In-Situ Monitoring of Hydrogen Embrittlement in Ferritic Steel Pipelines

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The Stress-Strain Microprobe® (SSM) system that utilizes an in-situ nondestructive Automated Ball Indentation (ABI) test technique was used to monitor the changes in tensile and fracture toughness properties of hydrogen transmission pipeline steels. Fracture toughness was calculated from integration of the ball indentation/deformation energy as a function of depth up to a critical depth. The latter depends on the critical fracture stress at the test temperature.

ATC's innovative hydrogen pressure chamber allows simultaneous exposure of six pipeline steels, and multiple ABI tests were performed on each disc at various times. The ABI-determined fracture toughness of X80 pipeline steel decreased by 12%, 25%, 32%, and 30% after 13.79 MPa (2,000-psi) hydrogen exposure times of 5, 25, 100, and 200 hours. The reduction in fracture toughness saturated at 100 hour for the 9.5-mm thick sample while the increase in tensile properties was very small for the X80 steel. No hydrogen embrittlement was detected for the other pipeline grades tested in this project.

1. Introduction

Monitoring hydrogen embrittlement is required for the safe transmission of hydrogen in ferritic steel pipelines. The chemical composition and microstructure of pipeline steels are key factors affecting their susceptibility to hydrogen embrittlement. Hence, several grades of pipeline have been investigated in this project, and their tensile and fracture toughness properties were non-destructively measured as a function of exposure time to hydrogen pressure. A small piece of grade X80 steel was received from a commercial vendor who performed destructive fracture toughness testing on the material (outer diameter of 406 mm and wall thickness of 14.6 mm). The conventional testing used 0.5T CT specimens that were soaked in hydrogen for 30 minutes before their testing according to ASTM Standard E1820 for initiation fracture toughness (J_{Ic}). Destructive testing showed that the fracture toughness decreased from 225 MPa√m (zero hydrogen pressure) to 118 MPa√m (13.79 MPa or 2000 psi hydrogen pressure), i.e. a 48% reduction due to hydrogen embrittlement. The chemical composition of the X80 seamless pipeline steel is given in Table 1. The microstructure of the X80 pipeline steel consisted of bainite or tempered martensite with fine grains and low inclusions.

Conventional fracture toughness testing has two disadvantages, namely, using large amounts of hydrogen for purging the autoclave prior to testing and pressurizing the autoclave during testing, and for exposing the specimen from both sides (although in a pipeline transmission the pipe wall is hydrogen pressurized only on the inside surface). The Automated Ball Indentation (ABI) method and the hydrogen chamber of this investigation have several advantages over conventional destructive testing: nondestructive, in-situ, use a very small volume of hydrogen per experiment, use multiple pipeline steel specimens exposed to the same hydrogen pressure and purity per single experiment, and conduct multiple ABI tests per each disc sample.

Table 1 Chemical composition of X80 pipeline steel used in this investigation

C	Mn	P	S	Si	Cu	Ni	Cr
0.23	1.37	0.011	0.002	0.23	0.01	0.01	0.126
Mo	Sn	Al	V	N	Ca	C.E.	
0.06	0.001	0.056	0.048	0.0043	0.0029	0.502	

2. Test Method

This article describes a new application of the innovative Stress-Strain Microprobe® (SSM) system that utilizes an in-situ nondestructive Automated Ball Indentation (ABI) test technique to measure the stress-strain curves and fracture toughness properties of in-service steel pipelines. The ABI test provides the actual/current values of these mechanical properties for base metal, welds, and heat-affected-zones. The SSM system is used in this research to quantify and monitor hydrogen embrittlement of several pipeline steel samples exposed to 13.79 MPa (2000 psi) hydrogen in a custom-built hydrogen chamber.

The SSM system utilizes an ABI technique that is nondestructive, fast (less than two minutes per test), and very accurate. The ABI test requires a reasonable, localized polishing of the test area for indentation testing. The spherical indentations produced on the pipeline surface are shallow, smooth depressions (i.e., no sharp edges and, hence, no stress concentration sites). Furthermore, the ABI test leaves a compressive surface residual stress that retards crack initiation (similar to the shot peening process used routinely in the aerospace industry). Therefore the ABI test, although a true/robust mechanical test, is considered for practical purposes nondestructive. Thousands of ABI tests have been conducted on ferritic steel samples, including grades from B to X100 of pipeline steels at various test temperatures. Also, numerous ABI tests have been conducted in the field (at ambient temperatures) on various metallic structures in the USA, Europe, Africa, and Asia (Fig. 1) [1-6]. The ABI test is based on a progressive indentation with intermediate partial unloadings until the desired maximum depth (maximum strain) is reached, and then the indenter is fully unloaded (Fig. 2a). The indentation load-depth data are collected continuously during the test using a 16-bit data acquisition system. The nonlinear spherical geometry of the tungsten carbide indenter allows increasing strain as the indentation penetration depth is increased. Hence, the incremental values of load (at the end of each progressive loading cycle) and plastic depth (associated with each partial unloading cycle and the upper part of the final full unloading data) are converted to incremental values of true-stress and true-plastic-strain values (Fig. 2b) according to established elasticity and plasticity theories (Equations 1-5 of Table 2)[1-3].

Fig. 1 The testing head of the SSM system is mounted on a 254-mm diameter gas pipeline using DC electric magnets.

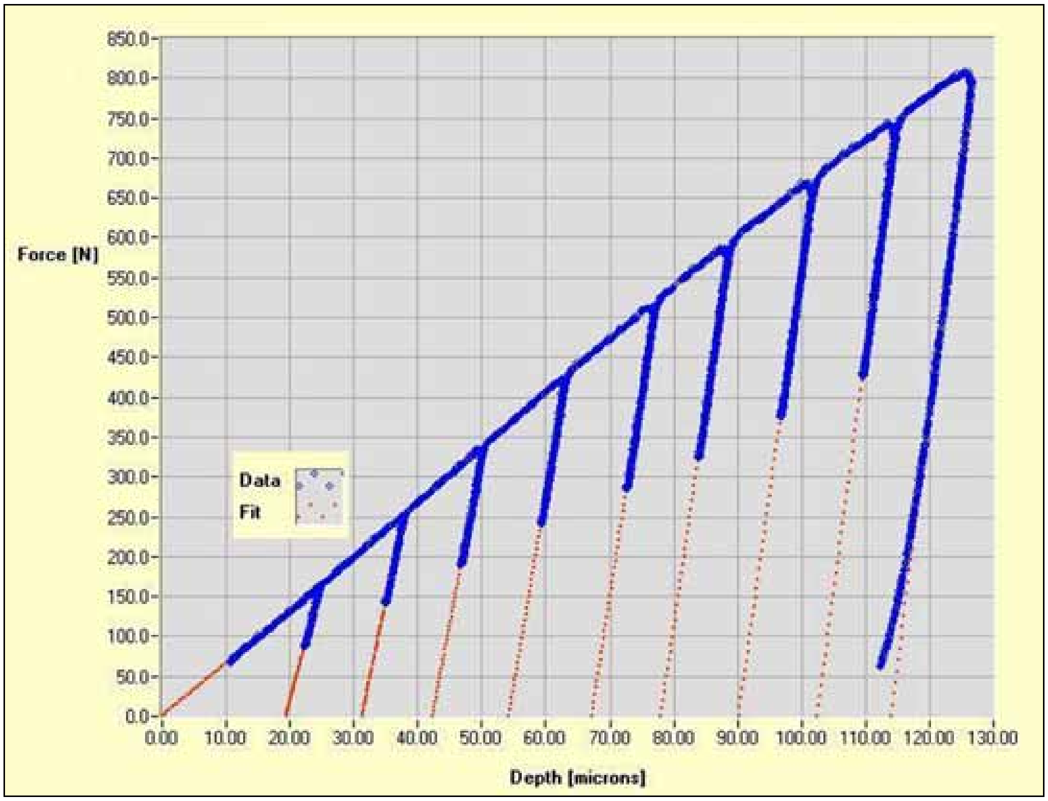

Fig. 2a Automated Ball Indentation Force versus depth using a 0.76-mm (0.030-in) diameter tungsten carbide indenter.

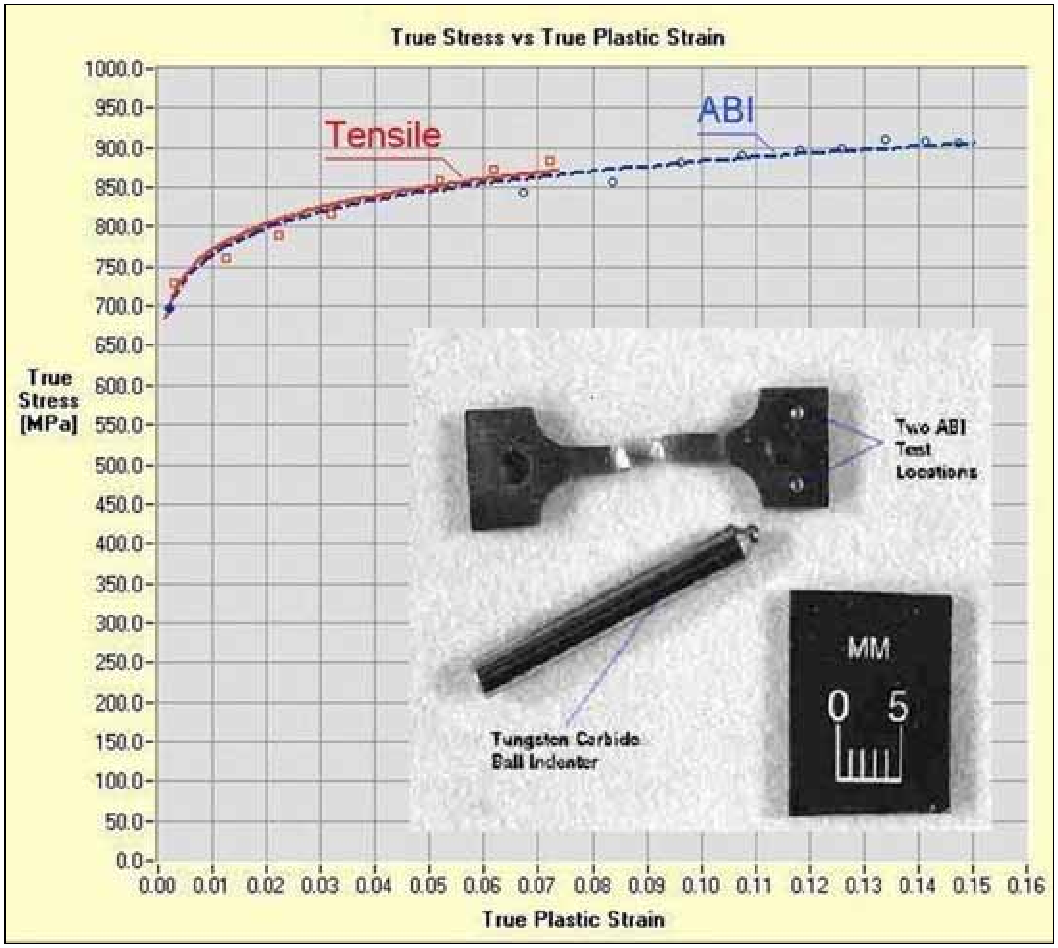

Fig. 2b Comparison of stress-strain curves from the ABI load-depth data and from a miniature tensile test of high strength steel.

The ABI test is fully automated (using a computer, data acquisition system, and a servo motor), and a single test is completed in less than two minutes depending on the desired strain rate. The ABI test is applicable to all materials regardless of the amount of material pile-up around the indentation (the pile-up volume depends on the thermo-mechanical treatment and/or Lüders strain behavior of the test material). Haggag's earlier work [6] used mechanical profilometry and optical interferometry to quantify the pile-up and/or Lüders strain in order to accurately determine yield strength and stress-strain values from ball indentation. Since these methods are cumbersome and not suitable for field applications, in 1989, Haggag [1] invented the progressive ABI test with its novel intermediate partial unloadings to make the test easier, automated, faster, more accurate, and applicable for field/in-situ test applications. Linear regression is performed on the load-depth data of each elastic/linear partial unloading and on only the upper part of the full unloading at the end of the test, and then the data are extrapolated to the X-axis to determine the plastic depth associated with maximum load of each loading cycle of the entire ABI test. The partial unloadings are linear with increasing slopes as the elastic recovery volume increases with increasing indentation depth. The innovative partial unloading technique accounts for the material plastic pile-up because it detects only the elastic recovery of the current spherical indentation of each progressive ABI cycle. Use of full unloading is incorrect for determining the plastic depth because of non-linearity of the last/lowest 20-30% of the full unloading data (experimental non-linearity results in lower plastic depth and consequently incorrect plastic-strain value).

Table 2a - Equations

$$\in_p = \frac{0.2 d_p}{D} \tag{1}$$

$$\sigma_t = \frac{4P}{\pi d_p^2 \delta} \tag{2}$$

$$d_t = 2\sqrt{h_t D - h_t^2} \tag{3}$$

$$\frac{P}{d_t^2} = A\left(\frac{d_t}{D}\right)^{m-2} \tag{4}$$

$$\sigma_y = \beta_m * A + B \tag{5}$$

$$IDE = \int_0^{h_f} P_m(h)\,dh \tag{6}$$

$$P_m = \frac{4P}{\pi d_t^{\,2}} \tag{7}$$

$$W = W_0 + W_T \tag{8}$$

$$(K_{Jc})^{ABI} = 30 + \sqrt{2E(W_T)} \tag{9}$$

$$K_{Jc}(med) = 30 + 70e^{0.019(T-T_0)} \qquad \text{MPa}\sqrt{\text{m}} \tag{10}$$

$$K_{Id}(med) = 30 + 70e^{0.019(T-T_0+T_{shift})} \qquad \text{MPa}\sqrt{\text{m}} \tag{11}$$

$$T_{shift}(^\circ F) = 215 - 1.5\sigma_y(ksi) \qquad \text{for 36 ksi} < \sigma_y < \text{140 ksi} \tag{12}$$

Table 2b – Definitions

ϵ_p = true plastic strain,
d_p = plastic indentation diameter,
D = diameter of the ball indenter.
σ_t = true stress,
P = applied indentation load,
δ = a parameter whose value depends on the stage of development of the plastic zone beneath the indenter.
h_t and d_t are the total indentation depth (plastic + elastic) and total indentation diameter while the load is being applied, respectively.

A is the material yield parameter and m is Meyer's index.
σ_y is the ABI-determined yield strength, β_m is the material yield slope, and B is the yield-strength offset-constant.
The Indentation Deformation Energy (IDE) is a function of ball indentation depth (h) and mean pressure (P_m)

The critical indentation depth (h_f) is the depth when the maximum stress equals the critical fracture stress of the ferritic steel at the low-test temperature.
The static fracture toughness, K_{Jc}, has a non-zero lower shelf even at very low-test temperatures.
The fracture toughness energy in J-Integral units is W.
W_o is the lower shelf energy per unit area (30 MPa√m).
W_T is the temperature-dependent energy ($W_T = IDE$).
T is the test temperature in ºC and T_0 is the reference temperature when $K_{Jc} = 100$ MPa√m
K_{Id} is the median dynamic fracture toughness

The plastic indentation depth and its associated cycle maximum load, the indenter diameter, and the elastic moduli of the test material and the indenter are used to calculate the plastic indentation diameter and consequently the true-plastic-strain [1-3]. Fig. 3 shows a final indentation produced using a 1.57-mm diameter tungsten carbide indenter on a 4142 steel sample. Despite pronounced pile-up shown in Fig. 3, the average plastic indentation diameter of 945 µm (from optical measurements of 0.943 mm and 0.947 mm) is within 1.1% of the calculated diameter of 935 µm using the innovative partial unloading technique. The precision of the ABI test method has been further demonstrated (Table 3) from a round robin study with participation of six organizations. The materials used in the round robin study are two aluminum alloys (6061 and 7075) and two steel alloys (1018 and 4142) with a wide range of flow properties. Other ball indenters (e.g., 0.25-mm, 0.51-mm, and 0.76-mm diameters) can be used to interrogate small volumes (e.g., spot or laser-beam welds); however, the choice of indenter diameter must consider the grain size of the test material in order to measure macroscopic tensile and/or fracture toughness properties.

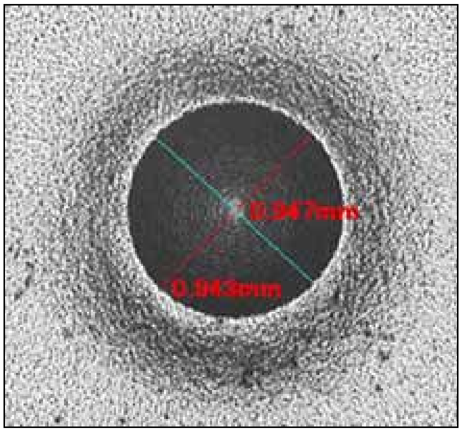

Fig. 3 Final indentation with pronounced pile-up from an ABI test on 4142 steel.

Table 3 Precision Values of the ABI Test Methods

	ABI-Yield Strength	ABI-Estimated Ultimate Strength	Strength Coefficient	Strain-Hardening Exponent	Uniform Ductility
CV $\%_r$	1.4	1.5	2.6	5.8	6.9
CV $\%_R$	1.7	2.3	3.4	6.7	7.8

CV $\%_r$ = repeatability coefficient of variation in percent within a laboratory
CV $\%_R$ = repeatability coefficient of variation in percent between six laboratories

3. Determination of Fracture Toughness Master Curve from ABI Tests

Indentation with a ball indenter generates concentrated stress (and strain) fields near and ahead of the contact of the indenter and the test surface similar to concentrated stress fields ahead of a crack tip; albeit the indentation stress fields are mostly compressive. The high value of the stress under the ball indenter is an example of ***plastic constraint*** when the rigid material surrounding the indentation volume does the constraining. Hence, at a certain critical ball indentation depth, there is a high state of transverse and lateral stresses similar to those in front of a sharp notch in an elastic material. Although the conditions for crack initiation might be attained, the high degree of plastic constraint will prevent cracks from developing during ball indentation of ductile metallic materials. Therefore, only initiation fracture toughness, not tearing modulus, can be determined from ball indentation (Equations 6-12 of Table 2). The initiation fracture toughness is calculated from the integration of the indentation deformation energy (*IDE*) up to the critical depth (when the maximum pressure underneath the ball indenter equals the critical fracture stress of the steel material at the test temperature or reaches a critical strain value of 0.12, whichever occurs first). An example of the ABI-measured fracture toughness results on pipeline steel is shown in Fig. 4.

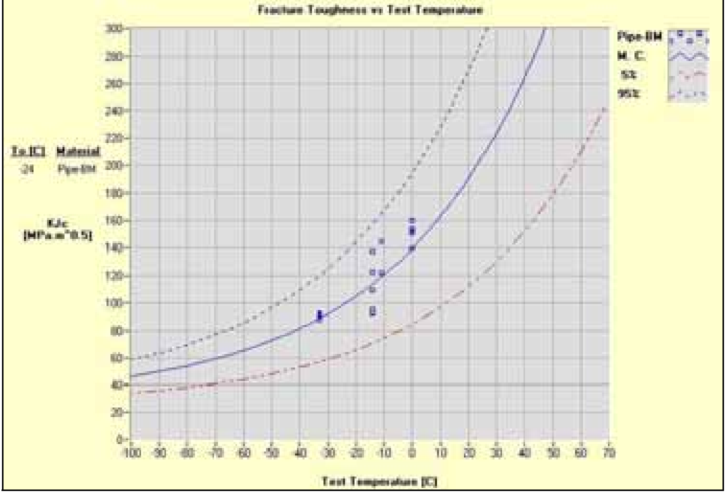

Fig. 4 Fracture toughness test results, their median master curve, and 95% and 5% confidence curves.

The ABI-measured fracture toughness capability is material-thickness independent since different size indenters can be used for all pipelines and pressure vessels to achieve valid results. Furthermore, the localized nature of ABI allows testing heat-affected-zones that cannot be tested destructively because of their irregular shapes and small volumes (Fig. 5). The determination of tensile and fracture toughness properties from each ***in-situ*** nondestructive ABI test allows deterministic structural integrity assessment or fitness-for-service evaluation based on robust fracture mechanics analysis.

Fig. 5 ABI tests on base metal, girth weld, and HAZ areas of high strength pipeline steel.

4. Results and Conclusions

The hydrogen chamber was designed to contain six disc samples, each of 25-mm diameter and 9.5-mm thick. It used a total volume of 36000 mm^3 (1.3 in^3). The testing head of the portable SSM system was temporarily attached to the top ferritic steel plate of the hydrogen chamber with a magnetic mount to perform ABI tests at various locations on each disc sample at various exposure times to monitor hydrogen embrittlement. The outer surface of each sample was sufficient to perform 16 ABI tests per disc sample. Due to the small volume of hydrogen, it was safely released in air after the end of the experiment (200 hours). The hydrogen chamber and the test setup are shown in Fig. 6. Several pipeline materials of Grade B, X52, X70, and X80 were included in the experiment, and in-situ ABI tests were conducted after 13.79 MPa (2000 psi) hydrogen exposure of 5, 25, 100, and 200 hours. Changes in the tensile and fracture toughness (see Fig. 7 for X80) properties due to hydrogen pressure exposure for various times were measured from the in-situ ABI tests.

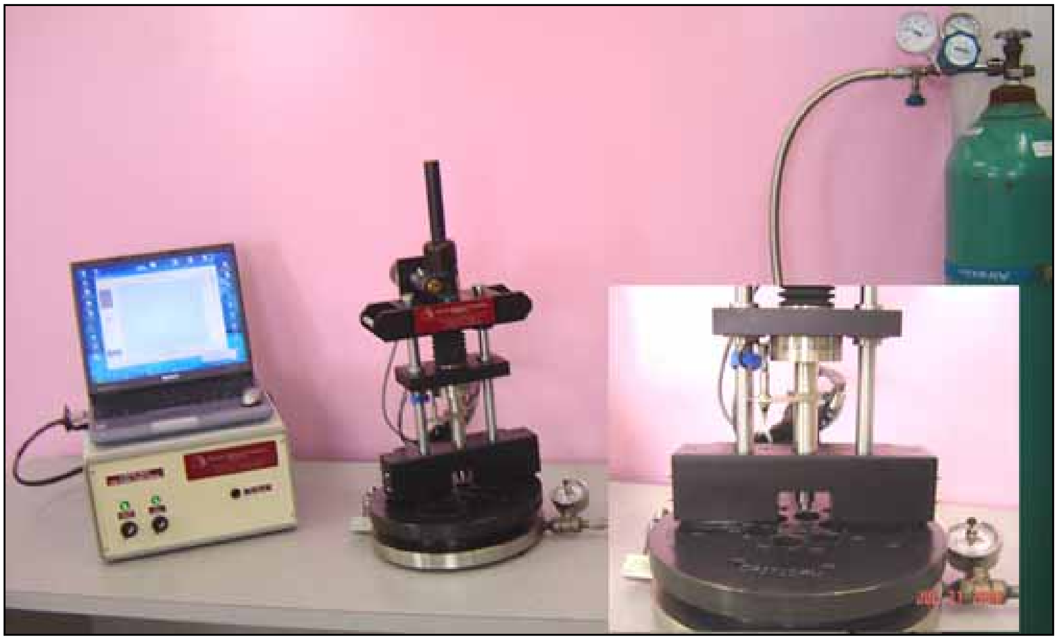

Fig. 6 Test set-up: SSM system, hydrogen chamber, and hydrogen cylinder.

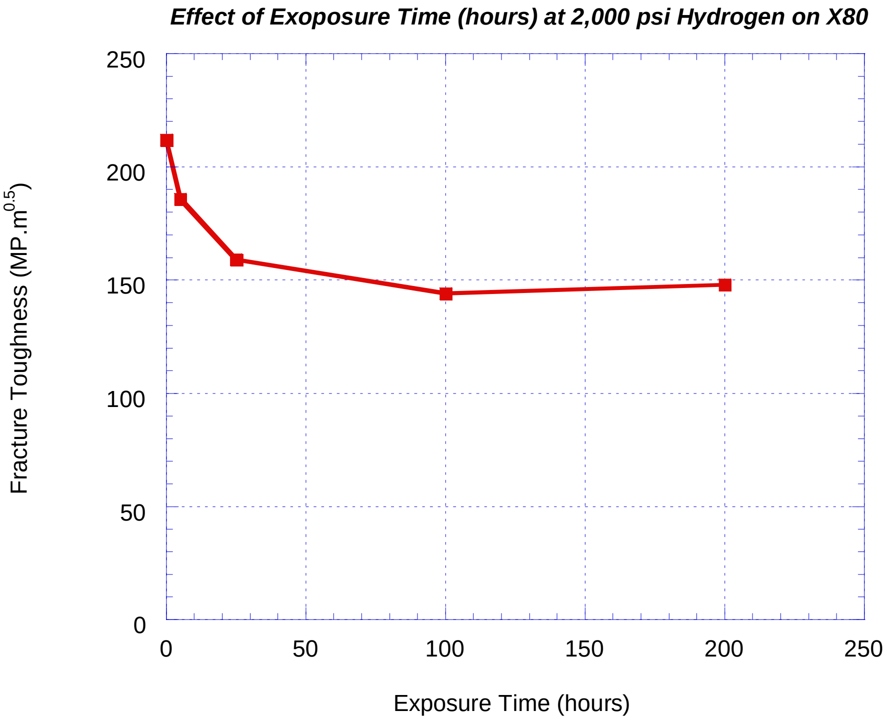

Fig. 7 Reduction in fracture toughness due to 13.79 MPa (2,000 psi) hydrogen exposure appears to saturate at 100 hrs for X80 steel.

The ABI-determined fracture toughness of X80 pipeline steel decreased by 12%, 25%, 32%, and 30% after 13.79 MPa (2,000 psi) hydrogen exposure times of 5, 25, 100, and 200 hours. The reduction in fracture toughness appears to saturate at 100 hours for the 9.5-mm (0.375-inch) thick sample.

This compared well with the 48% reduction in fracture toughness of 0.5T CT samples tested in an autoclave by the commercial vendor mentioned earlier. The complete immersion in hydrogen is more severe than actual hydrogen pipeline transmission application, and our hydrogen pressure chamber better simulates the real pipeline steel application since the samples are exposed to hydrogen pressure only on their inside surface. Furthermore, our portable/in-situ SSM system can be used to test the outer surface of in-service hydrogen transmission pipelines at various times to monitor their actual hydrogen embrittlement.

No reduction in fracture toughness was observed for disc samples manufactured from Grades B, X52, and X70 included in the same experiment with the X80 steel. The increase in tensile properties was very small for the X80 pipeline steel and no changes were observed for other grades.

References

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