DOI 10.1007/s11003-015-9745-8

Materials Science, Vol. 50, No. 4, January, 2015 (Ukrainian Original Vol. 50, No. 4, July–August, 2014)

METHODS FOR THE EVALUATION OF FRACTURE AND STRENGTH OF PIPELINE STEELS AND STRUCTURES UNDER THE ACTION OF WORKING MEDIA.
PART 2. INFLUENCE OF HYDROGEN-CONTAINING MEDIA

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UDC 620.191.33

We generalize contemporary methods used for the evaluation of the strength and fracture hazard of elements of pipeline structures by using the approaches of fracture mechanics of materials with regard for the specific features of the influence of hydrogen-containing media. We develop the methods and determine the characteristic values of hydrogen concentration in the metal depending on the applied stresses and physicochemical conditions of hydrogenation. On this basis, new methods are proposed for the evaluation of the strength of pipeline steels under the action of working hydrogen-containing media.

Keywords: pipeline structures, low-alloy steels, hydrogen-containing media, static and cyclic loads, hydrogenation of metal, hydrogen concentration, hydrogen embrittlement.

At present, the fact of the diversity of the influence of hydrogen on the mechanical properties of structural metals and alloys depending on the class of materials and conditions of their hydrogenation is generally accepted. This is confirmed by a large body of the available literature data (see, e.g., [1–3]). Nevertheless, there is no clear understanding of the entire spectrum of physical mechanisms of the diffusion of hydrogen into the metal, its concentration near the zones of elevated stress in the metal, and the fracture of metals under these conditions. Therefore, it is important to study the influence of hydrogen in the metal on the strength and serviceability of structural steels under their static and cyclic loading in hydrogen-containing media.

In recent years, in collaboration with other research teams, we performed complex investigations [4–8] of numerous low-alloy steels most often used in the systems of transportation of hydrogen-containing substances. Thus, in particular, we studied pipeline steels of the API class, namely, X52, X70, and X100 steels, and 20 and 16GS ferritic-pearlitic steels also used as materials of pipelines. The tests were carried out under the conditions of electrolytic hydrogenation of the metal in a special NS4 aqueous solution, which is a generally accepted and efficient method [4–6, 9–12]. Note that, in the process of hydrogenation, the specimens were loaded. The level of loading corresponded to the nominal tensile stress $\sigma = \sigma_{\text{exp}}$ formed in the pipe wall in the course of operation under the internal pressure $p_{\text{exp}} = 7$ MPa. The volume concentration of hydrogen in the material was found by using the modified electrochemical approaches [4–6, 12, 13] based on the analysis of oxidation of the hydrogenated metal [14]. In what follows, we present the generalized results of these investigations.

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Translated from Fizyko-Khimichna Mekhanika Materialiv, Vol. 50, No. 4, pp. 7–17, July–August, 2014. Original article submitted December 18, 2013.

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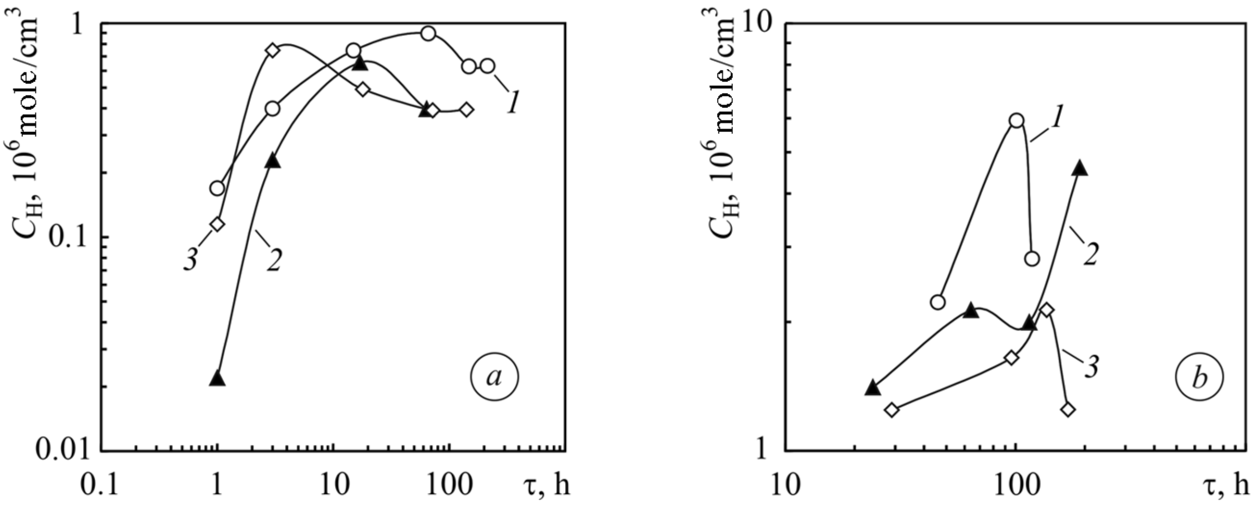

Fig. 1. Hydrogen concentration in the metal depending on the hydrogenation time for unloaded (a) and loaded (b) specimens of X52 (1), X70 (2), and X100 (3) steels.

Table 1. **Maximum Hydrogen Concentration $C_{H(max)}$ and the Time of Its Attainment $\tau_{C_{H(max)}}$**

Steel	Unloaded metal ($\sigma = 0$)		Under loading ($\sigma = \sigma_{exp}$)		$\frac{C_{H(max)}^{stressed}}{C_{H(max)}^{unloaded}}$
	$C_{H(max)}$, 10^6 mole/cm^3	$\tau_{C_{H(max)}}$, h	$C_{H(max)}$, 10^6 mole/cm^3	$\tau_{C_{H(max)}}$, h	
X52	0.895	66	5.924	101	6.62
X70	0.665	17	2.133	64	3.21
X100	0.747	3	2.136	136.5	2.86

Evaluation of the Susceptibility of Pipeline Steels to Hydrogenation

The evaluation of the mean volume hydrogen concentration in the material C_H and its dependence on the time of action of the medium τ and applied mechanical stresses σ_{exp} is of fundamental importance for the subsequent investigation of its strength and service life under the conditions of long-term operation in hydrogen-containing media.

For these types of steel, the efficiency of penetration of hydrogen is quite low and depends on the duration of hydrogenation [4, 15]. In this case, the typical dependences of the concentration C_H on the hydrogenation time τ are not monotonic (Fig. 1). The concentration of hydrogen in the metal first attains its maximum and then decreases with time. Moreover, the time $\tau_{C_{H(max)}}$ differs for different steels. In the process of hydrogenation of unloaded specimens $(\sigma = 0)$, the value $C_{H(max)}$ is attained in the following sequence: X100–X70–X52 (Fig. 1a); for loaded specimens $(\sigma = \sigma_{exp})$, this value is attained in the sequence X70–X52–X100 (Fig. 1b). The values of $C_{H(max)}$ and $\tau_{C_{H(max)}}$ are presented in Table 1.

Within the range $\tau \le \tau_{C_{\mathrm{H(max)}}}$, the rate of hydrogen absorption by steels can be evaluated according to the slopes of the curves $C_{\mathrm{H}} = f(\tau)$. The unloaded specimens of X52 steel have the lowest initial rate of absorption of hydrogen, and the unloaded specimens of X100 steel have the highest initial rate of hydrogen absorption. The loaded specimens of X70 steel are characterized by the highest initial rate of hydrogen absorption. On the other hand, the smallest time of attainment of the maximum hydrogen concentration is observed for unloaded X100 steel and loaded X70 steel.

The presented results reveal the difference between the processes running on the "medium–metal" interface for different types of steel caused by their microstructural features and chemical compositions [4–6, 15, 16], as well as a significant difference between the values of $C_{\mathrm{H(max)}}$ (Table 1). In general, this trend is expected and known from the literature. However, our data are of practical importance because they reproduce the influence of operating stresses for the analyzed pipes of standard sizes in the case where the internal pressure is given. It is shown that the absorption of hydrogen in X52 steel is especially sensitive to the applied stresses. In this case, $C_{\mathrm{H(max)}}$ becomes more than six times higher as compared with the unloaded metal. This fact should be taken into account in the development of new efficient methods for the evaluation of the serviceability of pipelines aimed at the transportation of hydrogen or hydrogen-containing substances.

Table 2. Values of Constants in Relation (1)

Steel	Unloaded metal ($\sigma = 0$)		Under loading ($\sigma = \sigma_{\mathrm{exp}}$)	
	A	m	A	m
X52	0.253	0.24	0.30	0.57
X70	0.049	0.67	0.40	0.42
X100	0.200	0.19	0.80	0.13

Table 3. Hydrogen Concentration in Steels Depending on Their Mechanical Properties (τ = 200 h)

Steel	σ_{y}	σ_{u}	C_{H}, 10^6 mole/cm^3
	MPa		
X52	410	528	6.14
X70	590	712	3.70
X100	866	890	1.59

In our opinion, the comparative evaluation of the susceptibility of these steels to hydrogenation should be performed within the range $0 \le \tau \le \tau_{\max}$ when the "virgin" state of the metal surface is still preserved for a certain time. Under these conditions, X100 steel absorbs hydrogen most efficiently. This fact is quite important for the investigation of hydrogenation of the metal from the "fresh" surface, e.g., in the course of development of cracklike defects. In this case, a substantial acceleration of crack growth is expected in X100 steel.

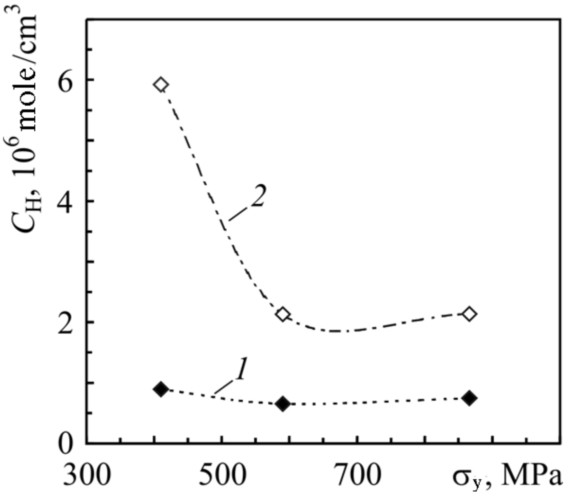

Fig. 2. Dependences of the maximum hydrogen concentration in X52, X70, and X100 steels on their yield strength σ_y: (1) $\sigma = 0$; (2) $\sigma = \sigma_{\exp}$.

For long-term holding, i.e., within the range $\tau \geq \tau_{C_{\mathrm{H(max)}}}$, the increase in the hydrogen concentration in the metal depending on the time of holding of specimens under the conditions of hydrogenation can be described by the following power dependence [6, 15]:

$$C_{\mathrm{H}} = A \cdot 10^{-6} \cdot \tau^{m} \quad [\mathrm{mole/cm^3}], \tag{1}$$

where A and m are constants of the material–medium system (Table 2).

According to the susceptibility to hydrogenation under these conditions, the investigated steels can be arranged in the following order: X52–X70–X100 (Table 3). In other words, their ability to absorb hydrogen is weakened in passing from the pearlitic–ferritic structure (X52) to the structure of polygonal ferrite (X70), and then to the ferritic–bainitic structure (X100).

Figure 2 illustrates the results of comparative evaluation of the susceptibility of X52, X70, and X100 steels to hydrogenation. It is easy to see that the difference between the values of $C_{\mathrm{H(max)}}$ for the unloaded metals is almost absent, although its value for X52 steel is somewhat higher than for X70 and X100 steels. However, for $\sigma = \sigma_{\exp}$, the hydrogenation resistance of X52 steel strongly decreases, whereas X70 and X100 steels resist to hydrogen absorption almost identically; however, the resistance of X100 steel to hydrogen absorption is somewhat higher. Thus, the hydrogenation resistance of the analyzed steels becomes lower as the yield strength of the material decreases.

It should be emphasized that the presented results were obtained under conditions very close to the actual operating conditions, which makes them suitable for the development of new criteria aimed at the evaluation of strength and serviceability of pipelines.

Influence of Hydrogen on the Fracture and the Local Strength of Steels near Defects (Stress Concentrators)

To guarantee the long-term integrity of hydrogen-transportation systems and determine the fracture hazard, it is necessary to know the local characteristics of strength of the materials near model defects, i.e., stress concen-

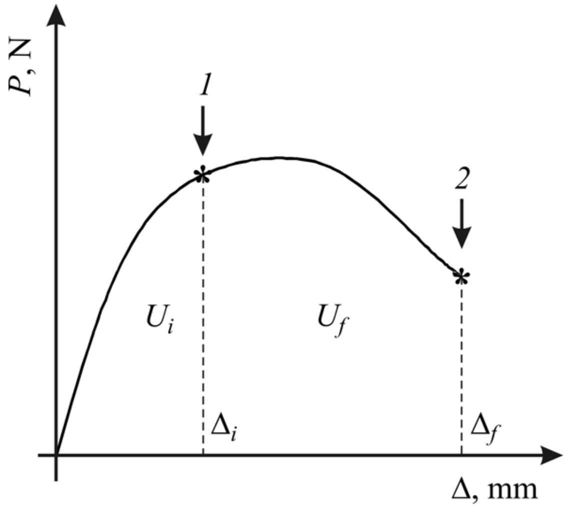

Fig. 3. Schematic diagram of evaluation of the parameters U_i and U_f: (1) onset of fracture (according to the AE-signal); (2) fracture of the specimen.

trators, in the presence of hydrogen. As an effective characteristic of the material, one can use the work of its local fracture near a notch for different bulk concentrations of hydrogen [5, 15]. On this basis, the corresponding dependences of the work of local fracture on hydrogen concentration are constructed for a given material–medium system. The corresponding experimental results are obtained as follows: After holding under given conditions of hydrogenation for time τ (for which the bulk concentration of hydrogen in the metal becomes equal to $C_{\mathrm{H}(\tau)}$), the specimens with defects, i.e., stress concentrators, are tested to fracture under monotonically increasing static loading. In computer-aided investigations, a load–displacement diagram is plotted and acoustic-emission (AE) signals formed in the process of deformation of the specimen are recorded. The onset of local fracture is detected by the AE method [5, 15]. The load–displacement diagrams are used to determine (Fig. 3) the work of the onset of local fracture in the material near a stress concentrator U_i and the total work of fracture of the specimen with this concentrator U_f [5, 15]:

$$U_i = \int_0^{\Delta_i} P(\Delta)\, d\Delta\,, \tag{2}$$

$$U_f = \int_0^{\Delta_f} P(\Delta)\, d\Delta\,. \tag{3}$$

The hydrogen concentration in the metal C_{H} is found by using relation (1) (Table 4).

Hence, (Fig. 4), there exists a certain critical time of hydrogenation of steels for which the corresponding critical concentration C_{H}^* is attained as a result of which their fracture resistance strongly decreases. Its value can be found from the dependences $U_i = f(C_{\mathrm{H}})$ and $U_f = f(C_{\mathrm{H}})$. It should be noted that they coincide (Table 5) because the parameter U_f can be regarded as characteristic for this class of steels under the indicated physicochemical conditions of hydrogenation.

Table 4. Values of the Parameters U_i and U_f for Steels Hydrogenated for Time τ

τ, h	U_i	U_f	C_H, 10^6 mole/cm^3
	N·m		
X52 steel			
0	27.23	226.47	0
46	32.12	170.84	2.660
101	26.57	126.33	4.165
118	15.53	132.98	4.551
167.3	10.47	124.90	5.553
X70 steel			
0	17.64	145.46	0
24	12.68	136.26	1.52
64	6.88	142.62	2.294
114.5	11.41	135.59	2.929
190	15.25	129.00	3.623
X100 steel			
0	119.00	318.00	0
29	114.75	312.63	1.239
96	102.86	296.78	1.448
136.5	64.29	258.46	1.516
169	12.72	241.77	1.558

Table 5. Evaluation of the Critical Concentration of Hydrogen C_H^* for Pipeline Steels

Steel	C_H^*, 10^6 mole/cm^3	
	from U_i	from U_f
X52	4.3	4.3
X70	2.3	2.3
X100	1.5	1.5

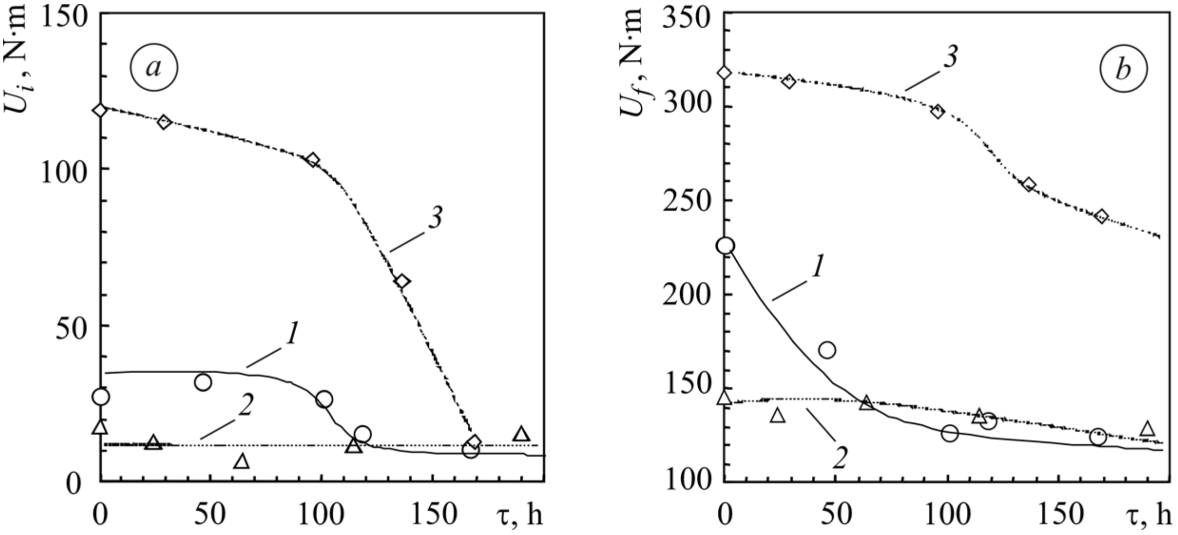

Fig. 4. Dependences of the parameters U_i (a) and U_f (b) for X52 (1), X70 (2), and X100 (3) steels on the duration of hydrogenation of specimens.

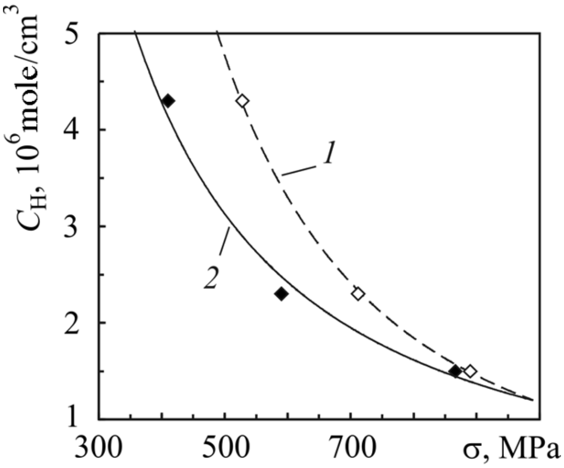

Fig. 5. Dependences of the critical hydrogen concentration C_H^* in the metal on the yield strength σ_y and the ultimate strength σ_u : (1) $C_H^* = 10^6(\sigma_u)^{-2.0212}$; $R^2 = 0.9994$; (2) $C_H^* = 19486(\sigma_y)^{-1.4058}$; $R^2 = 0.9848$.

Thus, among other important engineering parameters, the quantity C_H^* can be recommended for the evaluation of the reliability and service life of hydrogen transportation pipelines made of X52, X70, and X100 steels. By using this parameter, we performed a comparative analysis of the influence of hydrogen on the local strength of steels near stress concentrators. Note that the term “critical concentration” is often used in hydrogen materials science but its physical meaning is different in different works. In our case, this is the concentration of hydrogen in the metal upon attainment of which the local fracture resistance of the material near the notch strongly decreases.

The comparative evaluation of the parameter C_H^* for the investigated steels revealed the following fact: There exists a trend of its monotone decrease with the increase in the yield strength σ_y and ultimate strength σ_u

of the material (Fig. 5). These regularities can be described by power functions of the form [5]

$$C_{\mathrm{H}}^{*} = \frac{A_1}{(\sigma_{\mathrm{y}})^{n_1}}, \tag{4}$$

$$C_{\mathrm{H}}^{*} = \frac{A_2}{(\sigma_{\mathrm{u}})^{n_2}}, \tag{5}$$

where A_1, A_2, n_1, and n_2 are materials constants under given testing conditions. Here, it is worth noting that, for the indicated steels, the standard deviation is high ($R^2 = 0.98$–0.99), which confirms the reliability of the description of the obtained results by using relations (4) and (5):

$$C_{\mathrm{H}}^{*}(\sigma_{\mathrm{y}})^{n_1} = A_1 = \mathrm{const}, \tag{6}$$

$$C_{\mathrm{H}}^{*}(\sigma_{\mathrm{u}})^{n_2} = A_2 = \mathrm{const}. \tag{7}$$

In other words, there exists a certain critical combination of the parameters C_{H}^{*} and σ_{y} (or σ_{u}).

For this class of steels, in the first approximation, we can assume that

$$C_{\mathrm{H}}^{*} \approx \frac{1}{\sigma_{\mathrm{u}}^{2}}. \tag{8}$$

Dependence (8) can be recommended for engineering estimates in choosing the grade of pipeline steel in the initial stages of design of new pipelines or for the replacement of worn-out sections of the existing pipelines.

Evaluation of the Hydrogen Concentration Near Cracklike Defects and the Cyclic Crack-Growth Resistance of Materials in Hydrogen-Containing Media

The results presented above are restricted to the determination of the fracture resistance of steel specimens with stress concentrators modeling mechanical defects, such as notches, scratches, and dimples with rounded tips. For deeper understanding of the influence of hydrogen on the service life of structural elements of hydrogen power infrastructure under the actual operating conditions, it is also necessary to have the data on hydrogen-assisted crack propagation because we know, e.g., the cases of violation of the integrity of pipelines as a result of subcritical growth of cracklike defects [17, 18].

For this purpose, in collaboration with the Paton Institute of Electric Welding of the Ukrainian National Academy of Sciences, we performed a complex of physicomechanical investigations [7, 8] with an aim to experimentally determine the relationship between the bulk concentration of hydrogen in the metal and its local concentration near the tips of sharp stress concentrators (cracks) and the parameters characterizing the development of cracks in 16GS low-alloy pipeline steel under cyclic loading in hydrogenating media of different compositions.

To determine the hydrogen concentration near the crack tip, a special method of local laser microprobe mass spectrometry was used in [7, 8]. The procedure of investigations was as follows:

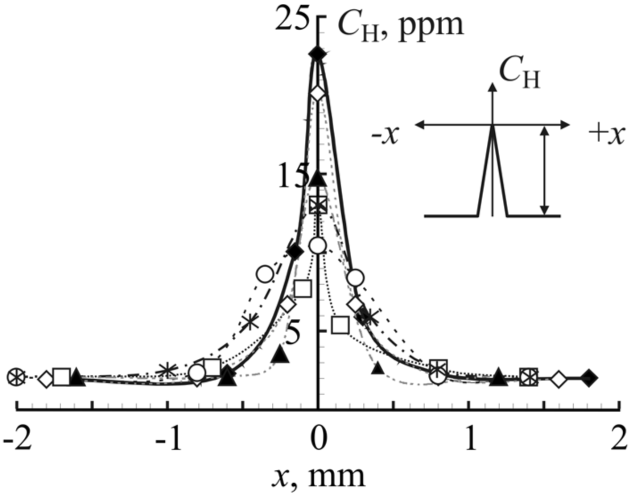

Fig. 6. Distributions of the local hydrogen concentration near the tip of a fatigue crack for different lengths a in 16GS low-alloy steel (bulk concentration of hydrogen of the metal $C_{H(v)}$ = 1.97 ppm; scanning was performed in the direction perpendicular to the plane of crack propagation at a distance of 0.1 mm from the crack tip): (◆) a = 3.18 mm; (◇) 4.78 mm; (▲) 7.53 mm; (□) 8.58 mm; (∗) 9.98 mm; (○) 12.53 mm.

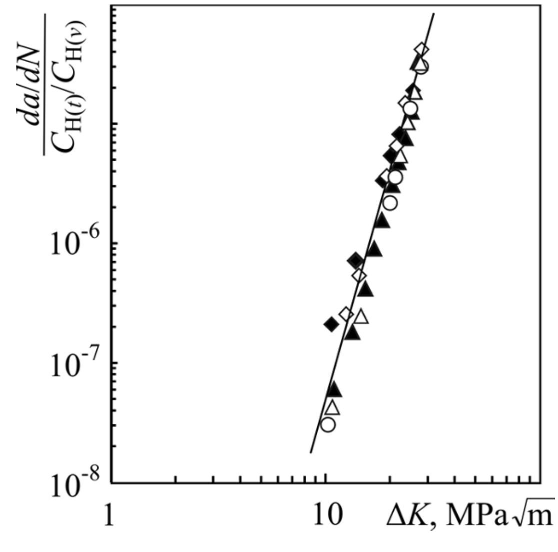

Fig. 7. Relationship between the crack growth rate, local hydrogen concentration at the crack tip, mean hydrogen concentration in the bulk of the metal, and the stress intensity factor [steel 16GS, $f = 1$ Hz, $R = 0$, H_2O + HCOOH; the line is described by relation (9)]: (◆) $C_{H(v)}$ = 1.97 ppm; (◇) 2.07 ppm; (▲) 2.20 ppm; (△) 2.47 ppm; (○) 2.68 ppm.

As soon as the length of a crack in a cyclically deformed hydrogenated specimen attained a certain prescribed value a, the test was terminated, and the local hydrogen concentration in the vicinity of the crack tip was found. In this case, scanning was performed starting from a distance of 0.1 mm from the crack tip both in the direction of crack propagation and in the direction perpendicular to the plane of crack propagation. It was shown that, for both directions of scanning, the hydrogen concentration in the metal abruptly decreases with the distance from the crack tip and approaches a certain value corresponding to the concentration of hydrogen in the bulk of the metal under the actual testing conditions (Fig. 6).

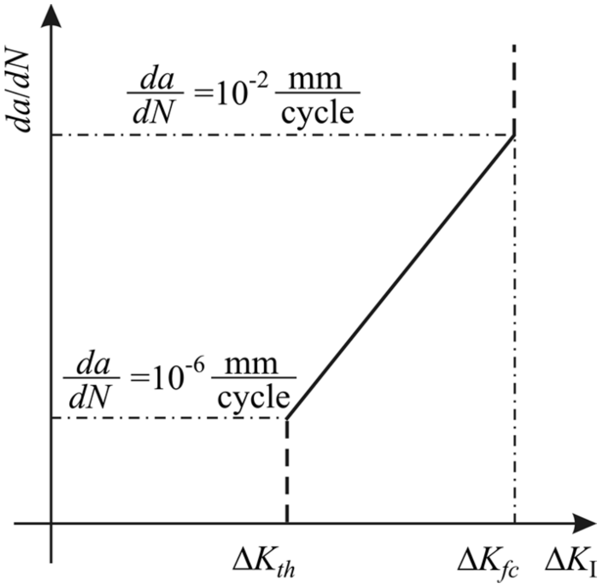

Fig. 8. Fatigue crack-growth diagram for pipeline steels in hydrogen-containing media; the solid line is described by relation (9); $\Delta K_{\mathrm{th}} = \Delta K_{da/dN=10^{-6}\,\mathrm{mm/cycle}}$; $\Delta K_{\mathrm{fc}} = \Delta K_{da/dN=10^{-2}\,\mathrm{mm/cycle}}$.

On the basis of the results of these tests, the relationships between the fatigue-crack growth rate da/dN and the range of the stress intensity factor (SIF) ΔK at the crack tip, between the local hydrogen concentration near the tip of the fatigue crack $C_{\mathrm{H}(t)}$ and the range of the SIF ΔK, and between the local hydrogen concentration near the tip of the fatigue crack $C_{\mathrm{H}(t)}$ and its growth rate da/dN in steel were established in [7] for five different values of the bulk concentration of hydrogen $C_{\mathrm{H}(v)}$ in 16GS steel.

The results of investigations were generalized in the form of a diagram (Fig. 7) presenting the dependence of the ratio $(da/dN)/(C_{\mathrm{H}(t)}/C_{\mathrm{H}(v)})$ on ΔK on the bilogarithmic coordinates (and showing that the indicated dependence is linear). In this case, the standard deviation r^2 of the experimental data from the analytic straight line is quite large and equal to 0.98. It is worth noting that the obtained diagram is common for all five values (see Fig. 7) of the bulk concentration of hydrogen $C_{\mathrm{H}(v)}$ in this steel.

Thus, the fatigue-crack growth rate da/dN in low-alloy pipeline steels under the conditions of hydrogenation was represented [7] in the form of a function of the local hydrogen concentration near the crack tip, hydrogen concentration in the bulk of the metal, and the range of the SIF caused by the external load

$$\frac{da}{dN} = A\left(\frac{C_{\mathrm{H}(t)}}{C_{\mathrm{H}(v)}}\right)(\Delta K_{\mathrm{I}})^{m}, \tag{9}$$

where $C_{\mathrm{H}(v)}$ and $C_{\mathrm{H}(t)}$ satisfy the following conditions:

$$C_{\mathrm{H}(v)} \neq 0; \qquad C_{\mathrm{H}(t)} \geq C_{\mathrm{H}(v)}. \tag{10}$$

Dependence (9) is an important result of physicochemical mechanics of materials in the field of determination and prediction of the service life of metal structures in hydrogen-containing media.

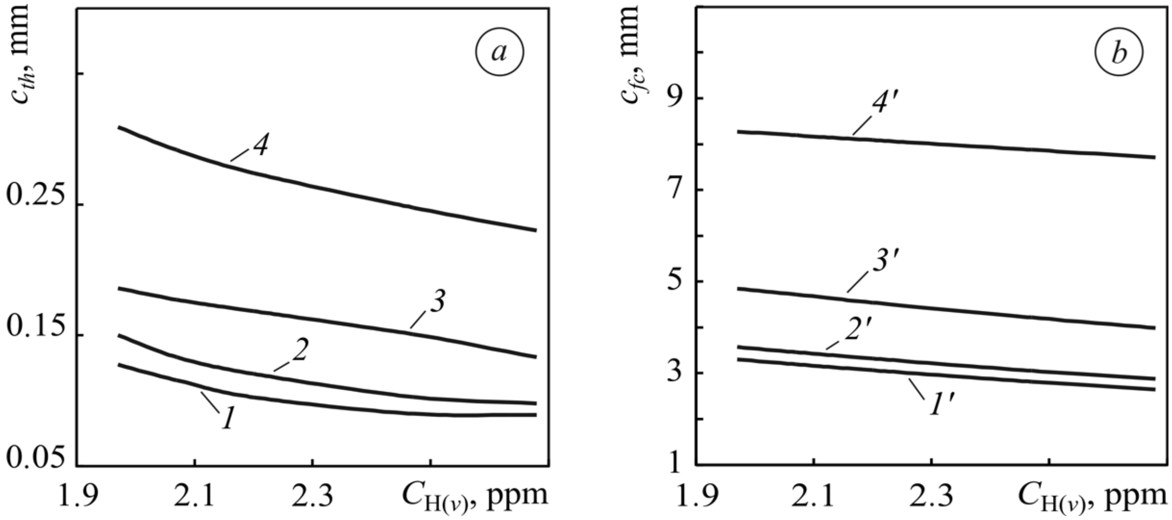

Fig. 9. Influence of the bulk concentration of hydrogen in the metal on the critical sizes of defects c_{th} (a) and c_{fc} (b) in a pipeline with cross section $D = 610$ mm, $t = 11$ mm: (1, 1′) $c/a = 0.01$; (2, 2′) 0.1; (3, 3′) 0.4; (4, 4′) 0.8.

Evaluation of the Serviceability and Fracture Hazard of Pipelines under the Action of Hydrogen-Containing Media. The obtained results can be used as a base for expert estimations of the reliability and the possibility of extension of the service life of pipeline systems with detected defects. By using dependence (22) and the results of experiments [7], we computed the local hydrogen concentration near the crack tip $C_{\text{H(t)}}$ for its different contents $C_{\text{H}(v)}$ in the bulk of the metal and different rates of crack growth da/dN. As basic, we used a simplified fatigue crack-growth diagram for pipeline steels under different conditions of hydrogenation (Fig. 8). The dependences of the threshold SIF K_{th} and critical SIF K_{fc} on the concentration $C_{\text{H}(v)}$ were established. Cracklike defects in a pipeline wall with internal diameter d and thickness t were modeled by a semielliptic crack with semiaxes a and c in size [17].

We also plotted the dependences of the threshold depth of the semielliptic defect c_{th} and the critical depth of the crack c_{fc} for a pipeline with sizes $D = 610$ mm and $t = 11$ mm on the mean hydrogen concentration in the bulk of the metal of the pipe $C_{\text{H}(v)}$ (Fig. 9). These diagrams can be regarded as basic for the evaluation of the serviceability of pipelines and fracture hazard under specific operating conditions and for the differential evaluation of the influence of sizes of the detected defects on the service life of the structure depending on the state of the material of pipeline (degree of its hydrogenation).

On the basis of the accumulated results, we developed an expert computer program for the evaluation of the serviceability and safe operation of defective pipelines. This program can be used for making expert conclusions concerning the fracture hazard for the analyzed structures [17, 19, 20–22].

CONCLUSIONS

We have generalized contemporary methods for the evaluation of strength of the elements of the pipeline structures and fracture hazard by using the approaches of fracture mechanics of materials and taking into account the specific features of the influence of hydrogen-containing media. We have developed a methodology and established the characteristic values of hydrogen concentration in the metal depending on the applied stresses and physicochemical conditions of hydrogenation. A certain critical hydrogen concentration in the metal leading to a strong loss of the local fracture resistance in the material was discovered, and its values were determined for low-alloy pipeline steels. Parallel with the other important engineering parameters, this characteristic value can be recommended for the evaluation of the reliability and service life of hydrogen-transportation pipelines. By using the approaches of fracture mechanics, we propose and justify criteria for the determination of the possibility of safe operation of a pipeline with cracklike defects in hydrogen-containing working media of different compositions. The influence of the bulk concentration of hydrogen in the metal of pipelines with cracklike defects on their strength and crack resistance is established.

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