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Evaluation of Electrochemical Hydrogen Absorption in Welded Pipe

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Abstract

The assessment of ability to absorb hydrogen by welds components of API grade pipeline steel X52 has been done. The factors of cathodic hydrogen charging, time of exposure on hydrogen concentration in base metal, heat affected zone and metal of weld were taken into account. It has been shown that all components of weld demonstrate the sensitivity to hydrogenating in deoxygenated, near-neutral pH NS4 solution under relatively “soft” cathodic polarisation, although the efficiency of hydrogen permeation in metal is relatively low and depends on time of exposure. The ability to absorb hydrogen decreases in the following sequence: heat affected zone – base metal – weld. The sensitivity to hydrogenation is higher for heat affected zone in comparison with base metal and weld.

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Keywords: welded pipe, hydrogen concentration;

1. Introduction

Process of free corrosion promotes the hydrogenating of steel, because hydrogen can evaluate on metal surface as result of cathodic counterpart of the anodic dissolution reaction, Cheng and Niu (2007) and Cheng (2007). Moreover, the hydrogen charging of steel is also possible when a cathodic protection system of pipeline works irregular, Shipilov and May (2006). Engineering practice of pipelines operation shows, according to Somerday et al. (2009) that the welds of pipelines are highly sensitive to the hydrogen influences from the reasons of their microstructural heterogeneity and difference of chemical composition of weld components (base metal, heat affected zone and metal of weld). A number of recent issues dedicated to different aspects of hydrogenating and hydrogen entry into welded joints of pipeline steels can be found in literature, Olden et al. (2012), Koenig et al. (2010), and

Briottet et al. (2012). In the work of Olden et al. (2012) the API X70 pipeline steel has been investigated with respect to hydrogen diffusion and fracture mechanics properties. Here it has been found that base metal and weld metal have marginal susceptibility to hydrogen embrittlement and the heat affected zone is clearly embrittled by hydrogen. Another aspect of considered problem is presented in the work of Koenig et al. (2010) where the non-destructive low-frequency impedance has been developed to determine hydrogen content in operating pipeline steel and weldment through a structural coating. The problems of the quantifying the hydrogen embrittlement of pipeline steels for safety considerations are pointed out in the papers of Briottet et al. (2012). Therefore, above-mentioned problem is real and the knowledge about hydrogenating ability of the weld components is required for reliable assessment of pipelines for transportation of hydrogen and natural gas mixtures. In presented paper the weldment of steel API X52 was studied, as it is the most used in existing European gas pipelines network, Capelle et al. (2008).

2. Experimental procedure

The material used for the study is API X52 that is the most usable in existed gas pipelines. The specimens were cut from a buried in-service pipeline. This pipeline with the outer diameter $D=610\text{mm}$ and wall thickness $t=11\text{mm}$, was in service approximately 50 years till it was cut for testing. The chemical composition of steel and its mechanical properties in air are given in Table 1 and 2 respectively. The structural specificity of the components of welded pipe can be seen in Fig 2. The base metal consists of grains of ferrite-pearlite, typical in all carbon steels (Fig. 1a). The weld metal showed the acicular ferrite columnar grains (Fig. 1b). The HAZ is composed of refined grains of ferrite-pearlite (Fig. 1c), product of the transformation of base metal with the heat provided by the welding process.

Table 1. Chemical compositions of steel API X52 (wt %).

C	Mn	Si	Cr	Ni	Mo	S	Cu	Ti	Nb	Al
0.206	1.257	0.293	0.014	0.017	0.006	0.009	0.011	0.001	<0.03	0.034

Table 2. Mechanical properties of steels API X52 in air.

Steel grade	σ_U, MPa	σ_Y, MPa	Elongation, %
X52	528	410	30.2

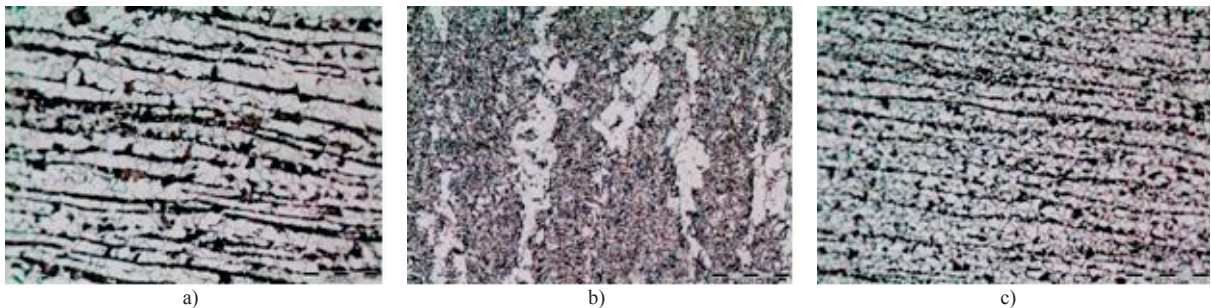


Fig. 1. Structural specificity of the components of welded pipe (x 200): a – base material; b – weld; c – heat affected zone.

The hydrogenation of specimens was made by electrochemical method. For this aim a special deoxygenated, near-neutral pH NS4 solution was chosen, Capelle et al. (2010). At these conditions hydrogen atoms are generated on the steel surface by the electrochemical reduction of water molecules according to known reactions, Cheng (2007). Before testing, the specimens were coated by special dielectric insulation and after the circular “windows” ($d=5\text{mm}$) with open surface of metal were made. This open surface of metal served as work electrode for electrochemical charging of hydrogen. The location of the “windows” with open surface of metal depends on the subject of test: base metal, weld metal or heat affected zone, Capelle et al. (2013). The electrolytic hydrogenation of specimens was made with application of the standard three-electrode electrochemical cell (open surface of metal (working electrode), auxiliary counter electrode and reference calomel electrode) where the Potentiostat VMP has been used. For modelling the hydrogen entry conditions in real operating pipelines, where there is the situation of

freely corroding system, the same procedure has been applied, Capelle et al. (2010).

3. Results and Discussion

With using the proposed experimental procedure the values of hydrogen concentration in the weld components of steels API X52 were determined depending on exposure time. The most general observations, which can be made from these results, are the followings. All components of weld demonstrate the sensitivity to hydrogenating in deoxygenated, near-neutral pH NS4 solution under soft cathodic polarisation and processes of hydrogen evolution and absorption have own specificity for each component. Here should be also pointed on some scatter of received data, which reflects the dynamics of variable processes at environment-surface interface. The non-monotonic dependencies of hydrogen concentration C_H on exposure time τ have been found for all components of weld (Fig. 2). After achieving of maximum, the hydrogen concentration decreases in time with some fluctuations to some steady-state value $C_{H(stab)}$. As can be seen the time of exposure $\tau_{C_{H(max)}}$, which corresponds of maximal value of $C_{H(max)}$, is different for base metal, weld metal and heat affected zone.

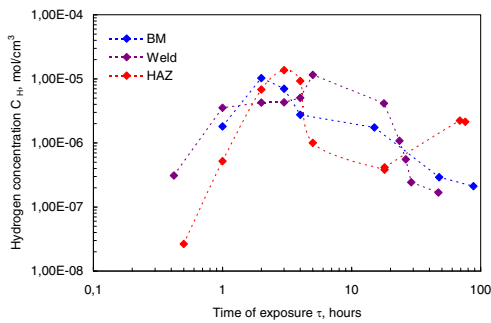


Fig. 2. Hydrogen concentration in components of weld.

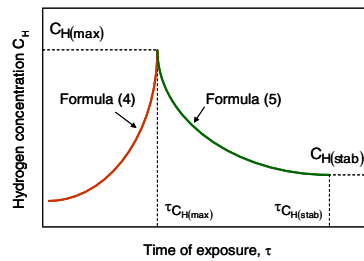


Fig.3. Schematic view of an experimental analysis.

Table 3. Value of parameters in formulas (1) and (2).

Component of weld	$C_{H(max)}, mol/cm^3$	$\tau_{C_{H(max)}}, hours$	n	m
Base metal (BM)	$1.03 \cdot 10^{-5}$	2	2.897	0.825
Weld metal (WM)	$1.15 \cdot 10^{-5}$	5	1.184	1.564
Heat affected zone (HAZ)	$1.36 \cdot 10^{-5}$	3	2.448	0.802

This fact can be used for the analysis of received results. When we chose $\tau = \tau_{C_{H(max)}}$ as some specific point then two stages of studying processes may be considered (Fig. 3), namely : the stage of increasing ($\tau \leq \tau_{C_{H(max)}}$) and the stage of its decreasing ($\tau \geq \tau_{C_{H(max)}}$). Taking into account some scattering of received experimental data (Fig; 2) we have described these stages (Fig. 3) by power functions, the values are given in Table 3:

$$C_H = C_{H(max)} \cdot \left(\frac{\tau}{\tau_{C_H(max)}} \right)^n \quad \text{under } \tau \leq \tau_{C_H(max)} \tag{1}$$

$$C_H = C_{H(max)} / \left(\frac{\tau}{\tau_{C_H(max)}} \right)^m \quad \text{under } \tau \geq \tau_{C_H(max)} \tag{2}$$

Based on such scheme of the results analysis the diagrams $C_H = f(\tau)$ for each component of weld were calculated and compared with primary experimental data (Fig. 4).

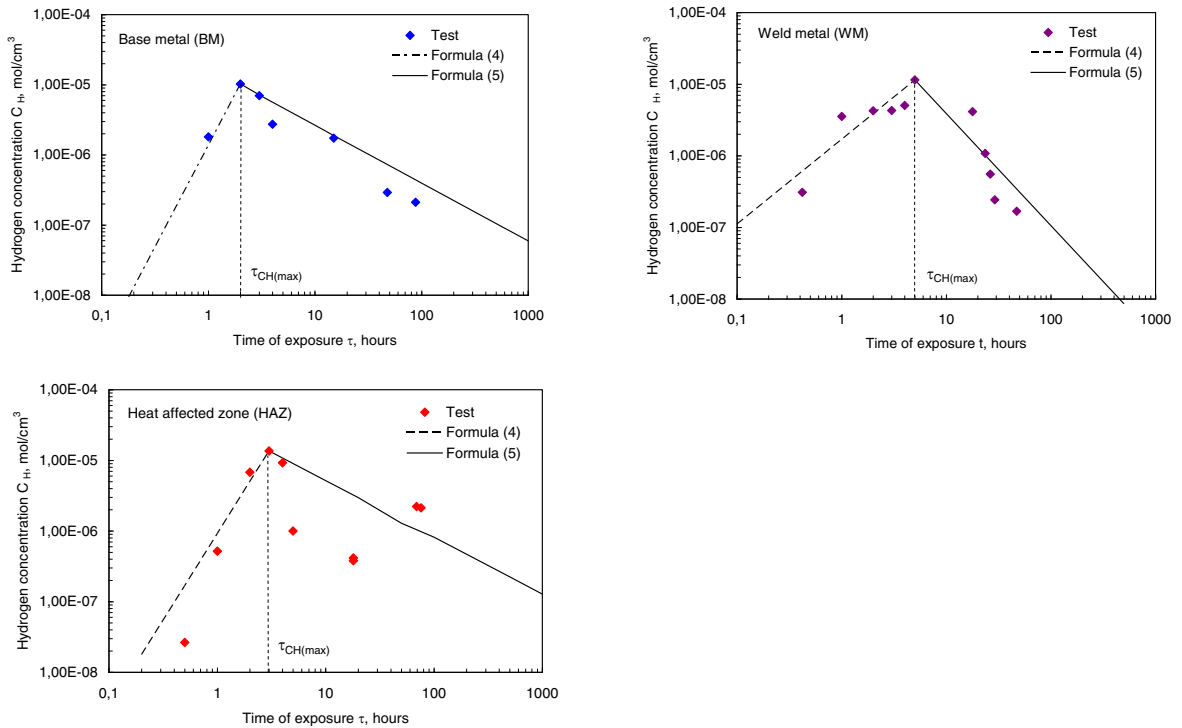


Fig. 4. Comparison of the calculated and experimental data on the hydrogen concentration in components of weld versus time of exposure.

Table 4. Maximal and steady-state values (under 100h) of hydrogen concentration C_H in components of weld.

Component	$C_{H(max)}$ mol/cm^3	$C_{H(stab)}$ mol/cm^3	$\frac{C_{H(stab)}}{C_{H(max)}}$
Base metal (BM)	$1.03 \cdot 10^{-5}$	$3.96 \cdot 10^{-7}$	0.038
Weld metal (WM)	$1.15 \cdot 10^{-5}$	$1.06 \cdot 10^{-7}$	0.009
Heat affected zone (HAZ)	$1.36 \cdot 10^{-5}$	$8.17 \cdot 10^{-7}$	0.061

During exposure time, a maximum of hydrogen concentration achieves in sequence: base metal – heat affected zone – weld metal (Table 4). The highest value of $C_{H(max)}$ was recorded for the heat affected zone and lowest for base metal. The steady-state value $C_{H(stab)}$ under $t = 100$ hours remains the highest for heat affected zone and

consequently this component of weld is the most sensitive to hydrogenating. If $C_{H(stab)}$ is considered as some characteristic value reflected the ability to absorb hydrogen then the sensitivity to hydrogenating has the following sequence: heat affected zone – base metal – weld metal. An analysis of the efficiency of hydrogen permeation in metal of weld components was done grounding on the coefficient $k = Q_{abs}/Q_{ev}$. The dependencies of efficiency coefficient k on exposure time have the similar view for all components of weld, namely: after achieving some early maximum k_{max} there is a tendency of monotonic decreasing of this parameter up to some steady-state value k_{stab} , Fig. 5. The dependencies $C_H = f(\tau)$ and $k = f_1(\tau)$ are very similar and values of $C_{H(max)}$ achieve at the time of exposure when $k = k_{max}$. It should be noted that the coefficient k is not considered as solely indicative parameter for direct assessment of absorption capacity of studied weld components. This value rather reflects the dynamics of processes at environment-surface interface, but it may be useful for forecasting of hydrogenating of welds during the long time of pipe operation.

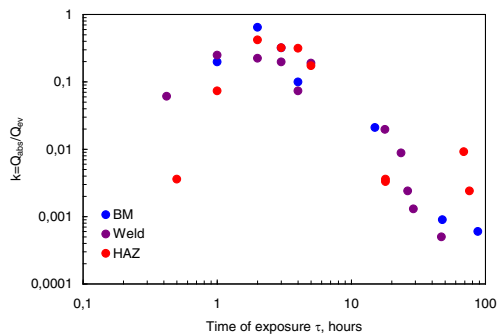


Fig. 5. Dependencies of the efficiency coefficient of hydrogen permeation on exposure time for components of weld.

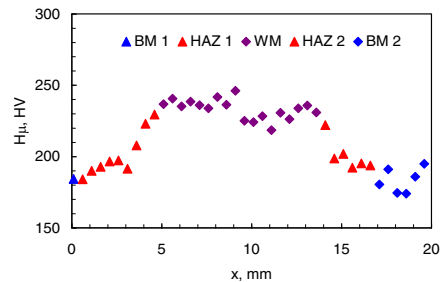


Fig. 6. Variation of the microhardness H_{μ} in the components of weld.

With the aim to find the possible correlation between the ability to absorb hydrogen and mechanical behaviour of the components of weld, the microhardness H_{μ} measurement was made along the welded zone. As it can be seen in Fig. 6 the metal of weld (WM) has the highest value of microhardness and the base metal – the lowest. Based on received data of measurement the average values of parameter H_{μ} were determined for each component of weld. These average values of H_{μ} are given in Table 5.

Table 5. Maximal and steady-state values of hydrogen concentration versus microhardness and yield stress of weld components.

Component	H_{μ} , HV	A , H_{μ}/MPa	σ_Y , MPa	$C_{H(max)}$, mol/cm^3	$C_{H(stab)}$, mol/cm^3
Base metal (BM)	183.4	0.447	410	$1.03 \cdot 10^{-5}$	$3.96 \cdot 10^{-7}$
Weld metal (WM)	233.2	0.447	522	$1.15 \cdot 10^{-5}$	$1.06 \cdot 10^{-7}$
Heat affected zone (HAZ)	200.5	0.447	448	$1.36 \cdot 10^{-5}$	$8.17 \cdot 10^{-7}$

The received above data may also be presented in terms of the engineering yield stress σ_Y , because the linear correlation between parameters H_{μ} and σ_Y is known from literature, Cahoon et al. (1971), Busby et al. (2005) and Gusev et al. (2006) :

$$H_{\mu} \approx A \cdot \sigma_Y \quad (3)$$

where A is some empiric constant. In our case the constant A was calculated from relation (3) for the base metal

inasmuch the value of σ_Y is known. After that it was conditionally assumed that this value of A is the same for other components of weld. On this base the corresponding values of σ_Y were determined for the weld metal and the heat affected zone (see Table 5). Analysis of data this table demonstrate non-monotonic dependence of parameters $C_{H(max)}$ and $C_{H(stab)}$ on the yield stress σ_Y . Despite on higher value of σ_Y , the heat affected zone remains the most sensitive component of weld to hydrogenating. This fact can be taken into account under diagnostics of in service pipelines. As concluding remark it may be pointed out on the following. This study doesn't include the fracture conditions or fracture criteria, which are very important for reliability and fracture risk assessment of the pipelines for transmission of hydrogen-contained agents. The most simple and usable criteria of hydrogen-induced fracture can be presented as some critical combination of hydrogen concentration C_H and applied stress σ , like to: $C_H \cdot \sigma^r = const.$, where r is some constant of material – environment system. In literature a number of versions of such criteria can be found, including the local fracture criteria at defects (cracks, notches, discontinuity of material, etc.). From this point of view presented work can be considered as some step for development of such criteria, because the knowledge about the hydrogen concentration in weld components is very important for this problem.

4. Conclusions

The ability to absorb electrochemical hydrogen by welded pipeline steel API X52 has been studied and the received results have led to the following conclusions. The weldment components of the given steel demonstrate the sensitivity to hydrogenating in deoxygenated, near-neutral pH NS4 solution under cathodic polarisation that is close to the situation of freely corroding system. In general, the efficiency of hydrogen permeation metal is relatively low for all components of weld and after achieving of some early maximum there is the tendency of monotonic decreasing of parameter with exposure time. The ability to absorb hydrogen decreases in the following sequence: heat affected zone – base metal – weld. The sensitivity to hydrogenation is higher for heat affected zone in comparison with base metal and weld. Presented study reflects the hydrogenating conditions, which simulate of real operating pipelines and therefore the received values of parameters may be used as reference data for assessment of hydrogenating of weld components during long-term operation of the considered pipelines.

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