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Evaluation of hydrogen degradation of high-strength weldable steels

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Properties

ABSTRACT

Purpose: of this paper is evaluation of susceptibility of a high-strength steel and welded joints to hydrogen degradation and establishing of applicable mechanism of their hydrogen embrittlement and hydrogen delayed cracking.

Design/methodology/approach: High-strength quenched and tempered steel grade S690Q and its welded joints have been used. Susceptibility to hydrogen embrittlement of steel and welded joints has been evaluated using monotonically increasing load. Slow strain rate test (SSRT) was carried out in hydrogen generating environment, i.e. artificial sea water under cathodic polarization. Susceptibility to hydrogen delayed cracking has been evaluated under constant load in artificial sea water under cathodic polarization. Fractographic examinations with the use of scanning electron microscope (SEM) were performed to establish suitable mechanism of hydrogen-enhanced cracking.

Findings: Tested high-strength steels and its welded joints are susceptible to hydrogen embrittlement when evaluated with the use of SSRT. The loss of plasticity is higher for welded joints then for the base metal. Tested steels and welded joints reveal high resistance to hydrogen degradation under constant load.

Research limitations/implications: Further research should be taken to reveal the exact mechanism of crack initiation.

Practical implications: Tested steel and its welded joints could be safely utilized in marine constructions under cathodic protection provided that overprotection does not take place. Tested steel could be safely utilized within elastic range of stress in hydrogen generating environments.

Originality/value: Hydrogen-enhanced localized plasticity (HELP) model is more applicable mechanism of hydrogen degradation for tested steel and its welded joints under monotonically increasing load in seawater environment. Under the critical load and hydrogen concentration notched samples premature failed and hydrogen-enhanced localised plasticity (HELP) model is a viable degradation mechanism.

Keywords: Crack resistance; High-strength steel; Welded joints; Hydrogen degradation

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1. Introduction

High-strength steels have been widely used in construction of large scale welded-structures. The principal advantage of these steels is good combination of strength and toughness, but also their good weldability. High-strength steels are especially suitable for application in pipelines, offshore facilities, and naval vessels and ships.

High-strength steels are produced as: quenched and tempered, direct quenched and tempered (the kind of TMCP - Thermo Mechanical Controlled Process), or precipitation hardened with copper. Especially, quenched and tempered steels are thought to be sensitive to hydrogen degradation. Significant limitation of use of extra high-strength steels could be their hydrogen degradation [1,2].

Hydrogen embrittlement has been the cause of failures in high-strength constructional steels used in many industry branches, e.g. the offshore industry [3,4], aircraft industry [5]. The problem is due to absorption of hydrogen from seawater, which is promoted when cathodic protection is applied to the steel to control corrosion. It is known that hydrogen uptake is increased substantially when sulphides are generated by active sulphate reducing bacteria (SRB) in marine sediments or biofilms on the metal surface [6].

Synergic action of stress and environment may result in various types of degradation of metallic materials, including hydrogen-enhanced degradation. Harmful influence of hydrogen at temperatures below 200°C is termed as low temperature hydrogen attack (LTHA). Hydrogen degrades properties of steels mainly by delayed cracking at stress below the yield strength and by the loss of ductility in a tensile test as reflected by a decreased reduction in area which is generally called hydrogen embrittlement (HE). When local hydrogen concentration is high enough (reaches critical concentration) it may cause hydrogen induced cracking (HIC) or may manifest as advancement of crack propagation (crack has been initiated by mechanical damage or corrosion).

Hydrogen effect is greater near room temperature and decreases with increasing strain rate. Hydrogen degradation is more pronounced with increasing hydrogen content or charging rate and with increasing strength of steel.

The sources of hydrogen in steel are many: gaseous hydrogen, liberation of atomic hydrogen by the iron-water or iron-H_2S reactions, decomposition of water molecules, electrolitic and corrosion processes including a cathodic reaction.

During pickling in mineral acids, cathodic electrolitical cleaning, cathodic polarisation protection, and zinc or cadmium plating hydrogen is formed. In all cases due to a cathodic reduction. The rate of hydrogen absorption can be greatly influenced by surface adsorpates called recombination poisons. The presence of poisons on steel-electrolyte interface promotes hydrogen absorption by exerting a blocking action on recombination of hydrogen. The poisons include the following elements and certain of their compounds: S, P, As, Se, Sn, Sb, Te. When hydrogen recombination is retarded, the ability of atomic hydrogen to enter steel is promoted [7].

Hydrogen degrades properties of steel under condition where cracking proceeds by all microstructural modes, including: ductile fracture - micro-void coalescence (MVC), quasicleavage, transgranular cleavage, and brittle intergranular fracture [7,8].

2. Models of hydrogen degradation

The numerous mechanisms have been proposed to explain LTHA phenomena, which reflect the many ways in which hydrogen was observed to interact with metals [7, 9-11].

Internal Pressure Model - Precipitation of molecular hydrogen at internal defects (nonmetallic inclusions, voids) develops high internal pressure. This pressure is added to applied stress and thus lowers the apparent fracture stress. The mechanism was initially proposed by Zapffe and Sims.

Hydrogen Induced Decohesion Model - Dissolved hydrogen (lattice hydrogen) reduces the cohesive strength of the lattice, i.e. interatomic bonds and thereby promotes decohesion. Mechanism proposed by Troiano and modified by Oriani. There is absence of direct experimental measurements supporting this mechanism. There are also a number of "open issues" relating to the observational base on which the decohesion model is founded. The most important is that fractography of transgranular fracture resulting from decohesion should be cleavage fracture, whereas most observations can be classified as quasi-cleavage.

Surface Energy Model (Adsorption Model) - Adsorption of hydrogen reduces the surface energy required to form a crack propagation and thus lowering of fracture stress. This model was first proposed by Petch. There are no direct experimental observation and reliable calculations that hydrogen can reduce surface energy.

Adsorption Induced Localised Slip Model - Adsorption of environmental hydrogen atoms at crack tip results in weakening of interatomic bonds facilitating dislocation injection from a crack tip and then crack growth by slip and formation of microvoids. Mechanism proposed by Lynch.

Hydrogen-Enhanced Localised Plasticity (HELP) Model - Absorption of hydrogen and its solid solution increases the ease of dislocation motion or generation, or both. Principle in this model is shielding of the elastic interactions between dislocations and obstacles by the hydrogen soluble atom (ions). Reduction of the interaction energies between elastic stress centers results in enhanced dislocation mobility. This phenomenon is supported by experimental evidences and has been observed in fcc, bbc, and hcp systems. Mechanism first proposed by Beachem and developed by Birnbaum et al. In many cases, the definition of hydrogen-related fracture as a "brittle fracture" is based on loss of macroscopic ductility (e.g. decrease of reduction in area and elongation). But careful fractographic examinations with high resolution technique shows, that hydrogen embrittlement of steel is associated with locally enhanced plasticity at the crack tip. Distribution of hydrogen can be highly nonuniform under an applied stress. Thus, the flow stress can be reduced locally, resulting in localised deformation that leads to highly localised failure by ductile processes, while the macroscopic deformation remains small.

Corrosion Enhanced Plasticity (CEP) Model - This model takes into account the generation of vacancies due to localised anodic dissolution and hydrogen evolution by cathodic reaction at the newly depasivated crack tip.

Thus, corrosion produces an enhanced localised plasticity. The activated dislocations along slip bands form pile-ups interacting with obstacles. The resulting high local stress can initiate cracking. Model was developed by Magnin et al.

This model has application mainly to passive metals and alloys like stainless steels, nickel and its alloys.

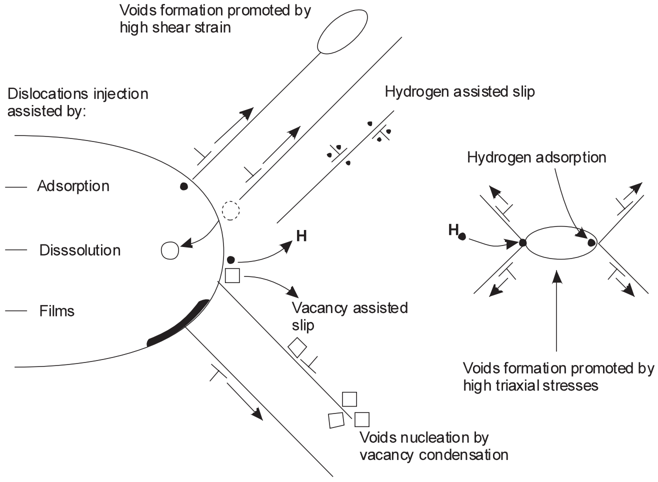

Fig. 1. Processes resulted in hydrogen assisted cracking by localized slip and microvoids coalescence [12]

Hydrogen Rich Phases Model - Formation of hydrogen rich phases - hydrides, whose mechanical properties differ from those of matrix. Cracking could proceed by the formation and cracking of brittle hydride near the crack tip. Model was generalised by Westlake. For iron it was found that no stable hydrides are formed up to hydrogen pressure of 2 GPa, so this model is not valid for steel hydrogen degradation.

Summary of possible corrosion-deformation interactions that could produce hydrogen assisted cracking is shown in Fig. 1 [12].

3. Materials

Quenched and tempered plate 12 mm in thickness made of 14HNMBCu steel grades – S690Q with minimum yield strength of 690 MPa according to PN-EN 10137-2 [13] was used. The chemical compositions of the tested steels is given in Table 1.

Welded joints were prepared with typical technology used in shipyards - submerged arc welded (SAW) and shielded metal arc welded (SMAW). Mechanical properties obtained from a tensile test performed according to PN-EN 10002-1 [14] are presented in Table 2.

Table 1.
Chemical composition of steel plate (control analyse)

Steel grade	Chemical composition, wt %						
	C	**Si**	**Mn**	**P**	**S**	**Cr**	**Ni**
S690Q 14HNMBCu	0.13	0.21	0.83	0.001	0.005	0.43	0.74
	Mo	**Cu**	**Ti**	**V**	**Al**	**B**	
	0.40	0.25	0.004	0.05	0.02	0.002	

Microstructures of the steel plate and welded joints were examined with the use of the optical microscope.

4. Methods

4.1. Estimation of susceptibility to hydrogen embrittlement

In order to estimate susceptibility to hydrogen embrittlement of tested steel and their welded joints, slow strain rate test (SSRT) according to PN-EN ISO 7539-7 [15] was conducted on round smooth specimens 4 mm in diameter.

Welded joints were placed in the centre of the specimens. Specimens were cut along the transverse direction. Tests were performed at ambient temperature either in dry air or in standard artificial sea-water grade A prepared according to PN-66/C-06502 [16]. The applied strain rate was 10^{-6} s^{-1}. Tests in sea-water were conducted at open circuit potential and under cathodic polarisation with constant current densities, chosen from the polarisation curves obtained in artificial sea-water for base metals with the potentiostatic method. The following cathodic currents were applied: 0.1; 1; 10; 20 and 50 mA/cm^2.

Elongation, reduction in area, fracture energy and additionally tensile strength were chosen as measures of hydrogen embrittlement. Then, relative parameters determined as the ratio of the appropriate value measured in air to that measured in artificial sea-water were calculated

Fracture surfaces of failed samples were investigated with the use of the scanning electron microscope (SEM) to determine mode of fracture.

4.2. Estimation of susceptibility to hydrogen delayed cracking

In order to estimate susceptibility to hydrogen delayed cracking of tested steel and its welded joints, the constant load test on round notched specimens 6 mm in diameter was conducted along with PN-EN 2832 [17]. The gauge length of samples was 50 mm. The geometry of a notch is presented in Fig. 2. For samples with welded joints, welds were placed in the centre of specimens and a notch was cut in the fusion line. All specimens were cut along the transverse direction. Tests were performed at room temperature in standard artificial sea-water grade A, prepared consistent with PN-66/C-06502 [16]. Tests in sea-water were conducted at open circuit potential and under cathodic polarisation with constant current densities chosen from the polarisation curves. The following cathodic currents were applied: 0.1; 1; 10 mA/cm^2 giving cathodic hydrogen charging of specimens during a test. Minimum two samples were used for each test parameters.

The constant load test was carried out with the use of a lever machine with leverage 25:1 and maximum load capacity of 20 kN. The machine was equipped with the environmental cell with platinum polarisation electrode. Time to failure of specimen was recorded. When a sample did not failure within 200 hours, the test was ended and result was signed as negative (-) according to PN-EN 2832 [17]. When a sample failed premature (before 200 hours), the result was signed as positive (+). Presence or lack of delayed failure of samples was chosen as measures of hydrogen degradation - susceptibility or resistance to delayed hydrogen cracking.

Table 2.
Mechanical properties (transverse direction) of steel plate and its welded joints

Steel grade	Samples	Yield Strength MPa	Tensile Strength MPa	Elongation %	Reduction in Area %
S690Q 14HNMBCu	Base metal	908	935	8.7	47.4
	SAW	601	631	7.2	55.5
	SMAW	599	687	6.6	61.9

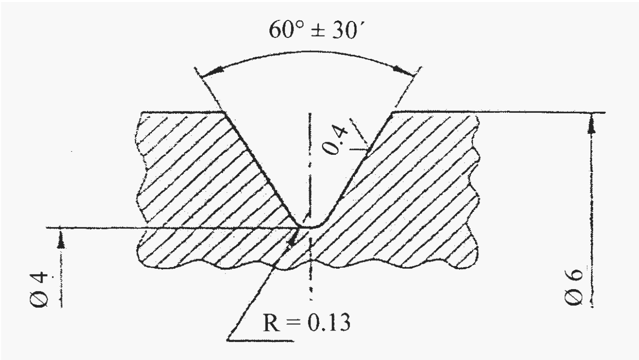

Fig. 2. The notch geometry of a specimen

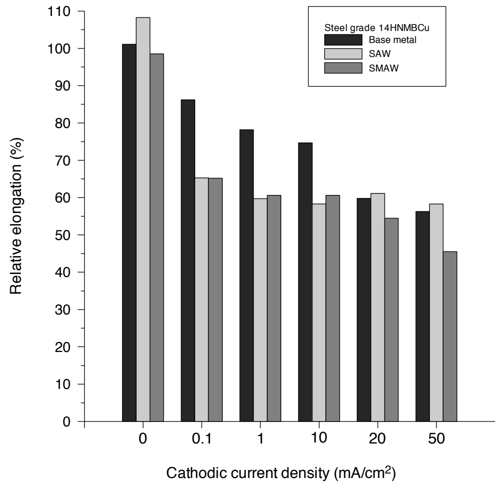

Fig. 3. Relative elongation versus cathodic current density for 14HNMBCu steel and its welded joints

Applied loads were calculated as a ratio of actual force (F) to the maximum force (F_m) obtained from a tensile test. Tensile test was performed with slow strain rate 10^{-6} s^{-1} in air using the same notched samples as for a constant load test.

Fracture surfaces of failed samples were investigated with the use of the SEM to determine mode of fracture.

5. Results and discussion

Microstructure of the steels composed of low carbon tempered lath martensite. Microstructure of the welded joint was typical for high-strength low-alloy steels. Weld metal microstructure composed of acicular ferrite and bainite. Microstructure of regions of HAZ (coarse grained region, fine grained region, and intercritical region) consisted low carbon lath martensite with various prior austenite grains size respectively.

5.1. Susceptibility to hydrogen embrittlement

Obtained results of loss of elongation, and reduction in area after SSRT test are presented in Figs. 3-4 [18-20].

Observed decrease of relative values of elongation with the increase of current density exhibits a certain minimum. Further increase of current density does not cause higher degradation due to deposits evolution on samples surface and hindering of hydrogen absorption.

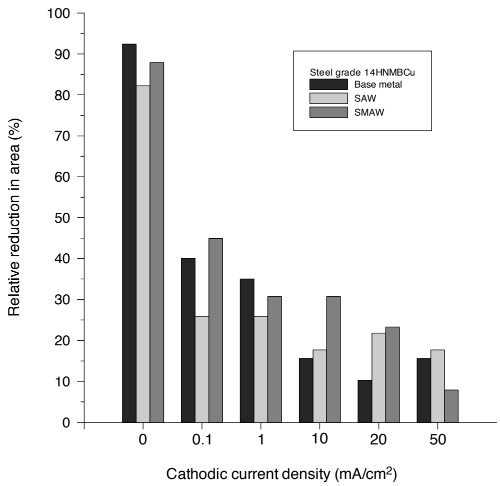

Fig. 4. Relative reduction in area versus cathodic current density for 14HNMBCu steel and its welded joints

The loss of elongation was as high as 45% for base metal, and 55% for welded joints. Reduction in area decreased of 90% for base metal, and 90% and 85% in the case of SMAW and SAW welded joints respectively. Changing of relative fracture energy with increase of current density is similar to shift of relative elongation. Tensile strength is at constant level which is typical for hydrogen embrittlement phenomenon. Failure of samples with welded joints occurred always in weld metal, where strength was lower comparing to base metal. The reduction of ductility by hydrogen was accompanied by a change in fracture mode. For samples tested in air crack growth occurred in a ductile mode.

Base metal samples tested in air had mixed - quasi-cleavage and micro void coalescence (MVC) fracture (Fig. 5). Under cathodic polarization base metal changed fracture mode, i.e. portion of quasi-cleavage fracture increased (Fig. 6), and cleavage fracture also appeared at higher current densities.

Samples with welded joints tested in air revealed ductile - MVC fracture mode (Fig. 7). When cathodic polarization was applied mixed - MVC and quasi-cleavage fracture was observed (Fig. 8). At higher cathodic current densities the presence of hydrogen induced microcracks and flakes in weld metal.

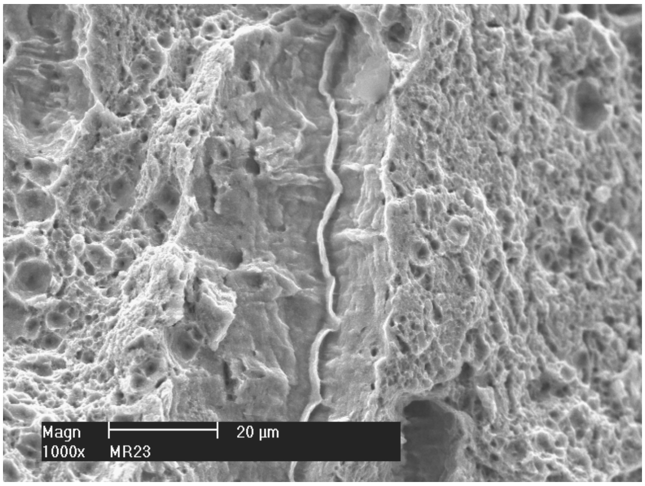

Fig. 5. SEM image of fracture surface of base metal sample after SSRT test in air

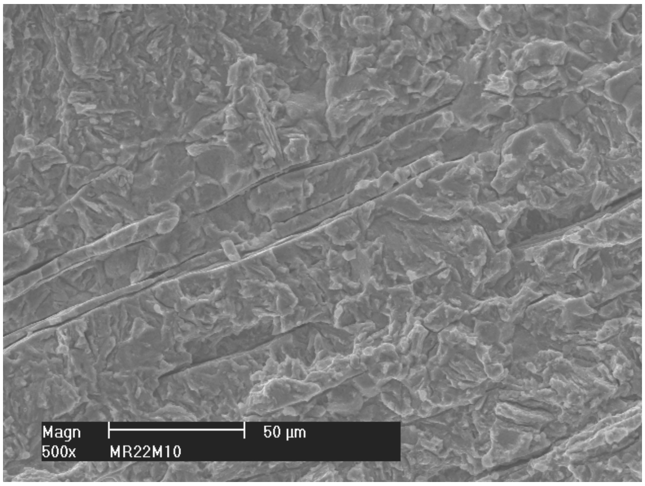

Fig. 6. SEM image of fracture surface of base metal sample after SSRT test in seawater, i = 10 mA/cm^2

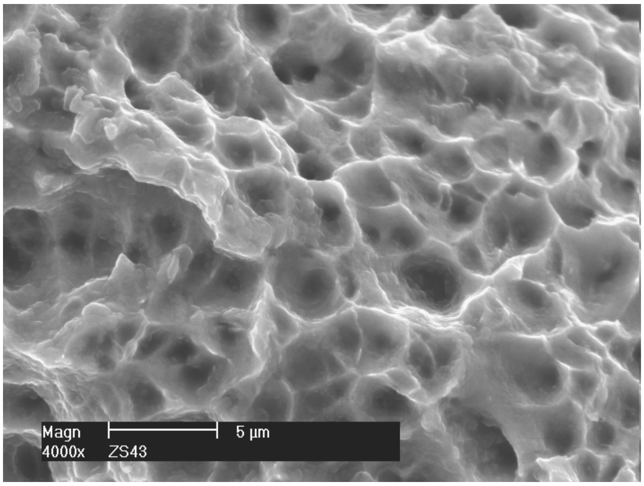

Fig. 7. SEM image of fracture surface of samplewith SMAW welded joint after SSRT test in air. Fracture localized in weld metal

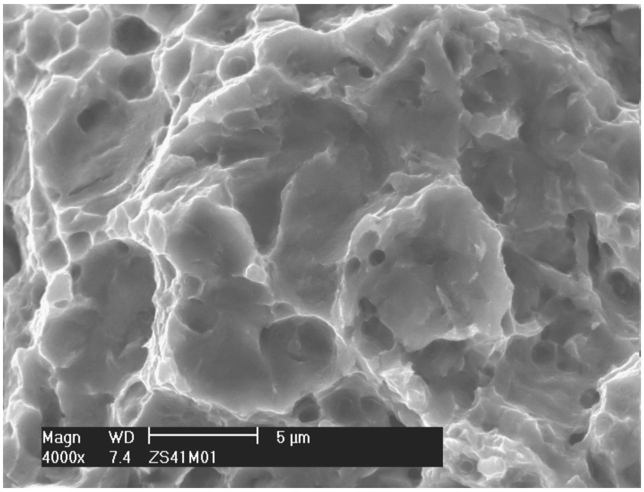

Fig. 8. SEM image of fracture surface of samplewith SMAW welded joint after SSRT test in seawater, i = 0.1 mA/cm^2. Fracture localized in weld metal

5.2. Susceptibility to hydrogen delayed cracking

Results of the constant load test are presented in Tables 3-5 [21]. Tables 3-5 present critical relative loads and cathodic current densities at which delayed hydrogen cracking occurs in 14HNMBCu steel and its welded joints. As it can be seen tested steel and the welded joints have high resistance to hydrogen degradation in seawater both at open circuit potential and cathodic polarisation. Additionally, high critical load at the level of 0.96 at open circuit potential shows that tested steel and its welded joints are not susceptible to pitting corrosion in seawater environment.

Table 3.
Resistance to delayed hydrogen cracking of 14HNMBCu steel under a constant load test in sea water

Cathodic current density mA/cm^2	Applied relative load F/F$_m$			
	0.84	0.88	0.92	0.96
open circuit potential	-	-	-	+
0.1	-	-	+	+
1	-	-	+	+
10	-	+	+	+

- means no failure within 200 hours and resistance to delayed hydrogen cracking
+ means premature failure and susceptibility to delayed hydrogen cracking

Table 4.
Resistance to delayed hydrogen cracking of welded joints (SAW) of 14HNMBCu steel under a constant load test in sea water

Cathodic current density mA/cm^2	Applied relative load F/F$_m$			
	0.84	0.88	0.92	0.96
open circuit potential	-	-	-	+
0.1	-	-	-	+
1	-	-	+	+
10	-	-	+	+

- means no failure within 200 hours and resistance to delayed hydrogen cracking
+ means premature failure and susceptibility to delayed hydrogen cracking

Table 5.
Resistance to delayed hydrogen cracking of welded joints (SMAW) of 14HNMBCu steel under a constant load test in sea water

Cathodic current density mA/cm^2	Applied relative load F/F$_m$			
	0.84	0.88	0.92	0.96
open circuit potential	-	-	-	+
0.1	-	-	+	+
1	-	+	+	+
10	-	+	+	+

- means no failure within 200 hours and resistance to delayed hydrogen cracking
+ means premature failure and susceptibility to delayed hydrogen cracking

Submerged arc welded joint (SAW) has higher resistance to hydrogen degradation than base metal. However, shielded metal arc welded (SMAW) joint is more susceptible than base metal. Differences in resistance to hydrogen delayed cracking could be explained by variations of microstructure present in steel and welded joints. The various microstructures resulting in different mechanical properties (strength, hardness) and different susceptibility to hydrogen degradation [22,23].

Fractographic observations of failed samples revealed mixed fracture mode composed of ductile and quasicleavage fracture.

Base metal samples of S690Q steel revealed quasicleavage fracture for test performed at open circuit potential, and under cathodic polarisations (Fig. 9). Samples with SMAW welded joints, tested at open circuit potential, had micro void coalescence (MVC) fracture in the weld metal (Fig. 10), which turned into transgranular cleavage in the heat affected zone (Fig. 11). Welded joints samples tested under cathodic polarisations showed brittle transgranular cleavage and intergranular fracture (Fig. 12).

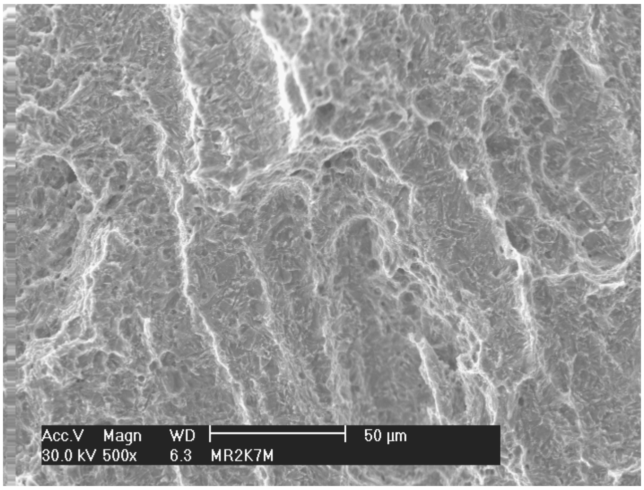

Fig. 9. SEM image of a fracture surfaces of S690Q steel. Sample after a constant load test in seawater. Relative load F/F$_m$ = 0.96, open circuit potential

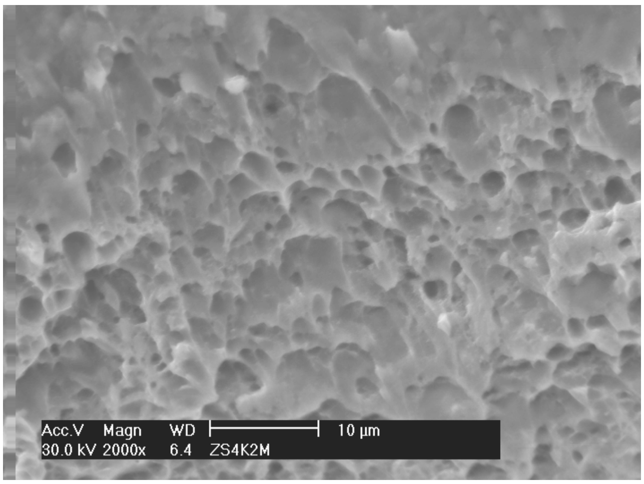

Fig. 10. SEM image of a fracture surfaces of weld metal (SMAW) of S690Q steelSample after a constant load test in seawater. Relative load F/F$_m$ = 0.96, open circuit potential

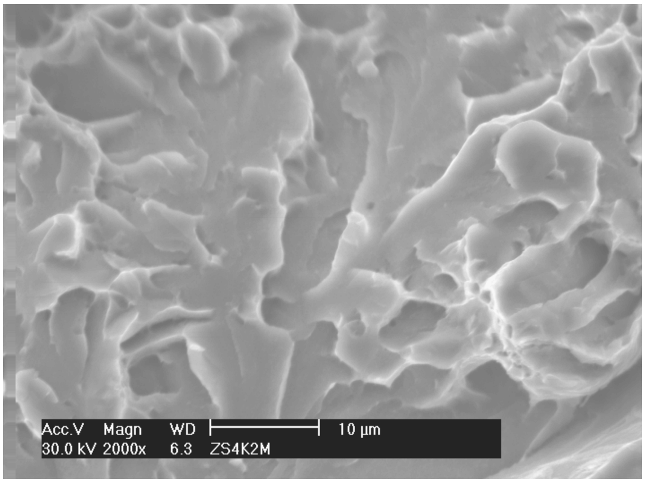

Fig. 11. SEM image of a fracture surfaces of HAZ (SMAW) of S690Q steel. Sample after a constant load test in seawater. Relative load $F/F_m = 0.96$, open circuit potential

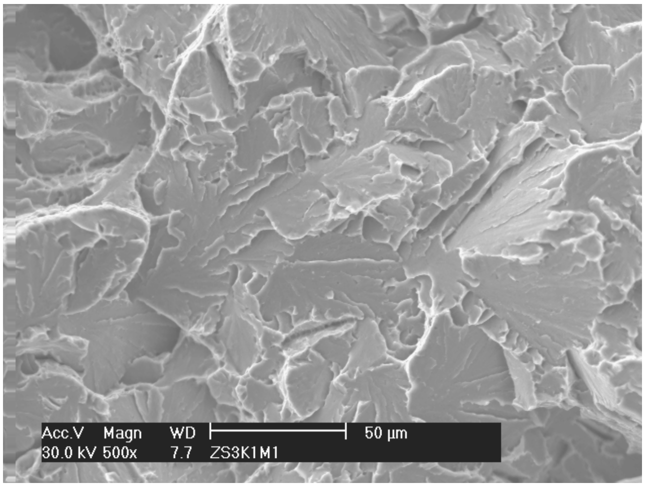

Fig. 12. SEM image of a fracture surfaces of HAZ (SAW) of S690Q steel. Sample after a constant load test in seawater. Relative load $F/F_m = 0.92$, cathodic current density 1 mA/cm^2

Obtained results of constant load test and fractographic observations suggest that hydrogen-enhanced localised plasticity (HELP) model is the more applicable mechanism of hydrogen degradation. Hydrogen delayed cracking occurs at load level as high as flow stress (yield strength) of tested steel and its welded joints. Ductile and quasicleavage fracture modes support suggestion that hydrogen interacts with dislocations and increase their mobility, and at the same time hydrogen is transported by mobile dislocations.

6. Conclusions

- Tested high-strength steel and its welded joints are susceptible to hydrogen embrittlement when evaluated with the use of SSRT. The loss of plasticity is higher for welded joints than for the base metal;
- Tested steel and its welded joints could be safely utilized in marine constructions under cathodic protection provided that overprotection does not take place;
- Hydrogen-enhanced localized plasticity (HELP) model is more applicable mechanism of hydrogen degradation than other for high-strength welded joints in seawater environment;
- High-strength low-alloy steel 14HNMBCu grade S690Q and its welded joints have high resistance to hydrogen delayed cracking in seawater environment;
- Under the critical load and cathodic current density notched samples premature failed and hydrogen-enhanced localised plasticity (HELP) model is a viable degradation mechanism.

Acknowledgements

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Additional information

Selected issues related to this paper are planned to be presented at the 16th International Scientific Conference on Contemporary Achievements in Mechanics, Manufacturing and Materials Science CAM3S'2010 celebrating 65 years of the tradition of Materials Engineering in Silesia, Poland and the 13th International Symposium Materials IMSP'2010, Denizli, Turkey.

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