

Chapter 38

Identification of Zones of Local Hydrogen Embrittlement of Metals by the Acoustoelastic Effect



Kseniya Frolova, Vladimir Polyanskiy, Dmitriy Tretyakov
and Yuri Yakovlev

Abstract In this paper, we present the results of the investigation of statistical correlations between local plastic deformation distributions, the acoustic anisotropy magnitude, and concentrations of hydrogen dissolved in corset samples. Pearson's correlation coefficients with values up to 0.95 were obtained. These coefficients designate the relationship between large hydrogen concentrations in the local areas of the samples and the acoustic anisotropy magnitude. The obtained results make it possible to establish that contribution of concentrations of the dissolved hydrogen to the acoustic anisotropy is much greater than contribution of the nonlinear elasticity (acoustoelastic effect). The obtained data allow one to develop new methods for the destructive control of hydrogen embrittlement.

38.1 Introduction

The share of new materials used in industry is permanently increasing. For example, mass fraction of high-strength steels in the body of a modern car is about 70%. Usually the effect of hydrogen existence on these materials is significant. Mass concentration of hydrogen equal to 0.2 ppm leads to decrease of the tensile strength of new high-strength steels by one and a half to two times [1]. In its turn, this entails a critical effect of hydrogen at a concentration of six atoms per million atoms of the metal matrix.

High hydrogen sensitivity leads to the changing of a role of the hydrogen embrittlement. In traditional metals, it was necessary to accumulate dozens of ppm of hydrogen [2]. The concentrations were equalized in the metal volume due to diffusion [2]. As a rule, large cracks propagated [3].

V. Polyanskiy (✉) · D. Tretyakov
Peter the Great Saint-Petersburg Polytechnic University, St. Petersburg, Russia
e-mail: vapol@mail.ru

K. Frolova · V. Polyanskiy · Y. Yakovlev
Institute for Problems in Mechanical Engineering RAS, St. Petersburg, Russia

The critical concentrations of hydrogen in modern materials are so small that they can accumulate locally. This fact significantly complicates the diagnostics of hydrogen embrittlement. Spectral analysis of ultrasonic signals [4, 5], sound velocity measurement [5] as well as ultrasonic scanning [6] do not have sufficient sensitivity. The hydrogen embrittlement detecting method based on measurement of the absolute sound velocity has another drawback. Namely, this velocity depends on many factors, and therefore in practice it is impossible to separate the influence of temperature, microstructure and hydrogen on its value [7, 8].

Our investigations allow one to consider the acoustoelasticity method [9–11] as one of methods for monitoring the metal damage under loading [12, 13]. An important feature of this method is that the measured data of the value of acoustic anisotropy depend only on the initial anisotropy of the material, mechanical stresses, plastic deformations and damage [11, 13, 14]. We obtained the relation between the acoustic anisotropy magnitude and the residence time of hydrogen charging of the sample in a corrosive solution.

The problem of local nature of the acoustic anisotropy method remains important from practical point of view, since according to the theory of this method [9, 10], the acoustic anisotropy magnitude is affected by the metal mechanical characteristics averaged along the path of the sound wave.

38.2 Research Method

38.2.1 *Acoustic Experiments*

The acoustoelasticity method, based on measurement of the acoustic anisotropy, makes it possible to evaluate mechanical stresses occurring in structures. In this sense, it is an alternative method to the tensometric ones. It is standardized [4] and widely used in modern technical diagnostics [5]. Unlike diagnostic methods using electro-magnetic properties of metals, the method of acoustoelasticity is standardized precisely as a method for monitoring internal mechanical stresses. The method is provided with serial measuring equipment.

In this regard, it is enough to have a reference sample or an unloaded section of a structure made of the same material. A great advantage of this method is the possibility to obtain mechanical stresses averaged along the material thickness. In this case, the thickness of the metal can be 5 mm or more. Thus, it is one more reason indicating that this method advantageously differs from the tensometry, since the last allows one to measure only deformations of the material surface. Measuring sensors are easily installed by using special grease. Measurements can be carried out quickly, in the express mode.

The essence of the method is to measure the relative difference of the propagation velocities of a transverse acoustic ultrasonic wave with different polarizations v_1 and v_2 :

$$\Delta a = 2(v_1 - v_2)/(v_1 + v_2).$$

Parameter Δa is called the acoustic anisotropy.

In the basic work [6], it was theoretically explained and experimentally confirmed that in the case of pure elastic deformations the acoustic anisotropy Δa is proportional to the difference of the principal stresses $\sigma_1 - \sigma_2$.

At the same time, the constructions use metal rolling. Consequently, the metal of the structure is preliminarily subjected to plastic deformation. Plastic deformations contribute to the value of the acoustic anisotropy. In [7–9] it was experimentally obtained that in this case the following expression holds:

$$\Delta a = a_0 + a_1 \cdot (\varepsilon_1^p - \varepsilon_2^p) + C_A(\sigma_1 - \sigma_2), \quad (38.1)$$

where $\varepsilon_1^p, \varepsilon_2^p$ are the principal plastic deformations, σ_1 and σ_2 are the principal stresses, C_A is an experimentally obtained value, $a_0 + a_1(\varepsilon_1^p - \varepsilon_2^p)$ is interpreted as the contribution of the anisotropic microstructure of the metal to the value Δa .

The effect of elastic deformations is described by the term $C_A(\sigma_1 - \sigma_2)$, which is physically related to the displacement of the atoms of the crystal lattice from the equilibrium position under the action of internal mechanical stresses.

According to our investigations [14, 15], the formation of a microcracks system can lead to appearance of an additional contribution to (38.1), which can be rewritten in the following way:

$$\Delta a = a_0 + a_1(\varepsilon_1^p - \varepsilon_2^p) + C_A(\sigma_1 - \sigma_2) + a_2 \quad (38.2)$$

where the term a_2 is associated with the formation of microcracks.

No every plastic deformation leads to metal cracking. At the same time, elastic deformations also are not related for the microcracks formation. So, the term a_2 cannot be included in other components of the equation and must be considered separately.

Acoustic anisotropy was measured by a IN5101A device, which realized the shock generation of ultrasonic waves in the tested body and received the reflected signal by piezotransducers. The three-component piezotransducer radiates and receives one longitudinal wave and two transverse waves polarized in mutually perpendicular directions. Appropriate software permits carrying out precision measurements of the time of propagation of the ultrasonic wave through the body thickness. The time lags between several reflected pulses are measured and averaged to increase the accuracy. The value of the acoustic anisotropy is calculated as the ratio of the difference between time lags of the reflected pulse of polarized transverse wave to the half-sum of these times. In this case, the first wave is polarized along of the line of the load application direction and the second wave is polarized in the perpendicular direction.

We used flat corset samples cut from industrial rolled metal to realize the experiments. Samples from commercially pure aluminum and three grades of steels,



Fig. 38.1 Measurement of acoustic anisotropy: 1—corset sample in the test machine, 2—IN5101A device with piezotransducers

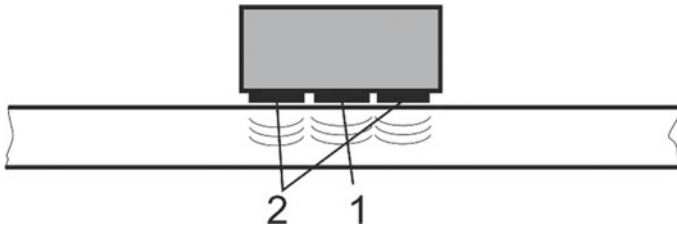


Fig. 38.2 Scheme of the piezotransducers of IN5101A device; 1—piezotransducer of a longitudinal wave, 2—piezotransducers of transverse waves

namely, A192, 5L Gr X-6, F-522, VNS-5 were investigated. The thickness of the samples is varied from 5 to 20 mm. Measurement of acoustic anisotropy is shown in Fig. 38.1.

In case of standard measurements, plates emitting three different types of waves are spaced approximately one centimeter apart, as shown in Fig. 38.2.

It makes difficult to look for local anomalies with the magnitude of the acoustic anisotropy. To increase the resolution, the device with the piezotransducers moved

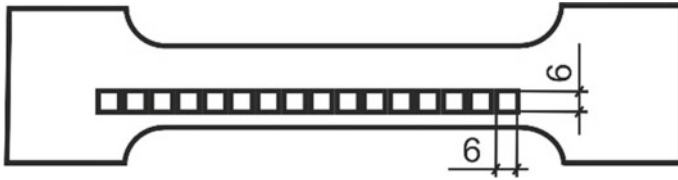


Fig. 38.3 Scheme of the samples cutting for hydrogen testing

along a special grid, as shown in Fig. 38.1. The grid spacing was the same as the spacing between the piezoelectric plates. Thus, resolution for detection of local anomalies of the order of 10 mm was obtained by data imposing measured for different positions of the piezotransducers.

38.2.2 *Hydrogen Experiments*

We carried out different mechanical tests for corset samples, namely, static stretching and cyclic loading. Measurement of hydrogen concentration was carried out by using the standard industrial mass spectrometric analyzer of hydrogen AB-1. The analyzer operation is based on the Vacuum Hot Extraction (VHE) method [16, 17]. The measurement process is described in detail in [18, 19].

In order to measure the volumetric distribution of hydrogen concentrations in corset samples, they were cut into small prismatic samples with a cross-section equal to $6 \times 6 \text{ mm}^2$. The cutting scheme is shown in Fig. 38.3.

The samples cutting was performed by a handsaw, so that the metal did not overheat and the hydrogen concentration was not changed.

A mass spectrometric analyzer of hydrogen allows one to realize measurements at various temperatures of hot vacuum extraction. In terms, it allows one to specify several temperature points for Vacuum Hot Extraction. Such points make it possible to separate hydrogen with different binding energies and to isolate diffusively-mobile hydrogen from the entire volume of hydrogen dissolved in the sample.

38.2.3 *Mechanical Testing*

Mechanical tests were carried out by applying the INSTRON machines.

One of the machines is present in Fig. 38.1. The load parameters were chosen in such a way that plastic deformation of the samples was occurring during the whole loading or one loading cycle. Some samples were loaded up to destruction, whereas some of them were not destroyed. Measurements of the distribution of the acoustic

anisotropy were made directly under the loading and after the destruction of the samples.

Measurements of the hydrogen concentration distribution were carried out for unloaded samples after measurements of the acoustic anisotropy.

In addition to the distribution of the acoustic anisotropy and hydrogen concentration, the thickness of the sample was measured in the site of location of the sensor. In case of the plastic flow, the thickness of the sample varies non-uniformly and the characteristic Lüder's bands are observed. Within these bands the deformation value is much bigger than the average one. The measurements were realized by an electronic micrometer with accurate to 0.01 mm.

38.3 Results and Discussion

The acoustic anisotropy distributions along the sample, made of technically pure aluminum and underwent significant plastic deformation equal to 28%, were obtained. Herewith, an appreciable non-uniformity of the plastic deformation magnitude was observed. Lüder's bands observable on the surface had a width of about 2–3 cm and were oriented perpendicular to the tensile forces applied to the specimen.

Distribution of the acoustic anisotropy in the sample is shown in Fig. 38.4. Values of the acoustic anisotropy were measured for two or three points placed at the same distance x from the origin of the working part of the sample.

Distribution of the thickness of the sample is shown in Fig. 38.5. The corresponding measurements were made in the middle of the site on which the acoustic anisotropy sensor plate was positioned.

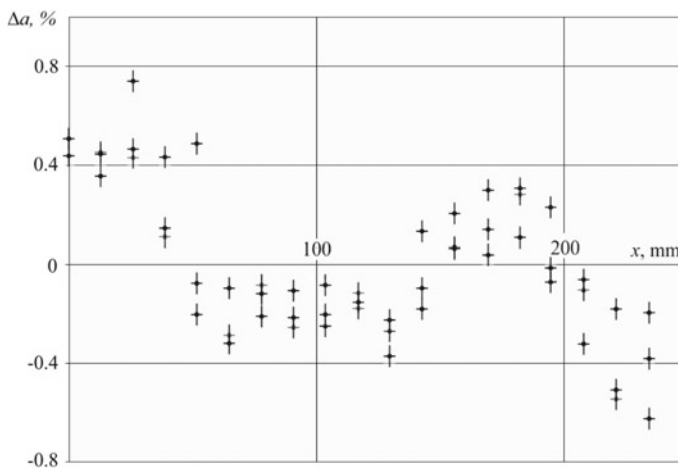


Fig. 38.4 Acoustic anisotropy versus coordinate x along the sample

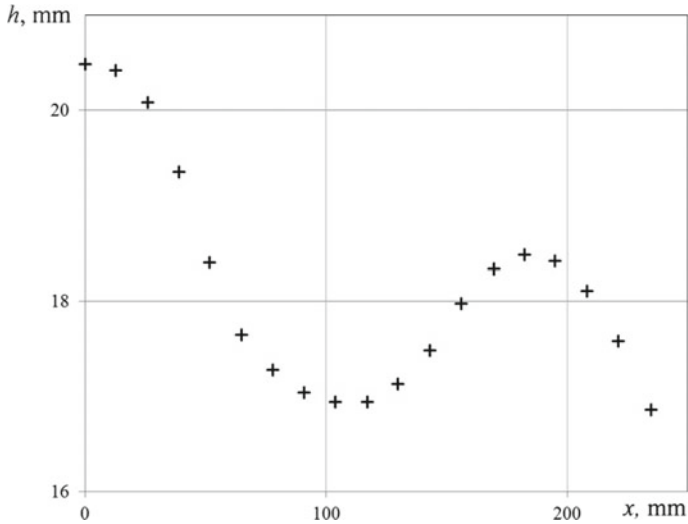


Fig. 38.5 Thickness of the sample after plastic deformation versus coordinate x along the sample

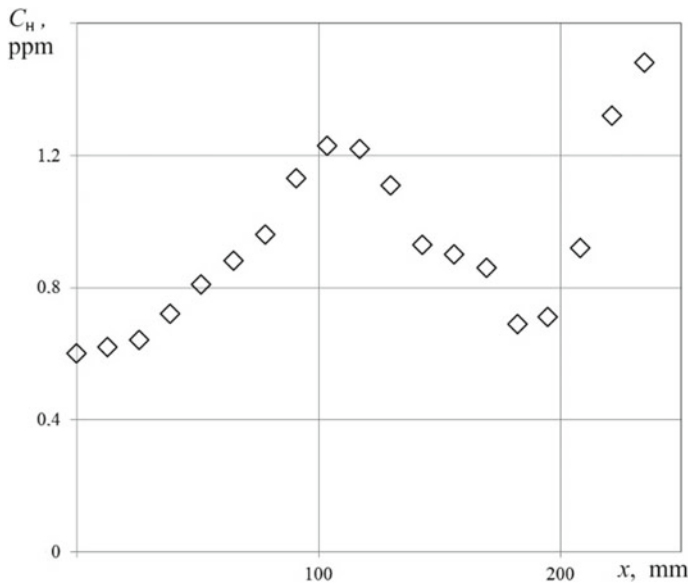


Fig. 38.6 Average concentration of dissolved hydrogen after plastic deformation versus coordinate x along the sample

Distribution of the average hydrogen concentrations in the same sample is shown in Fig. 38.6.

Pearson's correlation coefficients between the acoustic anisotropy and the sample thickness and between the acoustic anisotropy and hydrogen concentration are 0.83 and 0.79, respectively. In its turn, this makes it possible to assume a linear correlation of these quantities.

Similar results characterized by a correlation coefficient ranging from 0.72 to 0.78 were obtained for various steel samples.

It turned out that correlation between distribution of the acoustic anisotropy and distribution of the average concentration of dissolved hydrogen is better in case of aluminum alloys. For several samples, the correlation coefficient reached a value of 0.95.

Comparison of hydrogen concentrations before and after the plastic deformation showed that during the plastic deformation the average concentration of dissolved hydrogen increases.

We conducted an additional investigation. We manually ground the surface layer of the experiment samples to a depth of the order of 0.5–1 mm for aluminum samples and 60–100 μm for steel ones before measuring of hydrogen concentration. Removal of the surface layer led to the average hydrogen concentration decrease, as well as to the correlation coefficient drop to values of the order of 0.07–0.1.

Thus, an almost linear relationship between the hydrogen concentration in the surface layer and the value of the acoustic anisotropy takes place.

We would like to note that the acoustic anisotropy variations caused by the saturation of the thin surface layer of the metal with hydrogen are significantly (from 2 to 6 times) greater than the variations associated with the direct acousto-elastic effect arising due to mechanical stresses.

This makes it possible to establish reliable correlation dependencies between local areas of a metal with an increased hydrogen concentration and leaps observed for the acoustic anisotropy distribution over the metal surface.

This important result is in some measure confirmed by the data presented in [7]. In contrast with these findings, our measurements made for a hydrogenated material allow us to obtain the magnitude of the acoustic anisotropy, which is less subjected to temperature fluctuations and practically unrelated to the metal heat treatment method, as in case of large plastic deformations. Our results makes it possible to indicate the range of this magnitude variation associated with mechanical stresses for whole groups of metals [15, 20].

38.4 Conclusion

Multipurpose investigations consisting of mechanical tests, measurements of the acoustic anisotropy magnitude distribution and measurement of the distribution of hydrogen concentrations in metallic samples after mechanical testing were performed. We got a set of distributions of quantities along the axis of mechanical loading of the sample.

Statistical analysis of Pearson's correlation coefficients corresponding to the obtained distributions allows one to conclude that the value of the acoustic anisotropy correlates well with the concentration of hydrogen dissolved in a thin surface layer of the samples.

The correlation coefficient of several samples takes the value 0.95. This fact indicates availability of a linear dependence of the magnitude of acoustic anisotropy on the local hydrogen concentration. This dependence makes it possible to perform non-destructive testing of local hydrogen embrittlement by the acoustoelasticity method.

Acknowledgements The financial support by the Siemens scholarship program is acknowledged.

References

1. A.N. Solovyev, Y. Nie, Y. Kimura, T. Inoue et al., *Metall. Mater. Trans. A* **43**(5), 1670 (2012)
2. J.P. Hirth, *Metall. Trans. A* **11**(6), 861 (1980)
3. J.E. Costa, A.W. Thompson, *Metall. Trans. A* **13**(7), 1315 (1982)
4. S.E. Krüger, J.M.A. Rebello, P.C. De Camargo, *NDT & E Int.* **32**(5), 275 (1999)
5. P. Senior, J. Szilard, *Ultrasonics* **22**(1), 42 (1984)
6. J. Kittel, V. Smanio, M. Fregonese, L. Garnier, X. Lefebvre, *Corros. Sci.* **52**(4), 1386 (2010)
7. A.M. Lider, V.V. Larionov, G.V. Garanin, M.K. Krening, *Tech. Phys.* **58**(9), 1395 (2013)
8. S.K. Lawrence, B.P. Somerday, M.D. Ingraham, D.F. Bahr, *Ultrasonic methods. JOM* **1** (2018)
9. D.S. Hughes J.L. Kelly, *Phys. Rev.* **92**(5), 1145 (1953)
10. M. Hirao, Y.H. Pao, *J. Acoust. Soc. Am.* **77**(5), 1659 (1985)
11. Y.H. Pao, *Solid Mechanics Research for Quantitative Non-destructive Evaluation* (Springer, Netherlands, 1987), p. 257
12. A.K. Belyaev et al., *Mech. Solids* **51**(5), 606 (2016)
13. A.K. Belyaev et al., *Procedia Struct. Integrity* **6**, 201 (2017)
14. E.L. Alekseeva, et al., *Stroitel'stvo Unikal'nyh Zdanij i Sooruzenij* **12**, 33 (2016) (In Russian)
15. E.L. Alekseeva et al., *AIP Conf. Proc.* **1915**(1), 030001 (2017)
16. E. Petushkov, A. Tserfas, T. Maksumov, in *Secondary Emission and Structural Properties of Solids*, ed. by U. Arifov (Springer US, 1971), p. 107
17. J. Konar, N. Banerjee, *NML Tech. J.* **16**(1–2), 18 (1974)
18. A. Polyanskiy, V. Polyanskiy, Y.A. Yakovlev, *Int. J. Hydrogen Energy* **39**(30), 17381 (2014)
19. A. Polyanskiy, D. Popov-Diumin, V. Polyanskiy, in *Hydrogen Materials Science and Chemistry of Carbon Nanomaterials*, ed. by T. Veziroglu, S. Zaginaichenko, D. Schur, B. Baranowski, A. Shpak, V. Skorokhod, A. Kale, NATO Security through Science Series A: Chemistry and Biology (Springer, Netherlands, 2007) p. 681
20. A.G. Lunev, M.V. Nadezhkin, S.A. Barannikova, L.B. Zuev, *AIP Conf. Proc.* **1909**(1), 020121 (2017)