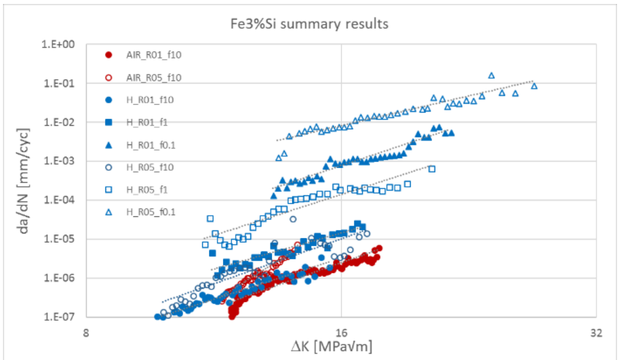
Results

Fatigue crack growth rates

Fatigue crack growth rate test were performed in air (10 Hz) and in-situ electrochemical charging at two load ratios and three frequencies. Stage II fatigue crack growth domain was characterized thorough the well-known Paris law (Paris et al. (1963)):

$$ {da}/{dN} = C \cdot \Delta K^m \quad (1) $$

where *a* is the crack length and *N* is the number of the cycles, giving *da/dN* the discrete crack extension/growth per cycle. *C* and *m* on the right-hand side of the equation are constants that depend on the material and the testing conditions, while *ΔK* is the range of the stress intensity factor experienced by the cracked material during the fatigue cycles.
The resulting FCG rate curves are shown in Figure 3a) together with a table summarizing the Paris constants *C* and *m* and the acceleration factor recorded at *ΔK* values of 16 MPa√m. Results clearly show a strong acceleration



b)

Environment	*R*	*f* [Hz]	*C*	*m*	$\frac{(da/dN)_H}{(da/dN)_{air}}$
Air	*0.1*	*10*	*2E-16*	*6.67*	*1*
Air	*0.5*	*10*	*3E-17*	*7.89*	*1*
H	*0.1*	*10*	*2E-14*	*6.81*	*1*
H	*0.1*	*1*	*5E-13*	*6.17*	*8.4*
H	*0.1*	*0.1*	*1E-12*	*7.03*	*5E+02*
H	*0.5*	*10*	*3E-15*	*7.90*	*0.8*
H	*0.5*	*1*	*5E-13*	*6597*	*28*
H	*0.5*	*0.1*	*7E-9*	*5.05*	*1E+03*

Figure 3: a) Fatigue crack growth rate curves of Fe3%Si material in air and under different load ration and frequency levels. Red curves indicate test in air and blue curves indicates in-situ electrochemically charged tests. Full markers indicate test conducted at R=0.1 while empty markers the ones at R=0.5. Round, square and triangle markers indictes test carried out at 10 Hz, 1 Hz and 0.1 Hz, respectively. b) Table reporting the Paris law constants *C* and *m* obtained at different conditions. The acceleration factor for *ΔK=16 MPa√m* is also reported.

The specimens tested under H-charging conditions generally exhibited a higher FCGR than that the one tested in Air. The strength of the hydrogen induced acceleration featured a strong dependency with respect to the test frequency: the lower the frequency, more pronounced is the crack propagation rate enhancement. The Paris' law parameters obtained from the curves and summarized in Figure 3b): the presence of hydrogen induce an acceleration in crack growth of about 1000 times while the low variation in *m* values indicated rather a shifting of the curves, i.e. the effect of hydrogen is independent on the *ΔK* level when focusing on the Paris' domain. The effect of the load ratio is also consistent: exept for the test performed at 10 Hz for which little to none hydrogen influence is registered, all the test performed at R=0.5 feature a stronger crack growth rate acceleration than the ones recorded at R=0.1.

Fractography

All the tested specimens have been subjected to fractography analysis; three different main fracture morphologies have been identified: Transgranular (TG), Intergranular (IG) and "quasi-cleavage" (QC) ones. Images representative of the three zones are presented in Figure 4.

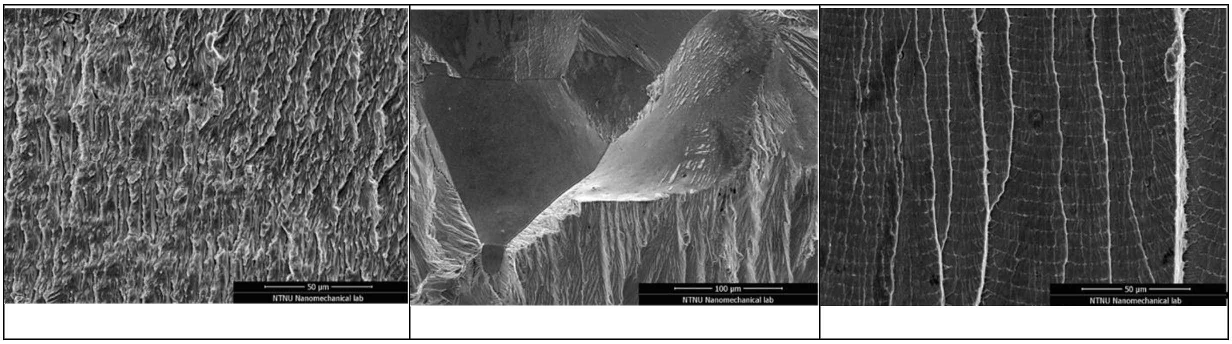


Figure 4: Representative images of the different fractographic morphologies: a) Transgranular, b) Intergranular, c) Quasi cleavage features.

Transgranular zones are characterized by the traditional ductile striation whose main directions are mostly aligned with the global crack growth testing direction (see **Error! Reference source not found.**a)). Intergranular features reveal grain-boundary-like surfaces (see Figure 4b)) while "QC" features are normally showing facets and smooth area between striations, sometimes accompanied by river-marks (Figure 4c)). The analysis of the striation morphology indicated a different striation appearance in relation to the different fracture modes and depending on the environmental conditions. In general, striations from TG zones are denser and deeper (Figure 5a)) indicating a strong plastic development in front of the crack during the load cycle and the crack advance while the ones featured in the "QC" fracture zones are sparser (Figure 5c)) , and with much smaller crests. In IG zones, no striations were observed (Figure 5b))

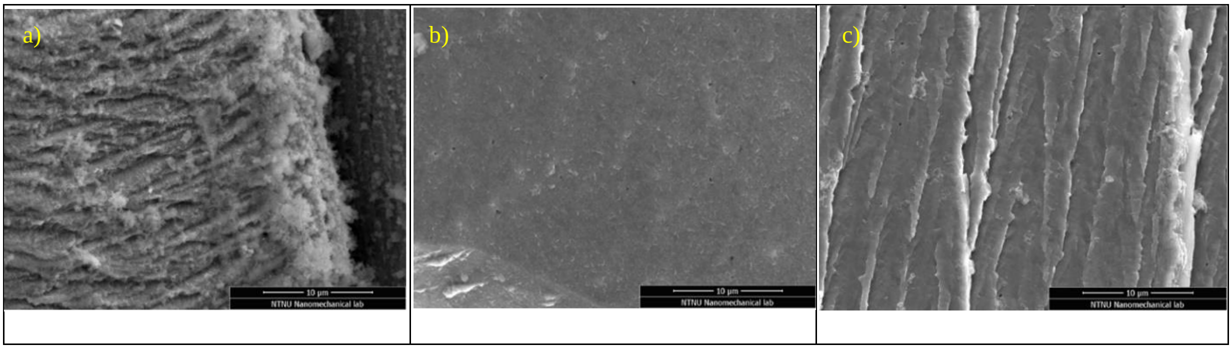


Figure 5: Representative striation morphology detectable in the three aforementioned fractographic features: a) Transgranular, b) Intergranular, c) Quasi cleavage. All the images are from the in-situ electrochemically charged specimen tested at 0.1 Hz and R=0.5. Crack growth is from up to down.

In order to have a quantitative idea of the impact of the frequency on the mechanisms inherent to FCG rate acceleration in relation to the different fracture morphologies observed on the specimen surfaces, a statistical distribution of the presence of the latter for the in-situ electrochemically charged specimens in the 13.5-17,5 ΔK range for the specimens tested at $R=0.5$ is presented in Figure 6a). It is evident that development and the exclusive ration of the fracture modes are strongly dependent on both test frequency and the ΔK level. When the ΔK level increased, the fracture mode changed from TG to "QC", while IG type fracture generally took only a small fraction independently on the testing conditions. The same trend could be observed when as the load frequency is lowered. In addition, the distance between fatigue striations was measured and verified against the global *da/dN* data from the same FCG rate tests. It was found that the distance between striations (representative of the local crack extension per cycle *da/dN*) in the "QC" fracture areas is always several times higher than the globally measured *da/dN* data, while that in the TG fracture zones is approximately the same or a slightly smaller than the measured global *da/dN* data, independently to the test frequency. The measurement was subjected to the same ΔK range and the result is shown in

Error! Reference source not found.. It is inferred that the H changed the FCG mode from TG type to “QC” facets by a different amount depending on the charging conditions and the load. IG type fracture is also not so common in H-charged specimens, suggesting GB is not the most critical material feature with respect to hydrogen embrittlement in this material.

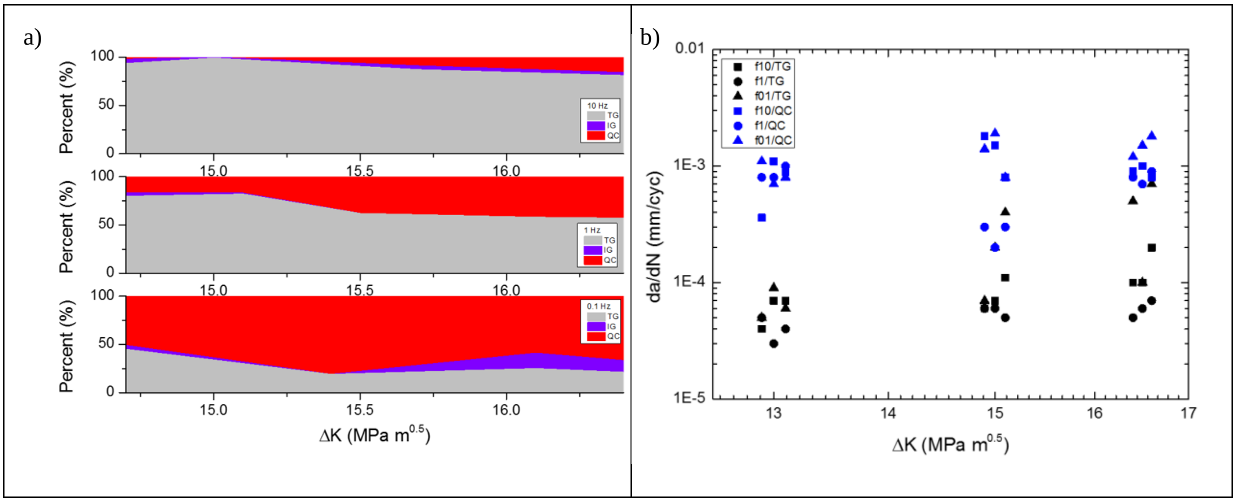


Figure 6: Fracture modes statistical distribution vs. ΔK level for 13.5 MPa√m < ΔK < 17.5 MPa√m; b) Measured striation distance vs. ΔK level in different fracture modes.

Discussion

It has been observed in other works that hydrogen can change the fatigue fracture mode from ductile striations to brittle cleavage facets. proposed that H can be trapped to the intensive stress field ahead of the crack tip, cyclically arresting the sharp cleavage crack advance by blunting it. This would result in the cleavage fracture surface shown as facets. Vehoff (Vehoff et al. (1986)) observed similar structures in Fe-2.6%Si single crystals under gaseous H_2 environment. In order to explain the sharp cleavage fracture structure, they postulated that crack grows stably in a stepwise micro-cleavage manner as well as through enhanced dislocations emission from the crack tip. Similar features were also observed in Fe-Si alloys Takahashi (Takahashi et al. (2010)) and in low carbon steels Nishikawa (Nishikawa et al. (2011)), Matsuoka (Matsuoka et al. (2011)) and Yamabe (Yamabe et al. (2016)) (just to mention few) and Fe-Si alloys with FCG tests in H_2 gaseous environments.

Another specific feature of the fracture surface investigation is the low presence of IG features. Hajilou (Hajilou et al. (2017)) did microcantilever bending test on the same Fe-3wt.%Si alloy with in-situ electrochemical H-charging, and showed that although grain boundaries will promote HE, the crack propagation is highly dependent on the relative position of the stress concentration with respect to the GB. According to McMahon (McMahon (2001)), the H-assisted IG fracture in steels depends strongly on the impurities segregated to the GBs. However, most H-assisted failure events relates to hydrogen diffusing into the region at high hydrostatic stress. In the present study, the materials were purposely heat-treated so that little to no grain boundary segregation could be expected. Therefore, the GBs are “pure” enough to prevent from hydrogen-assisted IG fracture and most fracture will happen according to the local stress or stress intensity distribution.

Ogawa (Ogawa et al. (2017) and Ogawa et al. (2018)) investigated the HA-FCG behaviour of pure iron under different H2 pressure levels through multi-scale characterization. By comparing their fracture features with the ones obtained in this work, a strong similarity is evident. In their study it is explained that H, transported by mobile dislocations (Robertson (1999)) based on the HELP mechanism, produced the formation of dislocation tangles which, in turn, determine a local enhancement of the H concentration, leading to H-assisted cracking. Their characterization results showed most of the “QC” fractures were on the 100 cleavage planes of the α-Fe phase. However, it is important to note that while they worked on pure Fe while the material under investigation in this work is a Fe3%Si and, as already demonstrated by Nakasato (Nakasato et al. (1978)), the addition of Si tends to strongly favour cleavage cracking. Therefore, the mechanism behind the FCG rate acceleration may be different and the difference in acceleration factor (about 30X in pure Fe and up to 1000 time in Fe-3%Si here presented) seems to strongly support this thesis. The strong dependence with respect to the frequency as well as the load ratio seem to point toward a more stress-controlled based mechanisms rather than a strain-controlled based mechanism as in the case of pure Fe. Deeper understanding is needed to completely unveil the mechanisms. The authors are currently working toward the comparison and the qualitative determination of the dislocation structures between the Fe-3%Si H-free and the H-charged specimens in order to clearly determine the alloy HA-FCG mechanism.

Conclusions and further work

The effect of hydrogen on fatigue crack growth behaviour in a Fe-3wt.%Si alloy under in-situ electro-chemical H charging condition was studied. The fracture modes distribution was statistically summarized along the crack growth path and the effect of testing frequency on the fracture mode transition was discussed. Some conclusions could be drawn as follows:

- The HE effect was revealed by the in-situ cathodic charging method on the FCGR test, and the H-charging enhanced the FCGR by up to 1000 times compared to a test in air, depending on the test frequency and the load ratio.
- The Paris' law can describe the FCGR behavior of a structure well at continuum level but cannot precisely describe the local behavior at microstructure level, especially when special environmental conditions apply.
- Post-mortem fractography clearly showed a shift in crack growth mechanisms: the FCG mode of Fe-3wt.%Si changed from TG to "QC" type; IG type fracture is not common in this material strongly indicating how the GB in this material is not the preferred path for crack propagation.
- The test results show a time-dependent nature of the H-deformation interaction: a lower frequency in H environment will lead to more severe degradation of the FCG process. This points toward a stress-based HA-FACG rate mechanism.

Diverse charging methods and advanced characterization methods could also be applied to study the deeper mechanism of the H-assisted cracking phenomena. This would be the outlook of the next work.

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