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Assessing the Susceptibility of Existing Pipelines to Hydrogen Embrittlement

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Abstract With fossil fuels being phased out and growing global interest in a hydrogen economy, there is demand for re-purposing existing pipelines for transportation of hydrogen gas. However, hydrogen embrittlement (HE) can limit pipeline steel's performance. In this study, the effect of hydrogen on the mechanical properties of an X60 base metal (polygonal ferrite/pearlite) and its girth weld (acicular ferrite/pearlite) was measured with a novel slow strain rate tensile (SSRT) test in which hollow pipe-like specimens were internally pressurised with nitrogen and hydrogen gas from 0 to 100 bars. Results showed that exposure to H_2 gas at 100 bars reduced the ductility of the base metal by up to 40% and the weld metal by 14%. Reduction in cross-sectional area (%RA) reduced by up to 28% in the base metal and 11% in the weld metal. Fracture surface analysis showed micro-void coalescence as well as quasi-cleavage fracture characteristic of HE. Susceptibility to HE was also observed in the form of secondary longitudinal and internal transverse cracks.

Keywords Hydrogen embrittlement · Pipeline steel · In situ testing · Fractography

Introduction

Hydrogen has never been as relevant globally as it is today since it is a cost-effective and energy-efficient way of transporting and storing energy sustainably. Implementation of hydrogen as an energy carrier is now being pushed for on a global scale like in the European Green Deal [1]. A cost and material-efficient way of transporting hydrogen would be to use existing natural gas pipelines. Since natural gas is expected to be phased out in the coming decades, these pipelines would become available to transport hydrogen instead. However, current knowledge of the effect of hydrogen

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gas on pipeline steels and especially their weldments is lacking. This research studies the effects of hydrogen gas on the mechanical properties of X60 pipeline steel and its girth weld and assesses the HE susceptibility of both.

Through several mechanisms, gaseous hydrogen that is absorbed into a steel will embrittle it, reducing its ductility and possibly its strength. This is called hydrogen embrittlement (HE). Because hydrogen diffuses towards regions of high stress triaxiality like notches and areas in front of crack tips, they can cause extra deterioration of mechanical properties leading to a loss of strength [2]. The fracture mechanisms related to HE can be fundamentally plastic in nature, even though the behaviour that is observed in the material on a larger scale appears to be brittle [3–5]. Standardised tests to assess HE susceptibility of metals are listed in, for example, ASTM F1624 [6], which describes an incremental step loading technique to assess a HE threshold stress, and ASTM G142, which describes a slow strain rate tensile (SSRT) test to assess HE susceptibility [7]. Both tests are performed in situ in a hydrogen environment to eliminate any desorption of hydrogen during the test. SSRT or constant load tests are preferred since a high strain rate can limit HE, because sufficient hydrogen diffusion towards regions of high stress triaxiality is required in order to cause HE [8]. Many studies in the literature test their materials in situ in an electrochemical charging environment. This environment is often not representative of gaseous hydrogen environments because the hydrogen concentrations are often much higher in an electrochemical charging environment, as shown by Zhao et al. [9].

To assess the susceptibility of pipelines that are used to transport gaseous hydrogen, it is necessary to adequately represent the hydrogen environment that would be in place during operation. Therefore, in situ SSRT testing in a high-pressure gaseous hydrogen environment is desired. In this research, a novel test setup was designed that allows for assessment of both the base and weld metals to HE. The design removes the need of a pressure vessel around the sample to charge it with hydrogen, thereby providing an easy and cost-effective way of testing metals in a hydrogen environment and greatly reducing the amount of gas required. Fracture surface investigation by microscopy (SEM) was used in this study to determine HE mechanisms.

Materials

This research considers an X60 pipeline steel, including a girth weld that was made according to industrial pipeline welding procedures. The alloy contents of both steels are listed in Table 1. The start\stop regions of the girth weld as well as the longitudinal weld regions were both excluded from the research. The base metal as shown in Figure 1a consists of mainly polygonal ferrite with small regions of pearlite with an average grain size on the order of 10 µm. The weld metal, shown in Figure 1b, has a microstructure consisting of mainly acicular ferrite and pearlite with grain boundary Widmanstätten ferrite. The acicular ferrite phase makes up the bulk of the weld microstructure with an average grain size on the order of 1 µm. The base metal

Table 1 Alloy contents the base and weld metals in weight %, where rest is Fe

Element	C	Mn	Si	Cr	Nb	Al	P	S	Rest
Base metal	0.06	1.66	0.26	0.06	0.04	0.04	<0.01	<0.01	97.88
Weld metal	0.07	1.45	0.58	0.05	0.01	0.01	0.01	0.01	97.81

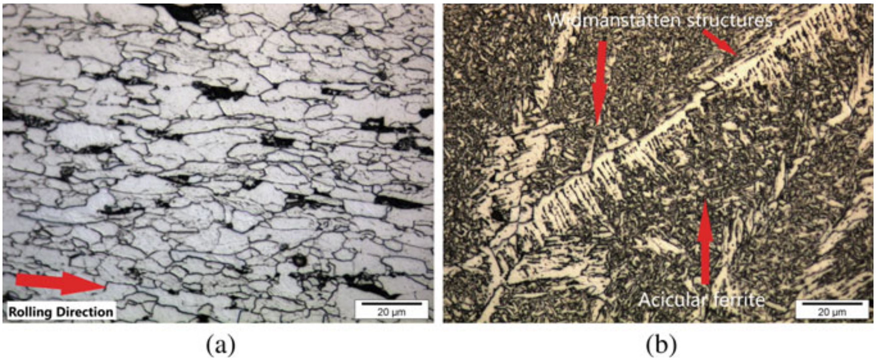

Fig. 1 **a** Microstructure of the base metal with ferrite (white) and pearlite (black) regions, and **b** the weld metal where the acicular and Widmanstätten structures are indicated. (Color figure online)

and weld metal have an HV1 hardness of 200 ± 3.7 and 248 ± 9.9, respectively. The X60 base metal has a yield strength of 461 MPa.

Methods

A novel test setup and specimen were designed in this research. The specimens, shown in Fig. 2, were machined from the X60 pipeline wall in longitudinal direction. Both specimens consisting of just the base metal as well as specimens including the girth weld zone were manufactured, in which case the weld metal was present in the notch region. The blunt notch was introduced to the specimen to enforce fracture in the weld zone without causing high stress triaxialities that will result in exaggeration of the HE susceptibility. A blind hole was machined into the specimen, so that it can be charged with hydrogen gas from the inside. In this way, the specimen acts as a miniature pipe where hydrogen gas is present on the inside, while the outside is left open to the laboratory environment. The specimens are machined in such a way that the girth weld remains in the same orientation as in the pipeline.

SSRT testing was performed on smooth base metal specimens, notched base metal and weld metal specimens under the following conditions: without any internal pressure, at 100 bar N_2 pressure and at different levels (0–100 bar) of H_2 pressures.

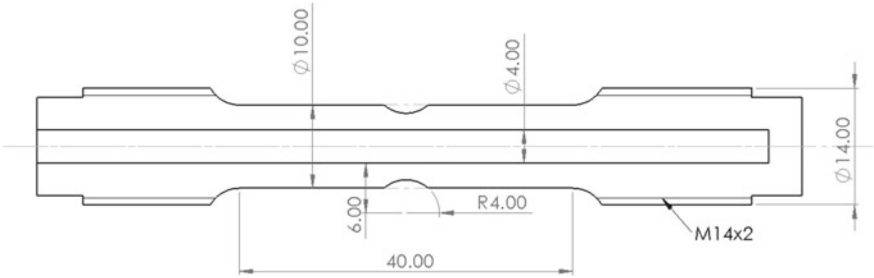

Fig. 2 Schematic drawing of the samples used in this research. Dotted lines in the notch represent the measurements of smooth samples

Every test was performed at a cross-head displacement speed of 1.5 mm/h, which translates to a strain rate of 10^{-5} s^{-1} for smooth (un-notched) specimens. Tests that were performed in H_2 gas also included a pre-charging step in which the sample was kept at the testing pressure for 17 h before the start of the test. At least three repetitions were tested for each combination of sample type and pressure, after which the tensile data was analysed. Fractographic analysis was performed in a SEM to highlight the differences in fracture behaviour between the different specimens. Furthermore, the area of each fracture surface was measured using a digital optical microscope so that reduction in cross-sectional area (%RA) could be calculated.

In addition to the experimental setup, two FEA models were created to support the design. Both models use the same geometry based upon an axisymmetric mesh type to model the cylindrical notched specimen. A diffusion model was used to estimate the pre-charging time, and a deformation model was created to model the behaviour of the sample during the tensile test. The deformation model shows the emergence of a zone inside the specimen wall where the stress triaxiality approaches 0.75 as a consequence of the notch and the axisymmetry of the specimen.

Results and Discussion

A significant influence of H_2 on the base metal and weld metal was found from tensile data and SEM fractography.

Effect of Hydrogen on Mechanical Properties

Representative tensile curves of the different specimens are shown in Fig. 3. The most prominent effect of hydrogen that was found is a reduction in ductility of both the base and weld metal. As can also be seen in Table 2, no apparent influence of the

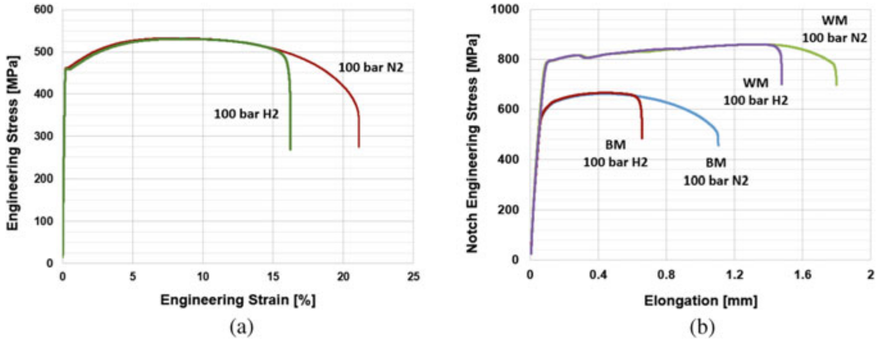

Fig. 3 **a** Tensile curves of smooth BM samples tested in 100 bar N_2 and 100 bar H_2, and **b** tensile curves of notched BM and notched WM samples tested in 100 bar N_2 and 100 bar H_2. (Color figure online)

hydrogen on either the yield strength or the UTS of the specimens was discovered. The notched base metal specimens lost 40.4% in ductility when subjected to 100 bar hydrogen gas. This is higher than that of the smooth specimens (27%), because of the higher stress triaxialities present in the notch root compared to a smooth specimen. Both the yield strength and the UTS of the weld metal exceed that of the base metal by approximately 30%. Moreover, the elongation of the weld metal samples at failure is 1.72 mm compared to 1.08 mm for the base metal. This means the weld metal is stronger and more ductile than the base metal, but still retains much more ductility when subjected to gaseous hydrogen. The specimens only lose 14% in ductility at 100 bar of H_2 pressure. This is because the weld metal has a more refined microstructure of small interlocking acicular ferrite grains and a high grain boundary density. This structure creates maximum resistance to brittle crack propagation which leads to a smaller reduction in elongation under influence of hydrogen compared to the base metal.

The notched base metal specimens showed a 28% reduced %RA in H_2 as compared to N_2. The weld metal specimens show an 11% decrease in %RA in H_2 compared to N_2. The %RA values for the weld metal were constant over different pressures, meaning that the trend that was observed of a reduction in elongation with increasing hydrogen pressure is not present in the %RA values. This can be explained by looking at the fracture surfaces.

Fractography

The reduction in %RA values for both steels in hydrogen is a direct consequence of the HE fracture mechanisms. Martin et al. study characteristic smooth features and ridges that arise on so-called quasi-cleavage (QC) fracture surfaces that is typical of hydrogen enhanced fracture [4, 5]. They observe nanoscale dimples on the smooth

Table 2 Yield strength, UTS, elongation until failure and %RA values for the notched base metal (BM) and weld metal (WM) specimens. Values are averages ± standard deviations of each set

	Notched BM 100 bar N_2	Notched BM 100 bar H_2	Notched WM 100 bar N_2	Notched WM 100 bar H_2
Yield strength (MPa)	626 ± 24.6	631 ± 18.3	810 ± 5.9	812 ± 4.8
UTS (MPa)	674 ± 15.1	677 ± 12.9	857 ± 6.4	861 ± 1.5
Elongation at failure (mm)	1.08 ± 0.04	0.64 ± 0.06	1.72 ± 0.16	1.48 ± 0.11
Reduction (%)	–	40.4	–	14.0
%RA	72.1 ± 1.0	52.0 ± 2.0	48.2 ± 1.5	43.0 ± 3.0
Reduction (%)	–	28.0	–	10.8

QC areas and a high density of dislocations under the fracture surface, pointing to a very localised ductile mode of failure rather than a brittle one. As can be seen in Fig. 4a, similar features were found on the base metal samples tested under 100 bar H_2 pressure. Such highly localised ductile failure acts before larger-scale necking and causes a reduction in %RA for the specimens. Both smooth and notched base metal specimens showed near identical %RA values, which could be explained by their fracture surfaces that were both QC dominated. It was also found that notched specimens show more secondary cracking perpendicular to the fracture surface in the zones of large triaxiality, but this does not influence the cross-sectional area.

As shown in Fig. 4b, weld specimens were found to only partially fracture in the QC mode before reverting back to a more ductile microvoid coalescence (MVC) mode near the outer surface. The size of the QC fracture area was found similar for

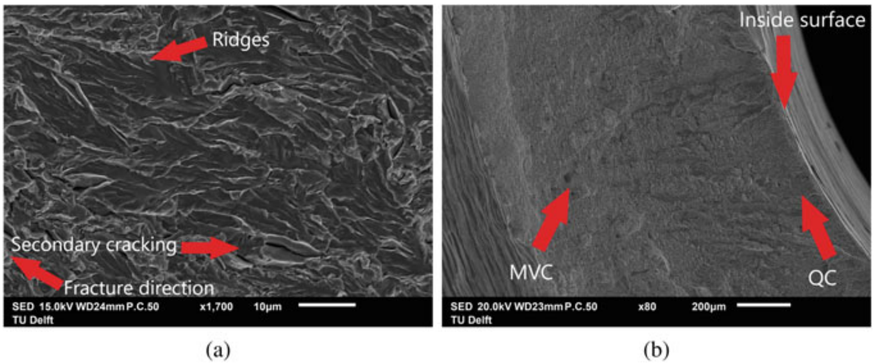

Fig. 4 **a** SEM image of the fracture surface of a smooth base metal sample tested in 100 bar H_2 gas showing smooth quasi-cleavage (QC) facets with ridges and secondary cracking, and **b** an overview of the fracture surface of a weld sample tested in 100 bar H_2 gas showing partial QC and partial microvoid coalescence (MVC) cracking. (Color figure online)

weld specimens tested in different hydrogen pressures, which could explain their similar %RA values. Because of the weld metal's small grain size, no characteristic QC features could be identified on their fracture surfaces. The fact that there is a transformation from QC to MVC fracture could be attributed to a more abruptly occurring fracture in the weld metal specimens. The base metal specimens showed a more gradual decrease in force before failure, pointing to a slower advancing crack front, while a faster advancing crack in the weld specimens might not have allowed the QC fracture mode to form over the entire fracture surface.

Secondary cracking (of up to 100 μm) parallel to the fracture surface was frequently observed in smooth samples tested in H_2 gas. In a gaseous hydrogen environment, the sensitivity of the material to sharp defects increases, and crack initiation can be accelerated. It is suspected that the secondary cracks initiated from machining defects. Although the inside surface of the smooth samples was mechanically reamed, it was still sensitive to accelerated cracking in a hydrogen environment. The outside surface, which was not reamed, did not show any signs of secondary cracking. It should be noted that notched specimens showed less secondary cracking, likely because strain in these samples was localised to the notch root where fracture occurred.

Conclusions

The following conclusions can be drawn from this research:

- A novel SSRT setup featuring in situ hydrogen gas and samples representing miniature pipelines was successfully applied for assessing HE susceptibility of both base and weld metals of pipeline steels.
- Both the base and weld metal were found susceptible to HE. Although the behaviour before fracture (yield strength and UTS) was found to be unaffected by H_2, a substantial reduction in ductility of 40% for base and 14% for weld metal under 100 bar H_2 was observed.
- The weld metal was found less susceptible to HE due to its fine acicular ferrite microstructure with its inherent resistance to crack propagation.
- Reduction in elongation of the weld specimens showed an increasing trend with increasing H_2 pressure. It should also be noted that %RA values did not show this trend, which indicates that %RA is not a suitable parameter to assess the effect of hydrogen pressure on the extent of HE.

Some aspects that are essential for the assessment of pipelines to HE were not researched in this work, but remain as recommendations for future work:

- Pipelines rarely operate in high plastic strain regimes, but rather at lower strains in cyclic loading. Therefore, cyclic testing is necessary to fully characterise the effect of HE on a pipeline in its application environment.

- Existing pipelines can have existing defects. These could be simulated by altering the notch geometry in the samples discussed in this work and thus result in useful fracture toughness data.

Details regarding the development of the test setup will be discussed in a follow-up journal paper.

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