Corrosion Science 78 (2014) 313–321

Contents lists available at ScienceDirect

Corrosion Science

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

Fatigue crack growth of two pipeline steels in a pressurized hydrogen environment ☆

Andrew J. Slifka *, Elizabeth S. Drexler, Nicholas E. Nanninga, Yaakov S. Levy, J. David McColskey, Robert L. Amaro, April E. Stevenson

NIST, Materials Reliability Division, 325 Broadway, Boulder, CO 80305, USA

ARTICLE INFO

Article history:
Received 2 July 2013
Accepted 13 October 2013
Available online 22 October 2013

Keywords:
A. low alloy steel
C. hydrogen embrittlement
C. intergranular corrosion

ABSTRACT

Fatigue crack growth tests were conducted on two pipeline steel alloys, API 5L X52 and X100. Baseline tests were conducted in air, and those results were compared with tests conducted in pressurized hydrogen gas. All tests were run at (load ratio) $R = 0.5$ and a frequency of 1 Hz, except for one test on X100, run at 0.1 Hz. Tests were conducted at hydrogen pressures of 1.7 MPa, 7 MPa, 21 MPa, and 48 MPa. Fatigue crack growth rates for both X100 and X52 were significantly higher in a pressurized hydrogen environment than in air. This enhanced growth rate appears to correlate to pressure for X100 but may not for X52.

Published by Elsevier Ltd.

1. Introduction

The use of hydrogen as a fuel and as an energy carrier is likely to increase in the next decade. According to the Fuel Cell and Hydrogen Energy Association, "Nearly all of the major auto manufacturers have 2015 as the projected release date for mass-produced models" [1], which means that economical transportation and distribution of hydrogen is needed prior to 2015. Hydrogen can be electrolyzed with excess electricity generated by solar and wind farms. The hydrogen can be stored and used during peak demand or transported to areas of need for Smart Grid applications. According to the U.S. Department of Transportation, "Pipelines are the safest and most cost-effective means to transport the extraordinary volumes of natural gas and hazardous liquid products that fuel our economy" [2]. Therefore, one would expect that pipelines would be the safest and most economical means of transporting large volumes of hydrogen. The primary issue with hydrogen is that it embrittles most materials, leading to a likely decrease in lifetime of a pipeline transporting hydrogen, compared to the transport of natural gas. The embrittlement of pipeline steels due to hydrogen is well documented in the case of tensile tests [3–17]. However, the literature contains far less information on hydrogen-assisted cracking or fatigue properties of pipeline steels [3–5,12,13,18–22]. There has been some recent work done on fatigue crack initiation with a 3-point bend test, but little work on fatigue crack growth in pipeline steels [23,24]. Some work has been done in recent years on measurement of fatigue properties of X60, X70 and X80 pipeline steels [8,13,20].

In order to safely and efficiently design pipelines for transport of hydrogen gas, codes such as ASME B31.12 on Hydrogen Piping and Pipelines are used [25]. Due to the dearth of fatigue information, codes rely upon tensile data in order to factor in design variables such as pressure. However, pipelines are not operated at or near material yield stress, such that embrittlement data from tensile tests may produce codes that are too conservative. Since safety is the primary factor in these codes, fatigue data from many different pipeline steels, encompassing many different microstructures, is needed if any modifications are to be made to the codes. These pipelines are expected to operate for decades, which will allow many or most of the hydrogen traps in the steel to be filled. This work will not address that issue. Future work is planned on samples that have been pre-charged either with high-pressure gas at elevated temperature or by cathodic charging. However, due to the rapid rate of diffusion of hydrogen in ferritic steels [26], being at least 5 orders of magnitude greater than in austenitic steels [6,27], hydrogen diffuses the few mm to the midpoint of a pipeline steel section in a manner of minutes [26]. An additional effect that is not accounted for in the results presented in this work is that of very slow stress cycling. A fuel pipeline would be expected to experience pressure cycles on the order of two per day. That type of slow cycling, done over some breadth of stress intensity ranges, would take a prohibitive amount of time to complete. However, some work has been done in air at very low cyclic frequencies [28]. That work includes fatigue crack growth rate (FCGR) measurements down to 0.001 Hz. Those authors

---

☆ Contribution of the National Institute of Standards and Technology, an agency of the US government; not subject to copyright in the USA.

* Corresponding author. Tel.: +1 303 497 3744.
E-mail address: andrew.slifka@nist.gov (A.J. Slifka).

0010-938X/$ - see front matter Published by Elsevier Ltd.
http://dx.doi.org/10.1016/j.corsci.2013.10.014

demonstrate that low temperature creep blunts the crack tip and reduces the crack growth rate. That phenomenon would work against the corrosion fatigue effect of hydrogen at the crack tip. Moreover, the strain hardening differences between X52 and X100 could cause a significant difference in creep behavior, because strain hardening effects can cause large differences in creep behavior of pipeline steels [29]. Measurements in hydrogen at very low cyclic loading rates need to be done to resolve the relationship between these competing mechanisms.

The work presented here deals only with diffusible hydrogen in the lattice that is near the crack tip. It reports measurements of FCGR of X52 and X100 pipeline steels at 1 Hz loading frequency, which is high compared to what an actual pipeline would see. These steels were chosen because X52 is material that is currently being used for hydrogen pipeline applications, and X100 represents the upper limit of strength that is likely to be used in the near future. The microstructures of these steels are very different, which may lead to differences in FCGR.

2. Materials and methods

2.1. The materials

This study investigates the fatigue crack growth properties of two very different grades of linepipe material, API 5L types X52 and X100. Sections of pipe were supplied from members of the pipeline industry. The X52 pipe was 0.51 m diameter and 12.7 mm thick and the X100 pipe was 1.3 m in diameter and 20.6 mm thick. The X100 pipe was an experimental pipe produced in the latter half of the 1990s. The X52 pipe was of early 21st century vintage and had never been in service.

Round specimens were made according to ASTM E8 [30] and were tested in air with a strain rate of 3×10^{-3} mm/s. The material was not flattened prior to machining the tensile specimens in order to measure the properties of the formed pipe. The tensile properties of the base material from each pipe are shown in Table 1. The "*L*" orientation, longitudinal, is along the pipe axis, and the "*T*" orientation, transverse, is along the hoop direction, perpendicular to the longitudinal direction.

As would be expected with materials with such different strengths, the microstructure was very different when comparing the two pipes. The X52 has about 10% pearlite in a polygonal ferrite microstructure, shown in Fig. 1a. This material shows some banding, but none of large scale, but shows a small amount of chemical segregation at low magnification, seen in Fig. 1a. Fig. 1b shows a higher-magnification optical image of the microstructure, which is predominantly polygonal ferrite plus carbide. The X100 material is shown in Fig. 2 at two magnifications. The low-magnification image shows that this material has considerable banding or macroscopic chemical desegregation, but it is not concentrated at any particular level through the thickness. The high-magnification image shows a fine-grained microstructure of lath-like components, including bainite and acicular ferrite. The small-scale banding, or chemical microsegregation, is evident even at high magnifications. Table 2 gives the chemical compositions of these two steels.

Table 1
Tensile properties of the base metals X52 and X100 used in this study.

Steel	Orient.	σ_y (MPa)	σ_{UTS} (MPa)	e_f (%)
X52	*L*	426 ± 10	491 ± 8	27.4
X52	*T*	436 ± 5	504 ± 5	27.0
X100	*L*	705 ± 40	803 ± 6	20.3
X100	*T*	794 ± 11	827 ± 5	19.3

2.2. The test matrix

Material from each linepipe was used to measure the FCGR in air, at various pressures of hydrogen gas, and at different frequencies. The X52 was tested at two different hydrogen pressures (7 MPa and 21 MPa); in addition to those pressures, the X100 material was also tested at 1.7 MPa. Only the X100 material was tested at different frequencies: 1 and 0.1 Hz. One specimen was tested for each condition.

The sub-sized specimens were designed according to the specifications for a compact tension (CT) specimen, found in ASTM E647-05 [31], with a $W = 26.4$ mm. Fig. 3 shows a drawing with the pertinent dimensions of the CT specimens. The surface roughness R_a was less than 0.25 μm for all specimens tested. The specimens are made in the *T–L* orientation, which means that the crack runs in the longitudinal (along the pipe axis) direction of the pipeline, and the cyclic loading is applied in the transverse (or hoop stress) direction, in accordance with ASTM E399 [32]. A fatigue crack is most likely to propagate in the longitudinal direction of a pipe due to the effect of the higher stress in the transverse, or hoop, direction.

The specimens were fatigue precracked (FPC) in air to generate a sharp crack at the notch tip with a load ratio $R = 0.1$ and a frequency of 10 Hz. The average length of the precrack was 3.12 mm, which corresponds to the width of the notch.

2.3. Pressurizing system

Specimens were tested in a stainless-steel pressure vessel (internal diameter 101.6 mm diameter and length 254 mm), capable of holding hydrogen gas pressures up to 140 MPa. The pressure vessel was closed on one end and had a sliding seal and pull rod on the opposite end. The pull rod was connected to the actuator of a servo-hydraulic test frame with 90 kN loading capacity. Fig. 4 provides a schematic of the test frame, pressure vessel, fatigue sample, and instrumentation used for testing. An internal load cell, designed for use in high-pressure hydrogen gas, was employed inside the pressure vessel. This was done because friction forces at the sliding seals, which vary with test pressure, can lead to differences between the force measured at the externally-mounted load cell on the test frame and that which would be exerted on the specimen, as seen in Fig. 5. The internal load cell uses a linear variable differential transducer to measure displacement, and was calibrated in air using the external load cell as a reference. All reported forces are those measured from the internal load cell. Control of cyclic loading was done with the internal load cell in the form of a sine wave, plotted in black in Fig. 5. Crack length was determined by compliance [$\Delta v/\Delta P$, where v = CMOD (crack mouth opening displacement) and P = force], as measured by a clip gauge tracking the CMOD. A relationship that uses the elastic modulus of the material, the geometry of the specimen, and the CMOD is used to calculate crack length, per ASTM E647-05 [31].

Upon installation and sealing the pressure vessel, the chamber was purged twice with ultra-pure helium (99.9999%) and three times with ultra-pure hydrogen (99.9995%) to 14 MPa, or the designated test pressure, whichever was lower, before the final pressurization to the test pressure. This entire purging/venting process is controlled by a computer program which automatically opens and closes air-operated valves in a pre-determined sequence to effect the cleansing of the system. Automating the cleansing and maintaining pressure eliminates the risk of human error and provides consistency from run to run.

This series of tests were conducted with hydrogen that was generated on-site with an electrolyzer unit. Analysis of the gas produced showed that it was at least as clean as the purest