Investigation of the corrosion process and destruction of metals by using acoustodamage method

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Abstract. The paper presents the results of investigation of acoustic anisotropy in industrial alloy made of steel 14HGNDC after hydrogen-induced cracking (HIC) tests according to the standard NACE TM0284-2003. It was found that location and parameters of corrosion cracks with size about 20 microns can be determined by distribution and value of acoustic anisotropy. A quantitative relationship between value of acoustic anisotropy and size of corrosion cracks in the range from 60 to 6600 microns was established. The obtained results have a great importance for improving methods of hydrogen-induced cracking tests and for non-destructive testing of brittle destruction of structures in oil and gas industry by using the acoustodamage method.

1 Introduction

Corrosion and stress-corrosion processes occurring in structures made of high-strength steels are often accompanied by brittle destruction. Its mechanism is not well understood. The results of recent studies [1-3] indicate that initiation of cracking during external loading occurs in entire volume of the material due to micro- and nano- pores and cracks. Thus, the problem of detecting zones of corrosion cracking of oil tanks, oil and gas pipelines and structures operating in corrosive mediums and sea water on the base of the existing methods of non-destructive testing is difficult. At the same time, the brittle failure of these structures is one of the main causes of accidents in oil and gas industry [4].

Ultrasonic methods [5, 6] have low requirements for the quality of preparation of diagnosed surface in comparison to other methods of non-destructive testing. In addition, they are applicable for diagnosis of structures in corrosive medium and sea water. They include methods based on investigation of the acoustoelastic effect in metals [7–12]. Acoustic anisotropy [13–15] is the relative difference in velocities of propagation of orthogonally polarized transverse waves. It is one of the main characteristics of the acoustoelastic effect.

The use of acoustic anisotropy as an indicator of surface and bulk cracking of metals has a great interest [16–19]. Measurements of acoustic anisotropy are not associated with the detection of additional signal reflected from cracks as in case of ultrasonic flaw detection [20]. It is related to measurements of the phase shift of two transverse waves of orthogonal polarization. The phase shift is caused by defects of arbitrarily small size. It is determined by precision measurements of average time delays between pulses of bulk shear waves. They are repeatedly reflected from the surface of structure opposite to the diagnosed one.

The aim of the work is to investigate the influence of surface and bulk cracking of metals on the value of acoustic anisotropy. The qualitative and quantitative correlations between acoustic anisotropy, cracking and time of corrosion tests of specimens made of industrial 14HGNDC steel alloy in corrosive medium were also investigated.

2 Experimental investigations of acoustic anisotropy during tests on hydrogen induced cracking of metals (HIC)

Tests on hydrogen-induced cracking (HIC) of steel specimens according to the standard [21] were carried out to investigate acoustic anisotropy in industrial alloy before and after corrosion. The specimens were kept in deaerated aqueous medium with 5% by mass of NaCl and 0.5% by mass of CH3COOH during different periods of time. The concentration 1000 mg / l of hydrogen sulfide H2S was provided by the method of bubbling with gaseous hydrogen sulfide.

Weather resistant 14HGNDC steel was selected for corrosion tests due to its cracking after hydrogenation during standard 96 hours [21]. Cracks in specimens were detected by microscopic analysis (in Fig. 1.). Prismatic specimens of size 15x20x100 mm³ (in Figure 2) were made from steel 14HGNDC. Their surface was polished on the machine to the roughness of Rz20.

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At the first stage of research, measurements of acoustic anisotropy in specimens before corrosion tests were carried out at six points at a distance of 12 mm from each other (as shown in Fig. 2). Acoustic anisotropy was measured using IN-5101A certified ultrasonic device. The distance between points is determined by the geometric dimensions of the piezoelectric elements of a standard 5 MHz ultrasonic sensor. Acoustic anisotropy was calculated according to Eqn. (1):

\[ a = \frac{2(t_2-t_1)}{(t_1+t_2)} \]  

where \( t_1 \) and \( t_2 \) are time delays between reflected pulses of two transverse waves of mutually orthogonal polarization.

In the second stage, specimens were kept in corrosive medium. Time intervals of 48, 72, 84, 108, and 152 hours were established in addition to the standard corrosion test time of 96 hours [21].

At the third stage, we measured acoustic anisotropy in specimens after corrosion tests at the same points as before the tests (as shown in Fig. 2). Macrocracks in specimens were detected by additional reflected pulses from shear waves. Changes in the distributions of acoustic anisotropy (as shown in Fig. 5) were the second indicator of metal cracking. The results of ultrasonic measurements in specimen without cracks and in specimen with crack of 1400 microns are presented in Fig.3 and Fig.4.

**Fig. 1.** Macrocra<ref>cks in specimen of steel 14HGNDC after 84 hours of hydrogenation in a corrosive medium.

**Fig. 2.** The geometry of specimens and location of measurement points of acoustic anisotropy.

**Fig. 3.** Reflected impulses in metal without macrocracks.

**Fig. 4.** Reflected pulses in metal with 1400 micron crack after 72 hours of corrosion tests.

At the fourth stage, microscopic investigations were carried out in parts of specimens with cracks detected by ultrasound diagnostics. As a result, dimensions and standard parameters of cracks CSR, CLR, and CTR were determined.

**3 Discussion**

Analysis of the acoustic anisotropy distributions \( a_0 \) before and \( \bar{a} \) after corrosion tests leads to several important conclusions.

It was found that the character of distribution of acoustic anisotropy \( a_0 \) before hydrogen absorption is preserved in case of absence of macrocracks inside the metal. Changes in the integral value of acoustic anisotropy \( \bar{a} \) in this case occur \( \Delta a_{\text{surface}} \) at all points (as shown in Fig. 5). This effect is observed in pure form in zones of specimens without corrosion macrocracks at points 2, 4 and 5 in Fig. 5 and at point 4 in Fig. 6. The average value \( \Delta a_{\text{surface}} \) does not depend on testing time in case of equal concentrations of the corrosive mediums. It is equal to the value \( \Delta a_{\text{surface}} = 0.0255\% \) for all specimens according to results provided by 10 thousand measurements of time delays. The component \( \Delta a_{\text{surface}} \) may be caused by hydrogen cracking of a thin surface layer of metal.

The second effect is correlated with formation of macrocracks in the volume of metal. It also affects the value of acoustic anisotropy. Let us denote the contribution of inner cracking of metal to the value of acoustic anisotropy as \( \Delta a_{\text{bulk}} \). The length of cracks detected during microanalysis is ranged from 113 to 6636 microns. The significant change in the value of acoustic anisotropy \( a_0 + \Delta a_{\text{surface}} \) by the value of \( \Delta a_{\text{bulk}} \) was observed. The example of influence of cracks with a length of 1700 and 700 microns on the value and distribution of acoustic anisotropy is shown in Fig.6.

**Fig. 5.** Acoustic anisotropy \( a_{\%} \) in specimen without macrocracks after 84 hours of corrosion tests.
Acoustic anisotropy $a, \%$ in specimen with 1700 micron and 700 micron cracks after 96 hours of corrosion tests.

It was found that the total change in acoustic anisotropy is caused only by influence of surface and bulk cracks during hydrogen cracking of metal.

$$\Delta a = \Delta a_{\text{surface}} + \Delta a_{\text{bulk}} \tag{2}$$

It is necessary to determine zones without macrocracks in specimens for independent estimating of the value $\Delta a_{\text{surface}}$.

The dependence of average cracks size on testing time of specimens in corrosive medium was obtained on the basis of microscopic studies (Fig. 7).

The dependence $\Delta a_{\text{bulk}}$ of modulus of acoustic anisotropy on length of cracks is shown in Fig. 8. It has a monotonous character (in Fig. 8). It allows to determine cracks size by applying results of acoustic anisotropy measurements.

Conclusions

It was found that change in the value of acoustic anisotropy during tests on hydrogen-induced cracking (HIC) of steel specimens made of 14HGNDC industrial alloy is observed even in case of absence of external loads and plastic deformations. This fact cannot be explained by the existing theory. It requires further research.

It was experimentally found that the location of crack in a specimen can be determined on the base of changes in the distribution of acoustic anisotropy. The value of change of acoustic anisotropy $\Delta a_{\text{bulk}}$ allows us to estimate a size of the crack. The total change $\Delta a$ of the value of acoustic anisotropy appears due to the mutual influence of surface and bulk cracks: $\Delta a = \Delta a_{\text{surface}} + \Delta a_{\text{bulk}}$.

The dependence of acoustic anisotropy on length of cracks in the range from 60 to 6600 microns for specimens made of weather resistant steel 14HGNDC was obtained. It has a monotonous character. It allows to estimate cracks size about $15 \pm 5$ microns.

The contribution of surface microcracks with average size of 50 microns to the value of acoustic anisotropy $a_0$ may be caused by the mutual influence of surface microcracks with length of 50 microns localized on two opposite surfaces of the specimen (in Fig. 8). The increase in testing time of specimens in corrosive mediums of equal concentrations does not lead to the growth of cracked layer above a certain level. This result is confirmed by recent studies of the surface effect of hydrogen concentrations in metals [22–25].

4 Conclusion

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It was experimentally found that the location of crack in a specimen can be determined on the base of changes in the distribution of acoustic anisotropy. The value of change of acoustic anisotropy $\Delta a_{\text{bulk}}$ allows us to estimate a size of the crack. The total change $\Delta a$ of the value of acoustic anisotropy appears due to the mutual influence of surface and bulk cracks: $\Delta a = \Delta a_{\text{surface}} + \Delta a_{\text{bulk}}$.

The dependence of acoustic anisotropy on length of cracks in the range from 60 to 6600 microns for specimens made of weather resistant steel 14HGNDC was obtained. It has a monotonous character. It allows to estimate cracks size about $15 \pm 5$ microns.

The contribution of surface microcracks with average size of 50 microns to the value of acoustic anisotropy is the same for all the studied time intervals. This is due to the fact that increase in testing time does not lead to the growth of damaged layer above a certain level for equal concentrations of corrosive mediums.

The obtained results allow to use measurements of acoustic anisotropy to diagnose corrosion cracking in metals. It can be used to improve the methods of hydrogen-induced cracking (HIC) tests and to diagnose...
the brittle destruction of structures in oil and gas industry. The research is carried out under the financial support by Russian Science Foundation, project 18-19-00413.

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