Experimental report

07/09/2022

Proposal: 1-05-105 **Council:** 4/2021

Title: Mapping localized hydrogen by in-situ neutron imaging

Research area: Engineering

This proposal is a resubmission of 1-05-41

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Samples: Steel specimen (4 identical)

Instrument	Requested days	Allocated days	From	To
NEXT	3	3	10/09/2021	13/09/2021

Abstract:

The main aim of this proposal is to investigate whether neutron transmission imaging can be used to detect spatially resolved concentrations of hydrogen (H) in steel under conditions approaching those relevant for advanced engineering applications. The purpose behind the investigation is the industrially highly relevant problem of hydrogen embrittlement (HE) where even minute amounts of H in steel will have a detrimental effect on the mechanical properties. Insights from fracture mechanics tests and subsequent SEM imaging of the fracture surfaces have led to further support for the hypothesis that while the specimen is under load the intense stress state in front of a crack tip promotes preferential diffusion of H into the crack tip region. The local concentration of H in front of the crack tip has never been measured and average values reported from TDS cannot be assumed to properly represent it. The proposed experiment aims to investigate if, and under what conditions, the locally increased H concentration can be detected.

Report for experiment 1-05-105: Mapping localized hydrogen by in-situ neutron imaging

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Experimental date: 10/09/21 to 13/09/21

Background and scientific motivation

The motivation for the experiment is the problem of hydrogen embrittlement (HE) where even minute amounts of H can make a steel brittle. Typically, thermal desorption spectroscopy (TDS) is used to investigate the H content in a steel specimen. However, values of H concentration from TDS only accounts for the average concentration in a specimen, whereas fracture and failure are local processes. In addition, TDS cannot be performed on specimens that are still under mechanical load. An intense mechanical stress state will promote H diffusion and accumulation in a region [1, 2] and may promote local embrittlement there. In some recent studies H have been quantified in ferrous metals using neutron transmission imaging [3, 4], and in Zr-alloys [5] but these studies are not representative of the operational conditions for high strength steel products.

Experimental procedure

Chemo-mechanical testing was performed on a single-edge-notch bend specimen and subsequently imaged with time series of neutron radiographs under in-situ conditions at NeXT. The specimens had been machined, pre-cracked and pre-deformed under normal operating conditions, see Fig 1(c). The material was a modern quenched and tempered high strength steel containing lath martensite with a yield strength σ_Y = 1348 MPa and Young's modulus E = 205 GPa [2]. The steel specimens had the following dimensions: height W = 16 mm, distance between roller supports s = 64 mm and thickness B = 8 mm, and the initial crack length a = 8 mm, which are in accordance with ASTM E180 [6]. The roller supports were made from a non-conductive ceramic material. Before loading, an electrode was attached to the specimen such that so-called cathodic polarization could be applied via a current. The electrochemical (EC) pre-charging was controlled by a galvanostat as a power source with a constant current of 2.5 mA. The specimen is submerged in an electrolyte made of water with a 3.5 wt. sodium chloride (NaCl) solution and a partially submerged platinum mesh is used (to increase the adsorption of hydrogen into the specimen) to close the circuit. A portable load frame was used to apply tensile loading. The environment container was a cylindrical cup made from Teflon (chemically inert and non-conductive)

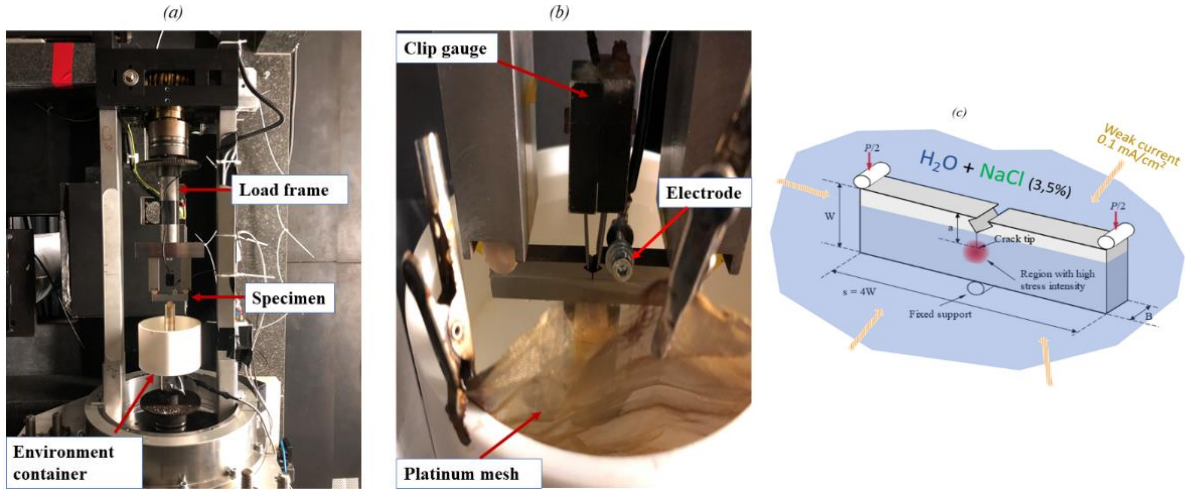

Figure 1: Experimental setup at NeXT showing in (a) the mobile load frame with specimen mounted in fixtures and the environment container in the down configuration for imaging, and in (b) a closeup of the specimen during chemo-mechanical charging to induce HE. A sketch of the electrochemical environment and the fracture mechanics specimen is shown in (c).

that could contain the specimen and the EC setup. The cup is mounted on the lower load transferring piston and can slide up/down without removing the load or leakage of the electrolyte. The purpose of this was to be able to charge the specimen with H during mechanical load and then, without removing the load, lowering the cup to expose the specimen to the neutron beam. Before imaging we also dried the specimen using compressed air to remove all the water from the surface and the crack. The experimental setup mounted at NeXT can be seen in Fig 1(a, b).

Experimental results

We hade 72 hours of beam access during which we tested several specimens. The first couple of specimens where mainly used for establishing the reference conditions without charging and testing the experimental procedure. Normally the samples are coated with a polyethylene lacquer to restrict the H-ingress to only a smaller exposed region close to the expected crack path. While evaluating the radiographs of the initial samples we noticed that tiny droplets of the lacquer had contaminated our region of interest and showed up as clear disturbances in the attenuation field. Therefore, on subsequent specimens we adjusted the lacquered region and adjusted the current of the EC charging such that the average current per exposed area remained the same as in all our previous experiments [2]. Without knowing we had now made a small, but significant, change to the experimental setup (see below). We will now report on the last three specimens where we learnt the most and got some useful data. We will refer to these as specimens A, B and C.

Specimen A

The specimen was charged for 4 hours while simultaneously increasing the mechanical load up to 10kN. Thereafter the displacement was kept constant. The environment was removed and once the drying was completed (in 3-4 minutes) we started taking radiographs using a 10mm pinhole and 10s exposures. Image acquisition proceeded over 10 hours to ensure good statistics and increase our chances of imaging any possible redistribution of H. Initially we thought the experiment was a failure since did not get the expected signal and the force steadily decreased. However, it turned out that we had managed to capture a related phenomena called delayed hydrogen cracking whereby H accumulated to the crack tip, the crack advances a distance and arrests when it has passed through the embrittled region, then the procedure is repeated. We did not manage to image and H during this experiment, only the effects of its redistribution (no other plausible fracture mechanics explanation exists for the observed behavior). Data and examples of the analysis from specimen A can be seen in Fig 2. A journal paper is under preparation based on this data set coupled with state-of-the-art modelling of crack propagation, embrittlement, and H diffusion from a moving stress field.

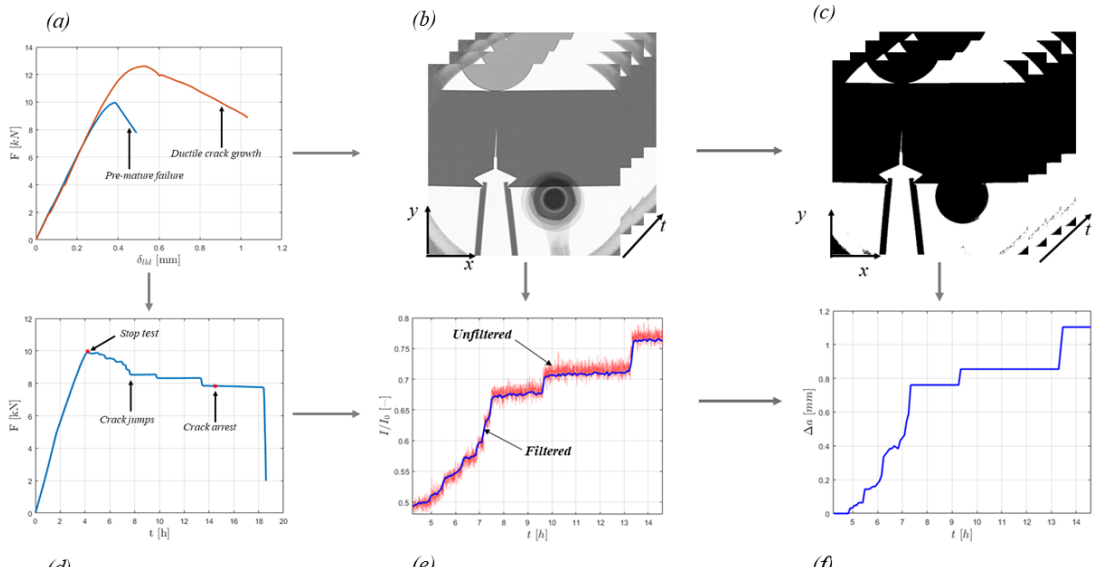

Figure 2: Data from specimen A showing (a) failure at 10kN (blue line) compared to reference sample (red), (b) imaging crack propagation over 10 hours, (c) image thresholding to automatically determine crack tip position, (d) force vs time showing load drop events correlated with (e) transmission profile of the crack propagation and (f) the obtained crack growth increments from the thresholding analysis.

Specimen B

The specimen was charged for 4 hours while simultaneously increasing the mechanical load up to 8kN to avoid the delayed cracking observed in A. Thereafter the displacement was kept constant, and the environment was removed. Imaging started after 3-4 minutes using a 30mm pinhole and 5s exposures to get a much larger neutron flux but also a better temporal resolution. Image acquisition proceeded for 2.5 hours. As shown in Fig 3(a, b) we could now see a region of larger attenuation in front of the crack tip. Subsequent data analysis revealed a signal that is both spatially and temporally resolved, see Fig 3(d, e). The signal decays to the surrounding levels (i.e. attenuation for steel without H) within about 30 minutes. The additional attenuation is consistent with a local H concentration of 25-35 ppm.wt. Obviously, we initially deemed these results as a success.

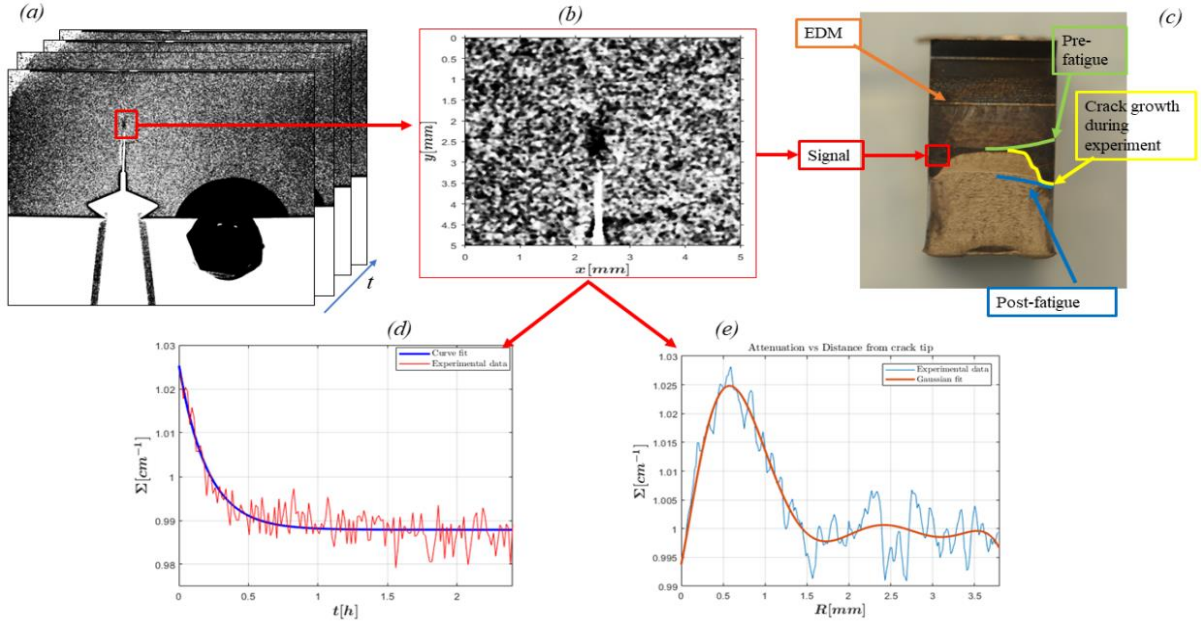

Figure 3: Data from specimen B showing (a) illustration of time series images, (b) closeup of increased attenuation signal in front of crack tip, (c) fractographic analysis of the anomalous crack propagation, (d) time decay of increased attenuation signal, and (e) spatial extent of increased attenuation in front of crack tip.

However, once we got the specimen back form ILL, we performed x-ray CT (not shown) and standard fractography, see Fig 3(c), on the specimen and noticed that the crack had grown unexpectedly on the sides (where the stress is much lower than in the middle). We have now confirmed that this was due to the modified lacquer (see above). We are still confident that the signal we saw is due to H but the presence of the side cracks complicates the analysis and adds too much uncertainty for a clear presentation of our results to the research community.

Specimen C

The same experiment as B but the drying procedure failed. Instead of getting more data we learnt what water in the crack region looks like (nothing like the signal in Fig 3) and got useful, albeit unintended, feedback on how to improve the drying procedure.

Conclusions

The experimental protocol combining neutron imaging with chemo-mechanical testing seems poised to be able to finally provide quantitative data of the local H concentration under operationally relevant conditions for steel. The uncertainty of the current results can almost surely be removed by minor, and well understood/investigated, changes to the protocol to attain results with a high level of confidence.

References

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