Scripta Materialia 151 (2018) 24–27

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Regular Article

Hydrogen embrittlement effect observed by in-situ hydrogen plasma charging on a ferritic alloy

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ARTICLE INFO

Article history:
Received 30 January 2018
Received in revised form 24 March 2018
Accepted 25 March 2018
Available online xxxx

Keywords:
Hydrogen embrittlement
In-situ test
Fe-3wt%Si
Plasma
Crack

ABSTRACT

To study the hydrogen embrittlement (HE) effect, a novel in-situ slow strain rate tensile test together with in-situ hydrogen (H) charging by H-plasma was conducted in an environmental scanning electron microscope (ESEM). The introduction of H-plasma gave a reduction in tensile elongation by about 5% in comparison with a reference test in vacuum. Fractographic observation clearly showed the difference in the resulted features on the fracture surfaces. Electron backscatter diffraction (EBSD) was conducted to elucidate the characteristics of the cracks. Such investigations can help to refresh the existing knowledge in HE study.

Hydrogen embrittlement (HE) is a well-acknowledged phenomenon in metals. It draws much scientists' and engineers' attention because it can lead to catastrophic failure of an industrial structure. Researchers use different methods to charge a test piece with hydrogen (H) and study the behavior of the material during or after charging. A common method is to use H_2 gas as the environment, for example Vehoff [1,2] used gaseous H to study the H effect on the fracture behavior of ferritic steels. More examples can be found in Ref. [3–12]. Furthermore, electrochemical cathodic charging is also commonly used, for example Wang et al. [13] used cathodic charging with an in-situ tensile test to study the effect of H on mobile dislocations, and the H concentrations were also determined. Other examples can be found in Ref. [14–22] as well. There is also a smart method for special materials, for example, Deng et al. [23,24] and Rogne et al. [25] used water vapor to charge Fe-Al intermetallic alloys since the H_2O molecule can react with Al, producing alumina and H to invade the matrix.

Due to the high diffusivity of H in body-centered cubic (BCC) lattice [13], it is always critical to have H pre-charging on the test piece because H can diffuse out even during a short transferring procedure. Therefore, in-situ H-charging is favored for the HE study of BCC structures.

Currently, in-situ charging of ferritic steel is either done in high pressure hydrogen gas or under electrochemical control by cathodic H-charging. This makes the high-resolution observation of the sample during deformation challenging.

Environmental scanning electron microscopy (ESEM) combined with miniaturized tensile stages provides a unique possibility to test the effect of H on mechanical deformation, however the limited maximum pressure inside the chamber and restrictions on the possible gases at that pressure in the chamber from the manufacturer hinders the HE tests inside the ESEM. At low pressures, the fugacity of H is not enough to have adequate physisorption and dissociation of the H_2 molecules on the surface to observe HE within a reasonable time scale. Converting H_2 gas to H plasma before letting it enter the ESEM chamber will increase the H fugacity in the chamber and provide possibilities to observe the HE processes inside the ESEM.

Narita [26] used H glow charging to study the embrittlement effect in a Fe-Si system with a tensile test on single crystals and concluded that this method is capable of charging the samples with enough H to reveal an embrittlement effect without severe surface damage. Kimura and Birnbaum [27] observed that that H-plasma charging can cause a softening effect in the flow stress of pure iron samples. A recent work from Malitckii et al. [28] used H-rich plasma at elevated temperatures to charge two kinds of steels and successfully revealed the HE effect on them.

In this paper, we show that the introduction of low pressure H-plasma in ESEM chamber can provide a possibility to study the HE of a ferritic alloy with in-situ mechanical testing. The limitation in in-situ investigation has been discussed. Moreover, ex-situ characterization was carried out to study the details of the tested specimen. Such investigations can help us refresh the knowledge in HE.

Simple ferritic Fe-3wt%Si alloy with the composition shown in Table 1 was used in this study. The as-received material has a coarse

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https://doi.org/10.1016/j.scriptamat.2018.03.038
1359-6462/

Table 1
Chemical composition of the investigated material.

Elem.	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
Elem.	Cu	Al	Ti	Nb	V	B	Zr	Fe
wt.%	0.013	0.015	0.001	0.002	0.001	0.0002	0.0010	Bal.

grain size of 300 μm. This simple microstructure makes the alloy a perfect model material to establish the methodology (e.g. [1,2,29–33]).Tensile specimens were cut from the raw material by electrical discharge machining (EDM) to a dog-bone shape with the gauge geometry of 20 mm × 6 mm × 2 mm. Grinding to #4000 emery paper followed by 3 μm and 1 μm diamond paste polishing plus final electropolishing was adopted to make sure the tested surfaces were flat and smooth without residual deformation. The tensile/compression module from Kammrath & Weiss GmbH (Germany) was used for mechanical testing inside a Quanta 650 ESEM (Thermo Fisher Scientific Inc., USA). The engineering strain rate was chosen to be 10^{-5} s^{-1} in order to provide enough time for H adsorption and diffusion. After fracture, the half-specimens were taken out for further characterization. For H-charging, an Evactron Model 25 Zephyr Plasma Cleaner (XEI Scientific, USA) was used with gaseous H_2 as a process gas from a hydrogen generator. It should be noted that remote plasma was applied in this cleaner. With this method, the interaction between plasma and material occurs at a location that is remote from the plasma afterglow. In other words, only active plasma would participate in the interactions with the material, such that the specimen surface can prevent from heating, contaminating and damaging effects.

Fig. 1 shows the experimental setup of the SEM chamber. The plasma cleaner was connected to the SEM chamber via a flange, and the hydrogen generator was connected to the working gas inlet of the plasma cleaner. The tensile specimen was installed into the tensile/compression module connecting to an external controlling unit via another flange. With this setup, in-situ mechanical testing with in-situ H-charging can be realized. In-situ observation by normal SEM mode is limited and we are discussing the possibilities for imaging at low pressure H-plasma with the ESEM manufacturer. To the authors' knowledge, no reports on imaging by high voltage electrons with H-plasma presenting have been found. According to the communications with the company, in-situ imaging with high-voltage in H environment is potentially unsafe. Since H is extremely flammable, there is a likelihood of arcing in the chamber from the detectors and this is extremely likely to cause the H to ignite. For this reason, the investigations in this study could only be done when the chamber is completely evacuated to high-vacuum state.

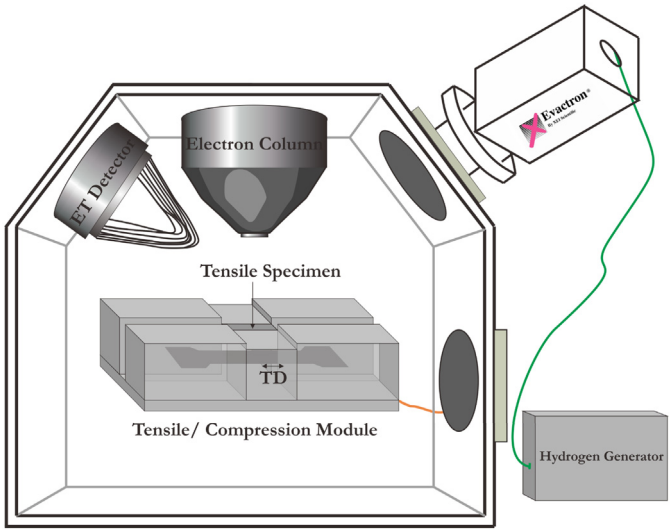

Fig. 1. Experimental setup of the SEM chamber (TD: tensile direction).

The stress-strain curves for the tensile tests are shown in Fig. 2. It should be noted that the stress is calculated by dividing the load over the cross-section area in the gauge range, which was re-measured after preparation since grinding and polishing could change the geometry of the specimen. The tests were denoted as Vac case (tested in vacuum) and H case (tested in H-plasma), respectively. The H test was done by stopping the test at the ultimate tensile strength (UTS) range from the Vac test for one hour to give more time for H uptake into the lattice under enhanced solubility effect of the tensile stress as well as exposure of the oxide free surface. From the curves, the two specimens had similar behaviors in the elastic range and behaved similarly up to the UTS range with a slight hardening effect in the H case. But the H-charged specimen had a reduction in the elongation to fracture by about 5% and the crack propagation procedure was much faster and more unstable. It is difficult to define the crack initiation from the stress-strain curves, but it can be seen qualitatively that the crack was growing in a more stable manner in Vac than in H case, which showed a sudden failure. A supplementary video was attached to this work, showing the relatively stable crack propagation procedure of the Vac case. This procedure corresponds to the part highlighted by blue dash-lines in Fig. 2. For the H case, due to technical limitations, no imaging is possible during the plasma charging and pictures can only be taken after interrupting the plasma and evacuating the chamber from H. We are currently discussing with the ESEM manufacturer for modification to try to make this imaging process possible.

Fig. 3 shows the fracture surfaces of the specimens after final fracture in Vac and in H case, respectively. The reduction in area from calculation is 61.4% for Vac and 50.1% for H case, respectively. The fracture surfaces showed generally transgranular type, regardless of H-plasma charging. The ductile feature (dimples) is highlighted by yellow dash-lines. A large area fraction (about 75% of the whole fracture surface) of dimples can be found in the Vac case, which indicates the crack growth procedure was in a more ductile manner when H was absent. While in the H case, most of the area shows brittle transgranular cleavage-like features with only a limited area fraction (about 10% of the whole fracture surface) of ductile features. Besides, many secondary cracks are observed on the fracture surface in the H case while these are not seen in the Vac case. Fig. 3b and c show the magnified dimple features in Vac case, with b showing inclusion spots in dimples and c showing pure dimple structure. This indicates a ductile behavior of this specimen in Vac, regardless of the presence of small inclusions. Fig. 3e and f reveal detailed fracture surface of the H case showing cleavage-like features.

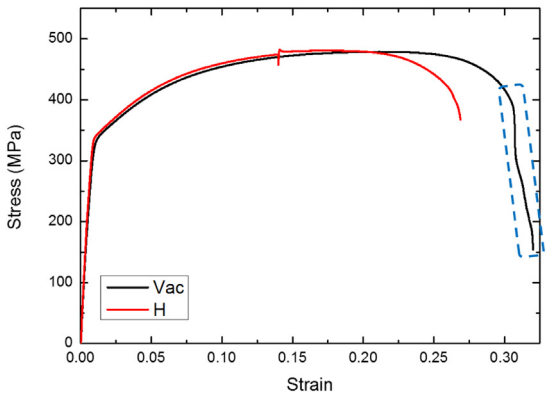

Fig. 2. Stress-strain curves of the investigated specimens (the highlighted part corresponds to the crack propagation procedure, which is shown by a video in supplementary files).