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Hydrogen embrittlement of welded joints for the heat-treatable XABO 960 steel heavy plates

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Properties

ABSTRACT

Purpose: In the paper, influence of hydrogen on mechanical properties of welded joints from heat-treatable structural XABO 960 steel plates was investigated.
Design/methodology/approach: The heat treatment of welded plate specimens was performed, and then the specimens were charged with hydrogen electrolytically generated from 1 N H_2SO_4 solution. The following studies were carried out: static tensile test, hardness investigations, macroscopic metallographic investigations as well as investigations with the use of a scanning microscope.
Findings: Hydrogen embrittlement of welded joints from XABO 960 steel plates was revealed by a distinct decrease of ductility and a slight decrease of strength. On the basis of metallographic investigations, it was found that in a fracture region there are fine pores created by the presence of hydrogen and its displacement due to formed stresses and plastic deformation. It was shown that welded joints are susceptible to hydrogen cracking in the heat affected zone and in the fusion zone.
Research limitations/implications: TEM investigations on structure of the steel were predicted.
Practical implications: The obtained results can be used for searching the appropriate way of improving the hydrogen embrittlement resistance of welded joints of the heat-treatable structural XABO 960 steel plates.
Originality/value: The hydrogen embrittlement of welded joints of the heat-treatable XABO 960 steel plates was investigated.
Keywords: Mechanical properties; Hydrogen embrittlement; Thick plate; Welded joint

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1. Introduction

HSLA type (High Strength Low Alloy) weldable constructional C-Mn steels, containing microadditions with strong chemical affinity for both carbon and nitrogen, i.e. Nb, Ti and V in an amount of up to 0.1%, sometimes with increased concentration of N and in case of heat-treatable steels - also B, meet in a wide range requirements expected from modern constructional materials. Metallic elements interacting with C and N form MX (M – Nb, Ti, V; X – N, C) stable interstitial phases with cubic lattice of NaCl type, allowing for formation of a stable fine-grained microstructure that decides about high mechanical properties of metallurgical products [1-6].

Weldable metal plates with high strength and desired crack resistance in a quenched and tempered state are produced from steels with reduced concentration of P and S impurities and with a limited portion of modified non-metallic inclusions. They are used for construction of highly loaded welded structures, particularly for wheeled cranes and road machines, heavy transport vehicles, high load capacity overhead travelling cranes, machines and devices for extractive industry and others. Initially, this type of plates with $YS_{0.2}$ ranging from 550 to 960 MPa and $KV_{-40\,^{o}C} > 27$ J was made from heat-treatable steels containing ≤0.2% C, ≤ 1.6% Mn, ≤ 0.8% Si, ≤ 1% Cr, ≤ 2% Ni, ≤ 0.6% Mo and ≤ 0.1% V. High value of carbon equivalent (C_E) for these steels equal from 0.72 to 0.82% created the necessity of preliminary preheating of components connected through welding to the temperature even higher than 200°C. It exerts a significant influence on the costs of produced welded structures.

Introduction of B, Nb, Ti and V microadditions into discussed group of steels has an essential impact on reduction of C_E carbon equivalent value at the simultaneous increase of mechanical properties of metal plates in the quenched and tempered state. It means that the liquid steel should be properly desulfurized and deoxidized using Al prior to the introduction of microadditions; it should also contain Ti (less often Zr) as a boron shielding, counteracting bounding of this element to oxygen and nitrogen. The microaddition of boron in an amount of 0.002 to 0.005% dissolved in a solid solution results in displacement of supercooled austenite transformation curves to longer times (Fig. 1) and also in increase of hardenability of fine-grained steels as a result of reduction in austenite grain boundaries energy caused by segregation of B atoms.

Chemical composition, %						
C	Mn	N	Cr	Mo	Ti	B
0.17	0.85	0.005	0.30	0.35	0.030	0.0016

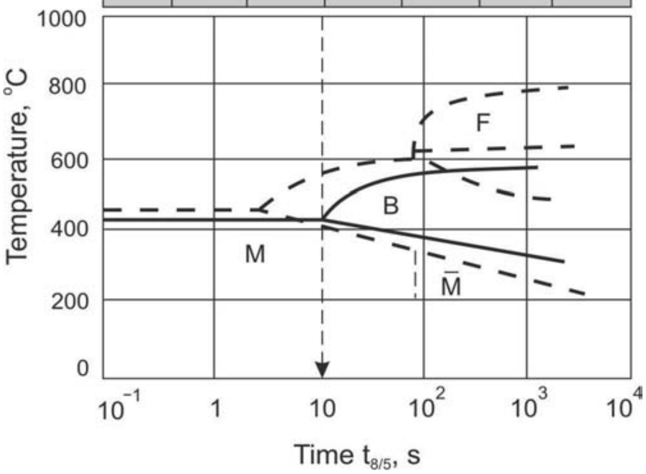

Fig. 1. The TTT-S diagram showing the influence of boron micro-addition on the curves of initiation of undercooled austenite transformation of a Mn-Cr-Mo-Ti steel as a function of the $t_{8/5}$ time: broken line - steel without boron content, solid line - steel with boron content, the arrow presents the cooling time of a 20 mm thick plate [7]

If the liquid steel is not properly deoxidized, also B_2O_3 is formed together with Al_2O_3 and in the solid state – AlN and BN stable nitrides; the part of boron bounded to oxygen and nitrogen is practically eliminated from having any impact on hardenability of steel. After nitrogen is used up, the residual portion of boron forms $M_{23}(C, B)_6$ carboborides in austenite [8-10]. The rest of microadditions, i.e. Ti, Nb and V form stable nitrides, carbonitrides and carbides which limit grain growth of austenite during hardening of steel. The consequence of the introduction of microadditions is a decrease of C, Cr and Si concentration and C_E equivalent, and moreover, a decrease of local preheating temperature of high-thickness plates before their welding to 150°C.

Weldability, being a very important technological feature of structural steels, apart from formability, is dependent on the metallurgical purity, total concentration of carbon and alloying components. These factors decide about crack sensitivity of welded joints already during solidification of the weld – hot cracking, caused by increased concentration of P and S and considerable portion of non-metallic inclusions or during their cooling – cold cracking, created with the participation of microscopic stresses arising from martensitic transformation of austenite in the heat affected zone HAZ. Structural micro-alloyed steels with low sulphur and phosphorus content and limited portion of non-metallic inclusions in comparison with non-alloyed steels present low hot-cracking susceptibility, but increased susceptibility to the formation of cold cracks, which are connected with the interaction of martensitic transformation, atomic hydrogen absorbed by the weld in the liquid state and stresses connected with the grade of welded parts fastening [11-13].

Atomic hydrogen, being formed during welding as a result of dissociation of water vapour coming from humidification of welded parts and additional materials and also from crystalline water they contain, dissolves in the liquid metal of the weld. The part of dissolved hydrogen precipitates during solidification of the weld as a result of vehement decrease of solubility of this element in Fe_δ. The remaining portion of hydrogen and iron form Fe_δ interstitial solid solution. Hydrogen solubility in the Fe_γ and Fe_δ solid solution decreases along with a decrease of temperature to Ar_3, at a decreasing value of diffusion coefficient (D_H) in austenite. It leads to supersaturation of the weld by hydrogen, at partial diffusion flow of this element from the weld to the heat affected zone (HAZ) (Fig. 2).

The D_H diffusion coefficient increases considerably in ferrite during martensitic transformation occurring in the weld, at the simultaneous decrease of hydrogen solubility in this phase. High concentration gradient H, generated on the fusion surface is the cause of H penetration from the weld to the adjacent area of HAZ austenite, dissolving a greater amount of hydrogen than ferrite. High hydrogen concentration in the at-weld area of HAZ austenite at the M_s temperature causes an increase of microscopic stresses accompanying martensitic transformation. It often leads to formation of time delay cracks in the heat affected zone adjacent to the weld. In order to decrease cold cracking susceptibility, the concentration of hydrogen introduced to the weld should be limited by utilization of low-hydrogen welding methods, use of proper technological solutions, limitation of stresses connected with the grade of welded parts fixing and by employing pre-heating of elements to be joined in desired temperature [15].

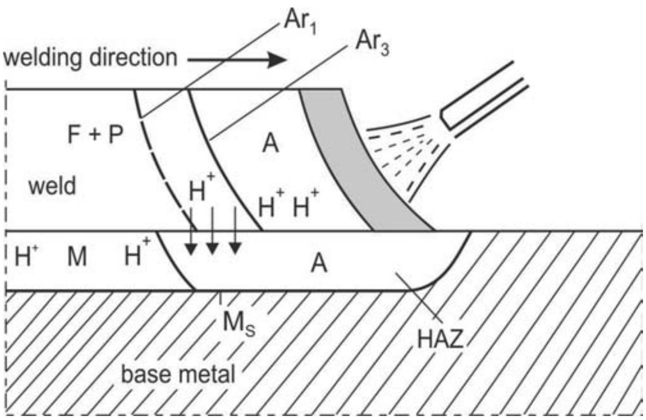

Fig. 2. Schematic diagram explaining the mechanism of atomic hydrogen penetration from the weld to the heat affected zone HAZ [14]: F – ferrite, P – pearlite, A – austenite, M – martensite, H^+ - hydrogen ions, A_{r3} – $\gamma\rightarrow\alpha$ transformation temperature, M_s – start temperature of martesitic transformation

Another cause of hydrogen embrittlement of welded joints is a cracking mechanism connected with adsorption of atomic hydrogen on the surface and its penetration inside the steel (HIC – Hydrogen Induced Cracking). Absorbed atomic hydrogen migrating in the steel is accumulated on fronts of non-metallic inclusions, microcracks and on other crystal defects, where conditions for atomic hydrogen recombination into molecular H_2 with heat releasing are suitable. Strongly exothermic recombination reaction of atomic hydrogen into the molecular form causes an increase of pressure in nascent bubbles of H_2 as well as nucleation and development of microcracks [16-20]. In order to counteract the formation of hydrogen induced cracks (HIC), a number of non-metallic inclusions and other crystal defects should be limited and low-hydrogen welding methods ought to be applied. This can be achieved through a limitation of carbon content in steel to 0.03%, of sulphur to 0.005% and phosphorus to 0.006%, at the simultaneous introduction of Nb, V, Ti and B microadditions [21].

The goal of the work was to investigate the influence of hydrogen on mechanical properties of welded joints in XABO 960 micro-alloyed constructional steel plates.

2. Experimental procedure

The research was carried out on samples cut out from the welded joint in XABO 960 micro-alloyed toughened steel plate with the thickness of 15 mm. This kind of plates finds its application for highly loaded structures and devices, particularly wheeled cranes and road machines, heavy transport vehicles, machines and devices for extractive industry. Welding process was conducted employing MAG method with the use of a shielding gas, PN-EN 439 M21. Chemical composition of the steel and the filler metal is presented in Table 1, while welding parameters are set together in Table 2. The scheme of the weld obtained is shown in Fig. 3.

Table 1.
Chemical composition of XABO 960 steel and binding agents

Material	Mass contents, %			
steel	C	Mn	Si	P
	0.18	1.60	0.50	0.020
	S	Cr	Mo	Ni
	0.010	0.80	0.60	2.00
binding	C	Mn	Si	P
	0.11	1.92	0.80	0.010
	S	Cr	Mo	Ni
	0.013	0.48	0.53	2.41

Table 2.
Parameters of a welding with the use of an electrode of 1.2 mm diameter and direct current DC(+)

Welding sequence	Current intensity, A	Voltage, V	Wire speed, cm/min	Heat input, kJ/cm
1	110 - 140	18 - 20	11 - 14	12 - 16
2 - 10	250 - 260	27 - 28	26 - 32	12 - 16

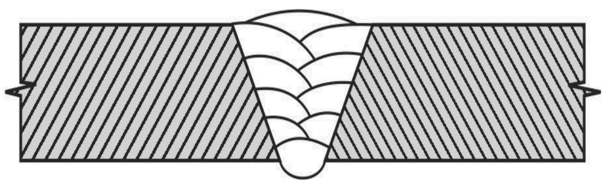

Fig. 3. Scheme of the used joint

In order to prepare test pieces, joints were cut into test samples after welding. For the purpose of achieving the goals of the work, the following were carried out:

- heat treatment of sections cut from the welded plate,
- electrolytic hydriding of samples in 1 N H_2SO_4 solution,
- static tensile test,
- hardness test,
- macroscopic metallographic examination,
- microscopic metallographic examination,
- scanning microscope observations.

Heat treatment of test samples was performed in Furnace 6000 type THERMOLYNE laboratory chamber furnace. Samples were tempered at the temperature of 350 and 600°C for 1h with successive air-cooling.

In order to investigate the influence of hydrogen on the welded joint, test pieces were charged with hydrogen electrolytically separated from 1 N solution of sulphuric acid. Samples were ground and degreased using ethanol prior to placing them in the electrolyte. The scheme of hydriding system is presented in Fig. 4 and parameters of hydriding are set together in Table 3.

The investigation of mechanical properties of welded joints was carried out using INSTRON 1195 universal testing machine, which is a part of the Laboratory equipment in the Institute of

Engineering Materials and Biomaterials of the Silesian University of Technology, applying advance speed of 5 mm · min^{-1}. The testing was performed in accordance with PN-EN 10002-1+AC1 standard on specimens with gauge length equal l_0 = 30 mm and 3 x 15 mm cross-section. Since the time between hydriding of samples and their fracture has an essential influence on the results of mechanical testing, the tensile test was conducted directly after hydriding of test pieces.

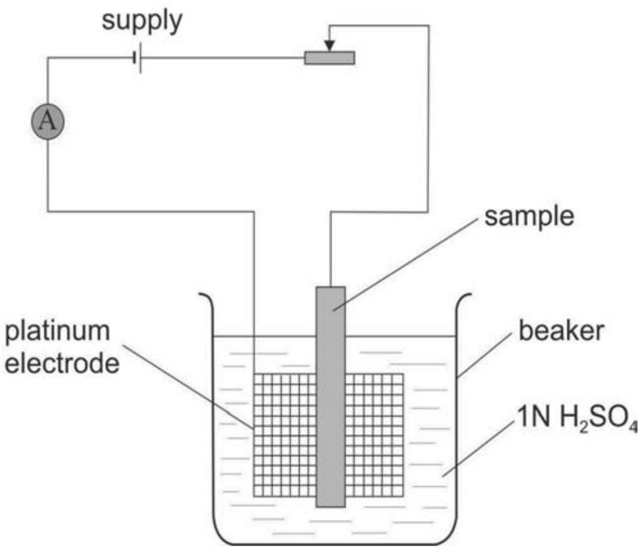

Fig. 4. Scheme of the system for hydrogen charging

Table 3.
Parameters of the hydrogen charging

Specimen mark	Tempering temperature, °C	Current density, A/cm^2	Time, h
1	raw condition	0.040	3
2	raw condition	0.035	3
3	350	0.040	3
4	350	0.035	3
5	600	0.040	3
6	600	0.035	3

Hardness test of the welded joint was done in accordance with PN-EN ISO 6507-1:2007 with the use of Vickers hardness testing machine supplied by HAUSER, applying load of 49 N. The specimen was etched prior to testing in order to reveal the heat affected zone. Hardness test was carried out along the line 3 mm distant from face of weld.

Macroscopic metallographic examinations were performed in order to compare fractures of specimens before and after their hydriding. Impurities located on the surface of analyzed samples were removed in ultrasonic washer using 20% solution of HCl in distilled water with the addition of “Tardiol D” inhibitor. Observations of fractures of test samples were carried out in LEICA MEF 4A light microscope, at the magnification of 10 and 15 x.

Microscopic metallographic examinations were carried out in order to reveal the character of corrosion damages in the fracture zones. Observations were performed on the ground and mechanically polished and Nital etched sections with the use of AXIOVERT 405M light microscope, at the magnification equal up to 500 x.

Fracture surfaces were examined using JCXA 733 scanning electron microscope supplied by JEOL with the resolving power of 3 nm and the accelerating voltage of 30 kV, applying magnification in the range from 200 x to 500 x.

3. Results and discussion

The results of static tensile tests (Table 4) indicate that hydrogen embrittlement of welded joints in investigated steel plates manifests itself with a distinct decrease of ductility and a slight decrease of strength. The highest decrease of ductility occurs after hydriding of sample that was not subjected to tempering. In this case, elongation of test pieces decreased from 10 to 6%, for the state before and after hydriding, respectively. The highest strength was noted for samples tempered at the temperature of 350°C. Non-hydrided specimen tempered at such temperature presents yield stress value $YS_{0.2}$ equal approximately 903 MPa and ultimate tensile strength value UTS of about 1035 MPa, whereas values noted for hydrided sample were as follows: $YS_{0.2}$ of approximately 854 MPa and ultimate tensile strength UTS equal around 970 MPa. Moreover, the elongation of test pieces tempered in a temperature range from 350 to 600°C changes from 8 to 6%.

Table 4.
Results of the static tensile test

Tempering temperature, °C	Specimen condition	$YS_{0.2}$, MPa	UTS, MPa	A, %
raw condition	non-hydrided	865	1053	10
	hydrided	834	1017	6
350	non-hydrided	903	1035	8
	hydrided	854	970	6
600	non-hydrided	850	993	8
	hydrided	833	974	6

Distribution of hardness changes in the welded joint is graphically illustrated in Fig. 5. Hardness of the fusion weld material ranges from 400 to 480 HV5. Performed hardness tests in both fused and heat affected zones demonstrated a clear increase of hardness in the vicinity of fused and HAZ zone. Hardness in this area increases to 528 HV5. This effect is caused by advantageous conditions of austenitizing and also by the fact that the rate of heat abstraction was sufficient for the occurrence of martensitic transformation in this region. The temperature was decreasing in the heat affected zone, further from the axis of the weld, what results in the hardness decrease on the hardness diagram. The material in this range was tempered and the tempering temperature decreased along with the increase of distance from the axis of the fusion weld. Hardness of the native material was equal 371 HV5.

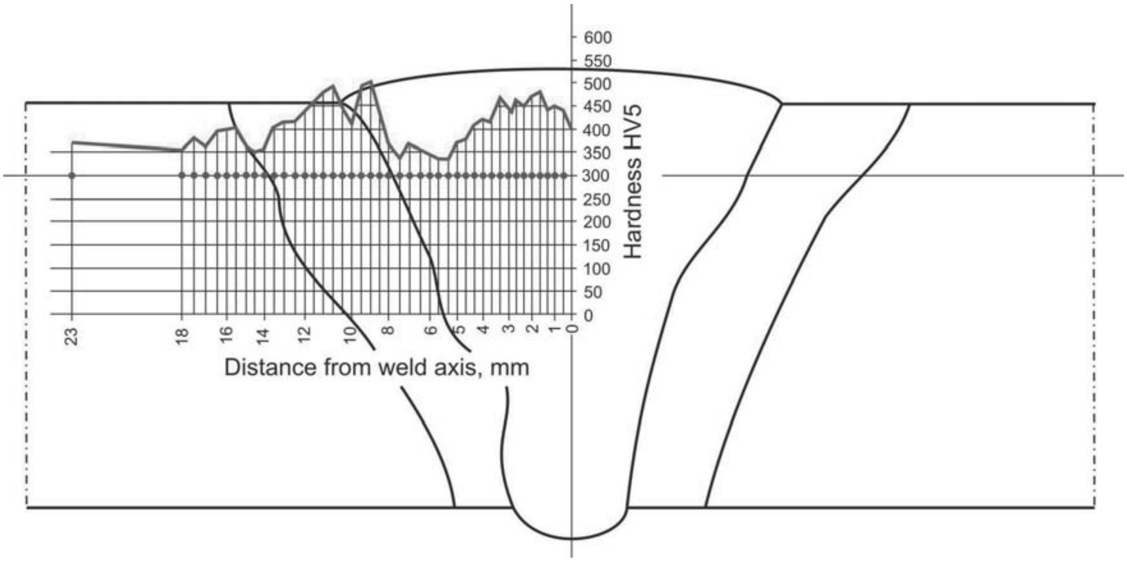

Fig. 5. Diagram of the hardness changes on the welded joint section

Macroscopic examinations of samples submitted to static tensile test directly after hydriding revealed that in most cases fracture occurred from the heat affected zone to the fused zone, independently on the tempering temperature. Brittle fracture of the sample tempered at the temperature of 350°C and hydrided is presented in Fig. 6.

Microscopic metallographic examinations of non-etched metallographic specimens showed slight portion of fine non-metallic inclusions in the heat affected zone, in the fused zone and in the fusion weld material (Figs. 7, 8).

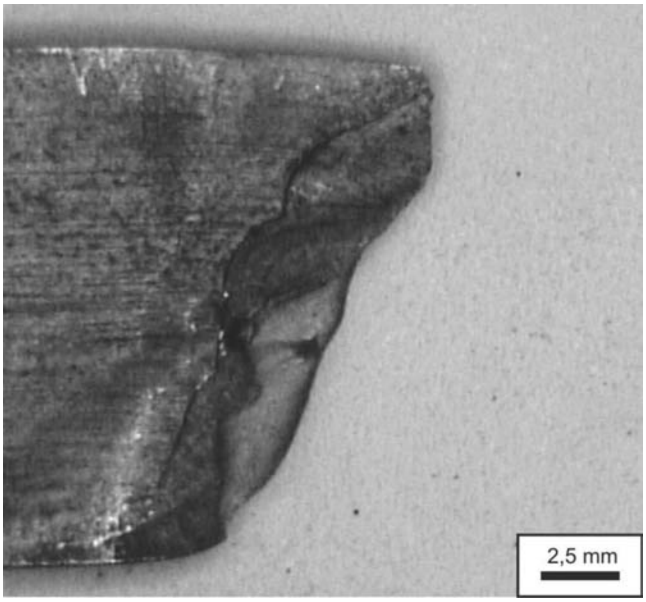

Fig. 6. Brittle fracture of the sample tempered at a temperature of 350 °C and subjected to hydrogen charging. The rupture occurs from the HAZ to fusion zone

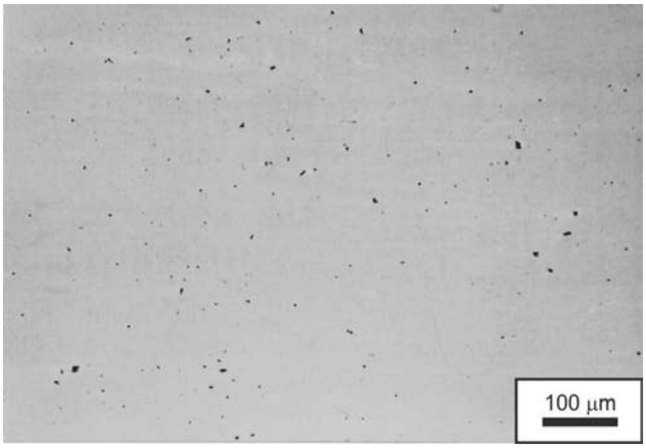

Fig. 7. Small non-metallic inclusions in the heat-affected zone

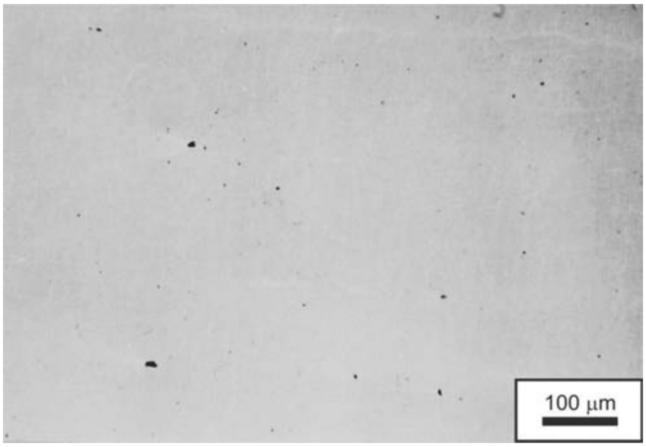

Fig. 8. Non-numerous non-metallic inclusions in the joint

Moreover, microscopic observations of samples that were hydrided and successively subjected to tensile test revealed numerous non-metallic inclusions with their increasing participation in the vicinity of the fracture, independently on the tempering temperature (Fig. 9) and also pores formed as a result of hydrogen influence (Fig. 10). High concentration of non-metallic inclusions observed in the vicinity of the fracture decides about facilitated transport of hydrogen to regions of crack development. Hydrogen entrapping on crystal defects is a reason why the total quantity of hydrogen needed for initialization of material damaging processes is low; these phenomena are local.

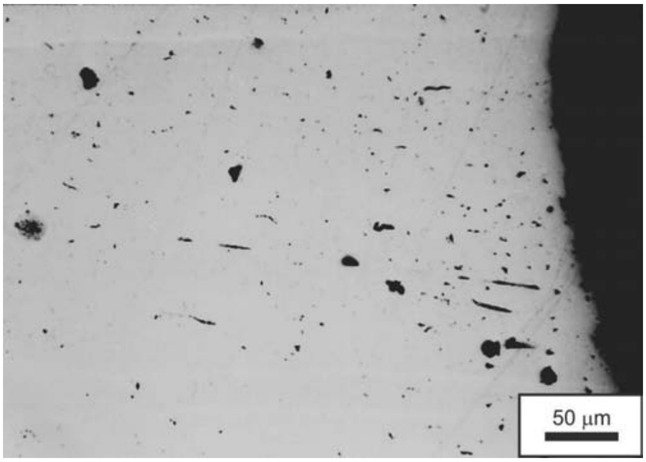

Fig. 9. Numerous non-metallic inclusions in the vicinity of the rupture in the sample tempered at a temperature of 350 °C and hydrogen charged

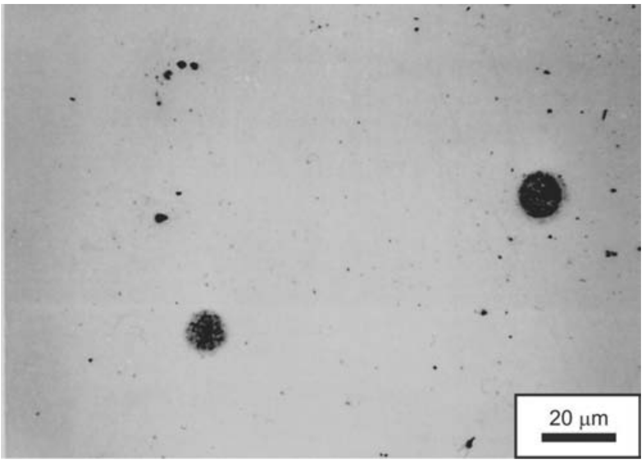

Fig. 10. Non-metallic inclusions and blisters formed due to the influence of hydrogen in the vicinity of the rupture in the sample tempered at a temperature of 600 °C and hydrogen charged

Fractographic examinations of fracture surface of hydrided and successively subjected to tensile test samples revealed numerous cracks, cavities and voids being probably the result of hydrogen interaction. These effects were revealed in both types of samples, in raw (Figs. 11, 12) and in tempered state (Figs. 13, 14), independently on the tempering temperature.

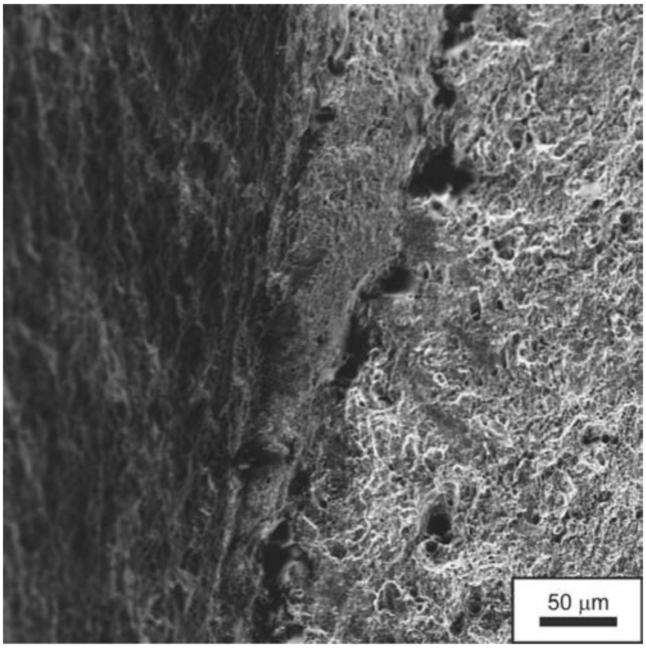

Fig. 11. Fracture surface of the fusion zone with the visible crack and numerous voids of the hydrogen charged

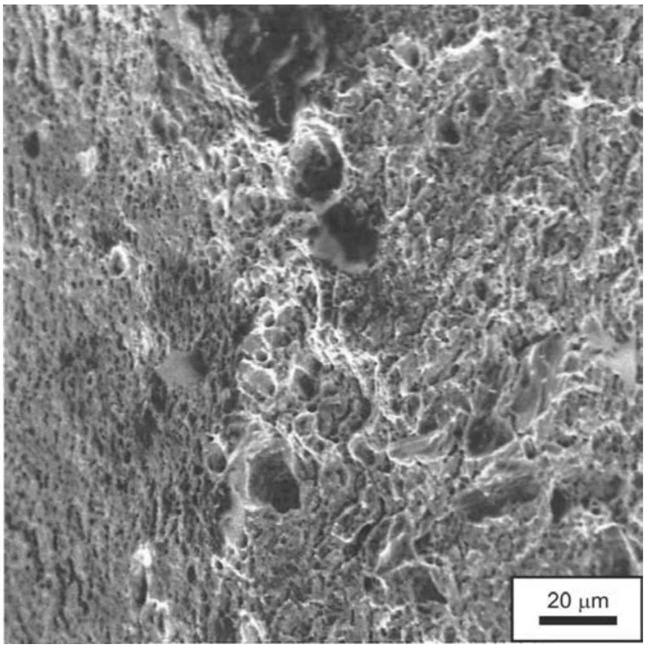

Fig. 12. Fracture surface of the heat-affected zone with the visible crack and numerous voids of the hydrogen charged

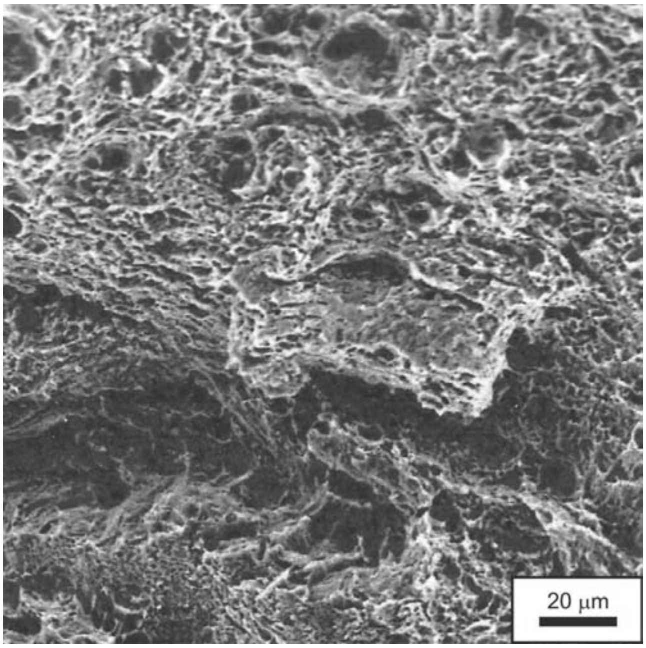

Fig. 13. Brittle fracture with visible hollows and voids in the sample tempered at a temperature of 350 °C and hydrogen charged

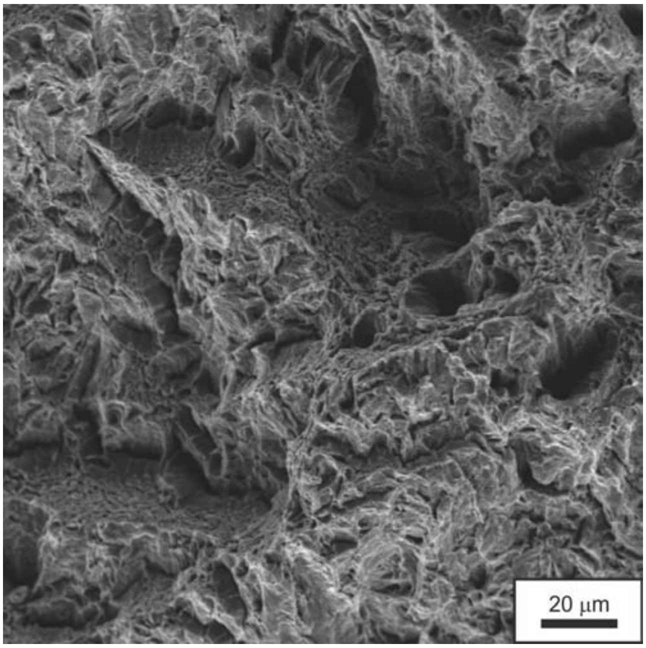

Fig. 14. Brittle fracture with visible hollows and voids in the sample tempered at a temperature of 600 °C and hydrogen charged

4. Conclusions

Performed metallographic examinations of samples subjected to prior electrolytic hydriding in 1N solution of H_2SO_4 revealed accumulation of hydrogen on fronts of non-metallic inclusions, microcracks and on other crystal defects, where powerful exothermal reaction of recombination of hydrogen atoms into hydrogen molecules takes place. It leads to hydrogen embrittlement of welded joints. In such processes, the state of stress and plastic strain formed during propagation of the microcracks plays an essential role.

It was found that hydrogen embrittlement of welded joints in XABO 960 steel plates was revealed by a distinct decrease of plastic properties and a slight decrease of strength. The highest decrease of plasticity occurs after hydriding of sample not subjected subsequently to tempering. Elongation of specimens submitted to hydriding is almost twice lower when comparing to the state before their hydriding.

It was pointed out that welded joints are susceptible to hydrogen cracking in both, heat affected and fused zone. Macroscopic metallographic observations of areas located in the vicinity of the fracture region revealed the presence of fine pores created by hydrogen and its displacement due to occurring stresses and plastic strain.

Moreover, it was found that the tempering temperature does not influence on the process of formation of pores connected with the migration of hydrogen.

In order to obtain optimal mechanical properties of welded joints in investigated steel plates, tempering at the temperature of 350°C should be applied.

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