



21st European Conference on Fracture, ECF21, 20-24 June 2016, Catania, Italy

Towards a unified and practical industrial model for prediction of hydrogen embrittlement and damage in steels

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Abstract

Bearing in mind the multiple effects of hydrogen in steels, the specific mechanism of hydrogen embrittlement (HE) is active, depending on the experimental conditions and numerous factors which can be grouped as environmental, mechanical and material influences. A large number of contemporary studies and models about hydrogen environment assisted cracking and HE in steels are presented in the form of critical review in this paper. This critical review represent the necessary background for the development of a multiscale structural integrity model based on correlation between simultaneously active HE micro-mechanisms: the hydrogen-enhanced localized plasticity (HELP) and the hydrogen-enhanced decohesion (HEDE) - (HELP+HEDE) and macro-mechanical response of material, unevenly enriched with hydrogen during service of boiler tubes in thermal fossil fuel power plant. Several different experimental methods and techniques were used to determine the boiler tube failure mechanism and afterwards also the viable HE mechanisms in the investigated ferritic-pearlitic low carbon steel, grade 20 - St.20 (equivalent to AISI 1020). That represent a background for the development of a structural integrity model based on the correlation of material macro-mechanical properties to scanning electron microscopy fractography analysis of fracture surfaces of Charpy specimens, in the presence of confirmed and simultaneously active HE micro-mechanisms (HELP+HEDE) in steel. The aim of this paper is to show how to implement what we have learned from theoretical HE models into the field to provide industry with valuable data and practical structural integrity model.

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Peer-review under responsibility of the Scientific Committee of ECF21.

Keywords: carbon steel; hydrogen embrittlement; mechanisms; impact strength; SEM

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1. Introduction

Because of the technological importance of hydrogen damage, numerous authors have recently explored the current state of science about the nature, causes and control of hydrogen-related degradation of metals (Dadfarnia et al., 2015a; Djukic et al., 2016; Robertson et al., 2015; Lynch, 2012). Hydrogen embrittlement (HE) of iron, steel and their alloys is extremely interesting phenomena since these materials are widely used in many industrial applications, while a fully developed and practically applicable predictive physical model still does not exist (Song and Curtin, 2013; Djukic et al., 2014; Djukic et al., 2016). Contemporary research revealed that material degradation caused by the presence of hydrogen in a material under load is manifested in a change of numerous interrelated mechanical properties including tensile strength, fracture toughness, elongation to failure, hardness, fatigue life, and crack propagation rate. Comprehensive reviews about the influence of hydrogen on the mechanical properties of steels have been published (Ahn et al., 2007; Bhadeshia, 2016; Borchers et al., 2008; Liu and Atrens, 2013).

The recently developed and proposed model for structural integrity analysis of industrial components by Djukic et al., (2016), whose overview is presented in this paper, is based on the correlation of mechanical properties to the fractography analysis of Charpy sub-sized, non-standard specimens in the presence of simultaneously active HE mechanisms: the hydrogen-enhanced localized plasticity (HELP) and the hydrogen-enhanced decohesion (HEDE), i.e. (HELP+HEDE) (Djukic et al., 2015), after reaching the critical hydrogen concentration. The aim of this paper is to show how to implement what we have learned from theoretical HE models into the field to provide industry with valuable data and practical structural integrity model that will actually prevent HEAC, HE and hydrogen damage of components in thermal power plant (TPP) and also provide predictions. A background for the analysis of the simultaneously active HE mechanisms in a ferritic-pearlitic low carbon, grade 20 - St.20 (equivalent to AISI 1020) steel and development of a model for structural integrity analysis is a concise literature overview and critical observations about the current state of the art in HE modelling, presented in the next section of this paper.

2. Towards a unified and practical industrial model

Unified practical industrial model for hydrogen assisted mechanical degradation processes in steels is as a part of development of a new paradigm for materials testing that will fundamentally alter the way industrial materials are assessed and thus how maintenance are performed. Unified industrial model for hydrogen assisted mechanical degradation processes in steels should consist of both – a comprehensive multiscale structural integrity model and a predictive maintenance model (Djukic et al., 2016). Environment in contact with metal during industrial component service is a potential source of external hydrogen due to corrosion process and from cathodic protection, while solubility, diffusion and local distribution of hydrogen in a metal are controlled by material properties and loading history (temperature distribution and fluctuation, global and local load and stress state) of industrial component.

Bearing in mind the multiple effects of hydrogen in steels, the specific mechanism of HE is manifested, depending on numerous factors which can be grouped as environmental, mechanical and material influences (Barnoush and Vehoff, 2010). Both mentioned factors, environmental and loading history of a particular TPP component, ultimately define preconditions for activation of a particular hydrogen damage (Carter and Cornish, 2001; Dayal and Parvathavarthini, 2003; Djukic and Sijacki, 2004; Djukic et al., 2005; Kolachev, 1999) and HE mechanism, including individual active mechanisms, like the HEDE (Oriani, 1972; Troiano, 1960) and the HELP (Birnbaum and Sofronis, 1994), or multiple simultaneously active mechanisms of HE (HELP+HEDE) (Katz et al., 2001; Gerberich et al., 2009; Novak et al., 2010; Djukic et al., 2015; Djukic et al., 2016). A recent study on the HE mechanism in iron indicates that a mechanism map for the prediction of HE is a function of numerous parameters, including the loading rate, hydrogen chemical potential, initial crack size, effective hydrogen diffusion activation enthalpy, temperature, and cleavage stress intensity (Song and Curtin, 2013).

A recent dislocation dynamics calculation study indicates that the possible change of HE mechanism in alpha iron, from HELP to HEDE, depends on the boundary environmental and mechanical conditions: hydrogen concentration and applied stress intensity rate. This study (Taketomi et al. 2013) also shows that at the high hydrogen concentration or the high applied stress rate conditions leads to the transition from HELP to HEDE. A comprehensive micro-mechanical model of fracture that accounts for the effect of hydrogen on the intergranular embrittlement of a high-strength steel and actual microstructure of the steel, advocates the synergistic interplay of both HELP and HEDE (Novak et al., 2010). Numerous contemporary studies confirmed that processes enhanced by the HELP mechanism (Robertson et al., 2015; Dadfarnia et al., 2015b), as well as a newly proposed hydrogen-

enhanced plasticity mediated mechanism (Robertson et al., 2015) can not only promote ductile behaviour and microvoid coalescence (MVC) modes of fracture in steels (Birnbaum and Sofronis, 1994), but also influence quasi-cleavage (QC) fracture (Martin et al., 2011) and the fracture pathway found on hydrogen-induced fracture surfaces from macroscopically brittle transgranular (TG) to intergranular (IG) failure mechanism (Wang et al., 2014). Recent molecular dynamics simulations of crack propagation in iron show that the various well-known HE phenomena can occur in the same material (Matsumoto et al., 2014), including HEDE and HELP, hydrogen-enhanced strain-induced vacancy model (HESIV) (Takai et al., 2008), adsorption-induced dislocation emission (AIDE) (Lynch, 2012) and the so-called defactant concept (Barnoush and Vehoff, 2010), depending on the boundary and the initial conditions.

Recently, based on the special approach applied in subsequent post-mortem investigations of samples unevenly saturated with hydrogen and damaged during actual operation of industrial component, the simultaneous action of the HELP and the HEDE mechanisms were detected and confirmed to be active (HELP+HEDE), depending on the local concentration of hydrogen in the investigated low carbon steel (Djukic et al., 2015; Djukic et al., 2016).

The main idea during the development of a structural integrity model presented in this paper was how to overcome the missing link between phenomenological research of hydrogen - metal interaction (theoretical HE models) and practical structural integrity model for industrial and predictive maintenance applications, Fig. 1.

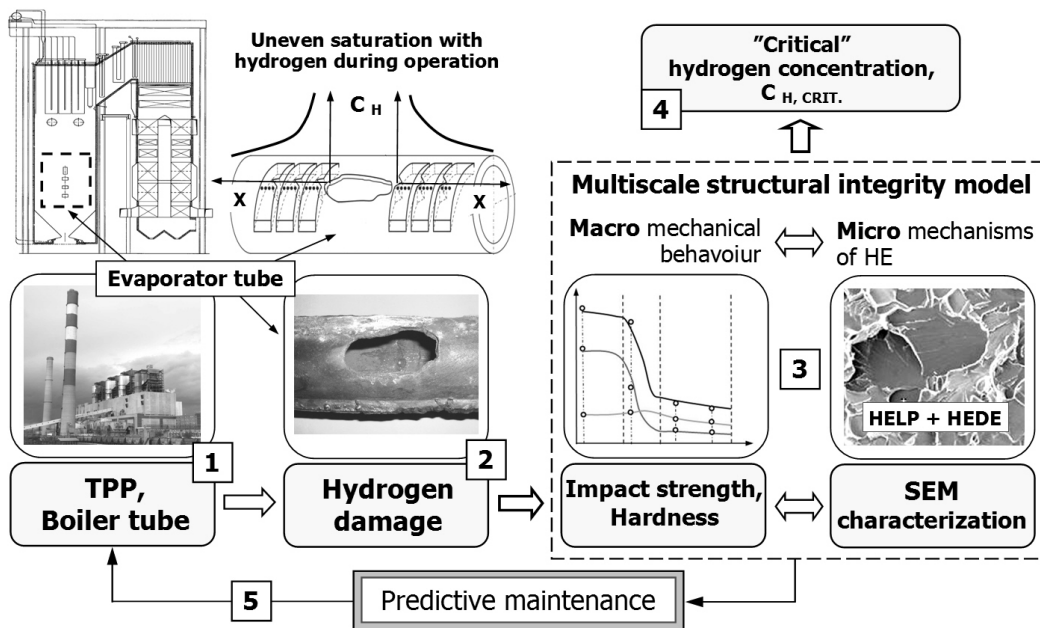


Fig. 1. Concept of a structural integrity model for prediction of hydrogen embrittlement and damage in steels: (1) Industrial component – TPP boiler tube; (2) Hydrogen damage of boiler evaporator tube unevenly saturated with hydrogen during exploitation; (3) Application of a structural integrity model - correlation between material macro-mechanical behaviour and simultaneously active HE micro-mechanisms (HELP+HEDE); (4) Assessment of hydrogen critical concentrations and (5) Predictive maintenance activities.

3. Background

During exploitation of one boiler in a 210 MW coal-fired TPP, significant failures of evaporator tubes occurred after 73,000 h of operation. Evaporator tubes were made of carbon steel, grade 20 - St.20 (or 20G, equivalent to AISI 1020). The chemical composition of low carbon steel, grade 20 - St.20 steel (GOST 1050-88) is given in Table 1. Hydrogen damage appeared on the boiler evaporator tubes located in the zone of membrane evaporator panels, exposed during operation to the peak thermal load (Djukic et al., 2015).

Our previous studies (Djukic and Sijacki Zeravcic, 2004; Djukic et al., 2005; Djukic et al., 2014) also showed that the simultaneous effect of hydrogen damage mechanisms in evaporator boiler tubes, must be considered from materials point of view, hydrogen-induced corrosion process and the complexity of design - exploitation characteristics of particular TPP unit.

Table 1. Chemical composition of St.20 steel (wt.%)

C	Si	Mn	S	P
0.24	0.28	0.48	0.025	0.013

4. Experimental concept and procedure

A critical experiment or computational model that would allow realistic simulation of the kinetics of the development of a certain type of hydrogen damage in steels including HE, which is in full compliance with the actual kinetics in the components of industrial plants exposed to hydrogen during service is very difficult to conduct. The experimental research was conducted in two distinctive phases: (1) a case study and failure analysis of the TPP boiler evaporator tube sample and (2) subsequent post-mortem analysis of the viable HE mechanisms in investigated St.20 steel unevenly enriched with hydrogen as a result of the development of intensive local hydrogenation of the tube metal during TPP boiler exploitation. A comprehensive failure analysis and case study of the boiler evaporator tube damaged during service, (phase (1)), due to the development of the hydrogen-induced corrosion process and the high temperature hydrogen attack (HTHA) were already carried and not shown here (Djukic et al., 2015). As a result of the uneven and local enrichment of metal with hydrogen, due to the activity of the HTHA process during boiler tube service, hydrogen embrittlement of the tube metal has appeared afterward in a varying degree.

Subsequent post-mortem analysis of the viable HE mechanisms, (phase (2)), represents an integral part of a model for structural integrity analysis of boiler tubes presented in this paper. Applied special experimental concept is based on the correlation of the hardness values with the corresponding macro-mechanical characteristics obtained by Charpy impact testing: impact strength values (KCV_{TOT}) and its components of crack propagation (KCV_P) and crack initiation (KCV_I) and SEM fractography analysis of fracture surfaces of Charpy specimens that enabled to define the activity and degree of influence of individual mechanism of HE and their simultaneous effects (HELP+HEDE) (Djukic et al., 2015). In this study, the term critical hydrogen concentration ($C_{H(Critical)}$) defines the concentration level which causes critical drop in the impact strength of the material (Kolachev, 1999) and a sharp ductile to brittle fracture (DBT) transition in the presence of hydrogen in steel (Djukic et al., 2015; Djukic et al., 2016). The similar approach to defining the effects of $C_{H(Critical)}$ on the mechanical properties of material was already successfully applied in the case of β -titanium (Teter et al., 2001). Also, such an interpretation allows the establishment of the new HE model in steel based on simultaneous action of both HELP and HEDE (HELP+HEDE) and also a structural integrity model for prediction of hydrogen embrittlement and damage in steels, see Fig. 1.

Numerous Charpy specimens were cut out immediately after normal shutdown and cooling of the boiler unit from the TPP evaporator boiler tube sample in the vicinity of the "window" type hydrogen damage fracture at a different distance from the fracture edge, Fig. 2a. The position and designation of Charpy specimens (S1-S6) in the vicinity of fracture are shown in Fig. 2a. The non-standard, sub-sized "Roman tile"-like geometry (Capelle et al., 2009), Charpy V-1 notched (CVN) specimens (1mm/45°) were used (dimension: 3x3x44mm and 3x6x44mm). Before impact testing CVN specimens were flattened as necessary at slow strain rate. The use of this particular specimen geometry is explained by the impossibility to prepare the standard Charpy specimen, due to the low thickness and boiler tube important curvature. The impact testing was performed in accordance with the EN 10045-1, i.e. ASTM E23-95 on the instrumented Charpy machine Schenck Trebel with maximum energy capacity of 150 J and 5.5 m/s hammer speed at 20°C. The total impact energy, as well as crack initiation and crack propagation energy components were estimated. For the hardness measurement (HV5), a stable Vickers hardness device type HPO 250 VEB-WPM, was used. Hardness measurement positions on the tube outer surface for all six Charpy specimens are also marked in Fig. 2a. The fracture surface of Charpy specimens was examined in order to identify the fracture mode and characteristic changes in fracture features caused by changes of the dominant HE mechanism. Fractography examination was carried out on a scanning electron microscope (SEM) unit, type JEOL JSM-6460LV at different magnifications, including the high one.

5. Results and discussions

Degree of hydrogen embrittlement was evaluated on the basis of decrease in the hardness with increasing distance (x , mm), from the edge of the "window" type hydrogen damage fracture, Fig. 2a. The mean hardness values for Charpy specimens (S2-S5) are shown in Fig. 2e.

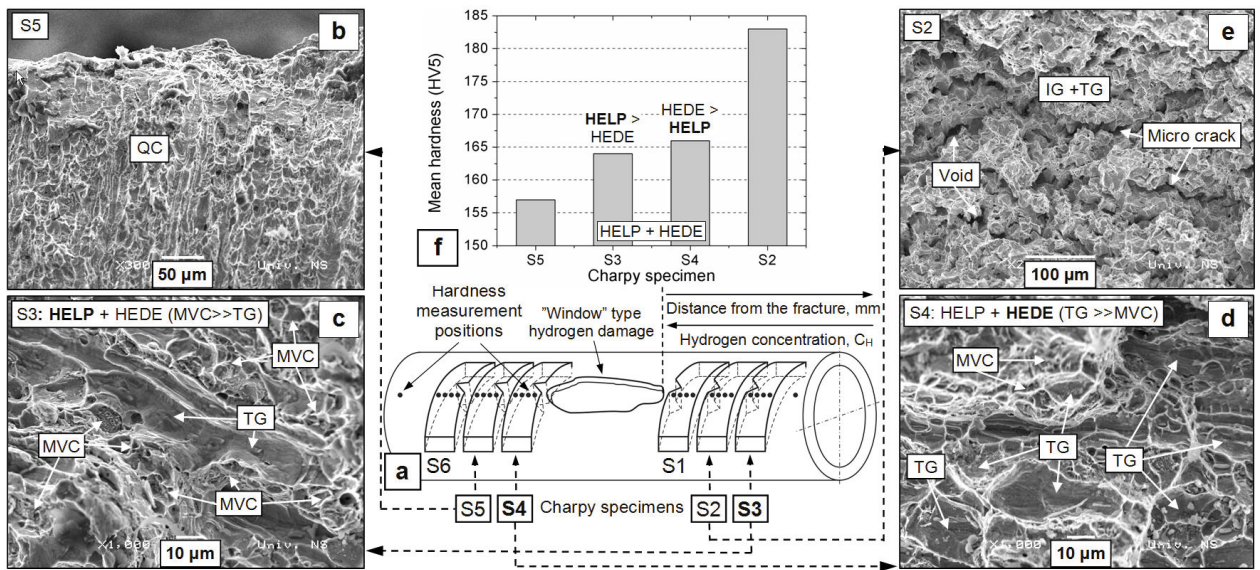


Fig. 2. (a) Position of Charpy specimens and hardness measurements in the vicinity of the "window" type hydrogen damage; (b-e) SEM fractographs of the fracture surfaces of a particular Charpy specimen (S2-S5); (f) Mean hardness of the Charpy specimens.

The highest values of mean hardness has specimen in the vicinity of "window" type hydrogen damage on the right side of fracture (S2:183HV5). With an increase of the distance from the fracture edge, hardness decrease on the both sides of the fracture (Djukic et al., 2015). The mean hardness values of specimen S3 at a distance of ~ 6 mm (right) and S4 in the vicinity of the opening (left), Fig. 2a, are similar (S3:164HV5; S4:166HV5) and higher than the maximum acceptable standard hardness value (max. 162HV5) for St.20 steel, Fig 2f. This indicates the material brittleness and change in the dominant hydrogen embrittlement mechanism, which was confirmed by impact toughness testing and fractographic studies of the fractured surface of Charpy specimens (Djukic et al., 2014).

The abrupt change in mechanical properties is a function of the content of hydrogen in the metal and whereby it appears when it exceeds a critical hydrogen concentration, $C_{H(Critical)}$ (Kolachev, 1999; Teter et al., 2001; Capelle et al., 2009; Djukic et al., 2016). Despite the fact that hydrogen content in steel was not determined in the vicinity of the "window" type fracture, the results of the present study show that the macro hardness value could be very well correlated with the hydrogen concentration in the metal, and that it is growing with an increase of hydrogen concentration (Djukic et al., 2015), regardless of the type of HE mechanism being dominant (HELP, HEDE or HELP/AIDE+HEDE).

The general fracture appearance for specimens S3-S5, Fig. 2b-d, is "quasi-cleavage" (QC) like. Over recent decades the term "quasi-cleavage", Fig. 2a, has been used to describe any fracture surface appearance that cannot be explained as either IG, MVC or a true cleavage. It is also important to note that the emergence of dominant macro brittle fracture (IG+TG) of the material enriched with hydrogen, Fig 2e, determined by the choice of experimental parameters, usually precludes the possibilities of proper detection of the phenomenon of hydrogen-assisted micro-local plasticity i.e. simultaneous effects of both HELP and HEDE mechanisms.

Simultaneous action of the HELP and HEDE mechanisms (HELP+HEDE) in St.20 steel is characterized by a distinctive mixed fracture mode with the simultaneous presence of locally ductile fine MVC fracture features of pearlitic microconstituent due to the HELP mechanism and brittle TG fracture features of ferrite, predominantly by the HEDE mechanism (MVC+TG), as shown in Figs. 2c and 2d. At lower hydrogen concentration (lower hardness), the HELP mechanism is dominant (HELP > HEDE), which is manifested by an increase in ductile MVC fracture features in the QC-like fracture surface of specimen S3, MVC >> TG, as shown in Figs. 2c and 2f. On the other hand, prevailing TG of ferrite of specimen S4 without much obvious traces of plasticity, TG >> MVC, Figs. 2d and 2f, is a consequence of increased activity of the HEDE mechanism (HEDE > HELP) with increasing in hydrogen concentration (Djukic et al., 2014; Djukic et al., 2015). Summary of simultaneously active HE mechanisms, fracture features and their effects on the decline in macro mechanical properties (hardness and impact strength) and ductile to brittle failure transition (DBT) caused by hydrogen in investigated St.20 steel is given in Table 2.

Table 2. Summary of active hydrogen embrittlement mechanisms and their effects on the decline in macro mechanical properties (hardness and impact strength) and ductile to brittle failure transition (DBT) in St.20 steel

Charpy specimen, Fig. 2a	Distance from the fracture edge (mm), Fig. 2a	Mean hardness (HV5), Figs. 2f and 3	KCV_{TOT} (J/cm ²), Fig. 3	KCV_P (J/cm ²), Fig. 3	KCV_I (J/cm ²), Fig. 3	Fracture mode and features, Fig. 2b-e	Coexistence of hydrogen embrittlement mechanisms
S5	3 (left)	157	30.10	21.11	8.99	MVC+TG, Fig. 2b	HELP ^d (AIDE)
S3	6 (right)	164	28.28 ^a	19.23 ^a	9.05	MVC+TG (MVC >> TG), Fig. 2c	HELP ^d + HEDE, HELP ^d > HEDE
S4	Close to the fracture (left)	166	13.23 ^b	3.14 ^b	10.09	TG+MVC (TG >> MVC), Fig. 2d	HELP + HEDE ^d , HEDE ^d > HELP → sharp DBT
S2	3 (right)	183 ^c	8.81 ^b	2.56 ^a	6.25 ^b	IG+TG, Voids and IG micro cracks, Fig. 2e	HEDE + HTHA

^aModerate drop; ^bSharp drop; ^cSharp rise; ^dPredominate

5.1. A structural integrity model for prediction of hydrogen embrittlement and damage in steels

The initial dominant HELP activity (HELP > HEDE) in specimen S3, TG >> MVC (Fig. 2c), is followed by a negligible drop in the impact strength (KCV_{TOT}) and its component of crack propagation energy (KCV_P) and steady value of crack initiation component (KCV_I), as shown in Table 2 and Fig. 3., and decreases with increasing in hydrogen concentration (hardness). On the other hand, prevailing TG fracture features of ferrite of specimen S4, TG >> MVC (Fig. 2d), followed by DBT fracture transition and a sharp drop in KCV_{TOT} . (reduced by a factor of two) and especially KCV_P (reduced by a factor of six), with a negligible increase in the KCV_I is a consequence of increased activity of the HEDE mechanism (HEDE > HELP), after reaching of $C_{H(Critical)}$, Table 2 and Fig. 3.

The proposed structural integrity model is based on testing of samples of a particular boiler tube that have undergone HTHA during service. Currently, this model is not useful for early detection of HTHA, prior to the first occurrence of failure, since it takes one or more of the damaged tubes in order to determine the critical drop in KCV_{TOT} , KCV_P and KCV_I , as a function of the mean specimen hardness, Table 2 and Fig. 3 (Djukic et al., 2016).

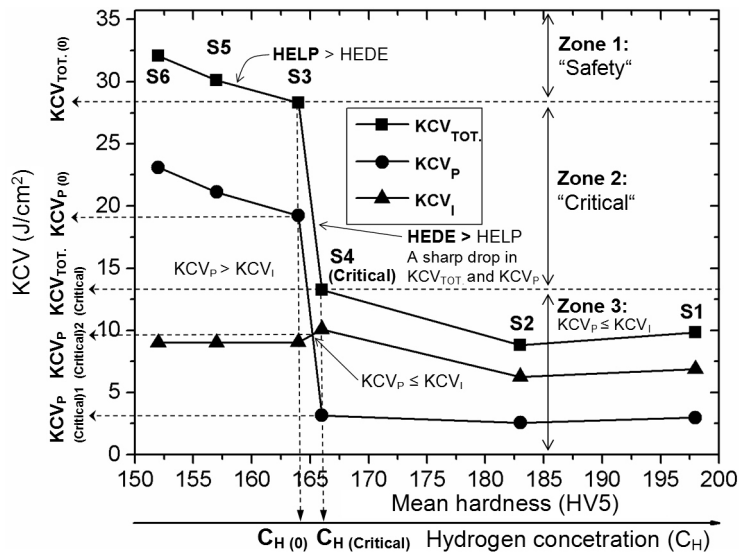


Fig. 3. The model for structural integrity analysis of boiler tubes exposed to hydrogen damage and hydrogen embrittlement: Zone 1 - "Safety" ($KCV_{TOT(0)} > KCV_{TOT(Measured)}$); Zone 2 - "Critical" ($KCV_{TOT(0)} > KCV_{TOT(Measured)} > KCV_{TOT(Critical)}$) and Zone 3 - $KCV_{TOT(Measured)} < KCV_{TOT(Critical)}$ and/or $KCV_P \leq KCV_I$. Two characteristic values of hydrogen concentration in a metal are $C_{H(0)}$ and $C_{H(Critical)}$. Variation of the impact strength (KCV_{TOT}) and its crack propagation component (KCV_P) and crack initiation component (KCV_I) of Charpy specimens (S1-S6), as a function of the specimens mean hardness (hydrogen concentration, C_H).

The general procedure of a structural integrity model for prediction of hydrogen embrittlement and damage in steels consists of six steps (Djukic et al., 2016): (1) Cutting of hydrogen damaged evaporator tube (one or more) with a characteristic "window" type hydrogen damage, Fig. 2a; (2) Determining the degree of hydrogen embrittlement in the vicinity of the fracture edge on the basis of decrease in hardness with distance from the edge of the "window" type hydrogen damage fracture, Figs. 2a and 2f; (3) Cutting sets of non-standard, sub-sized "Roman tile" geometry Charpy V-1 notched (CVN) specimens from the damaged tube in the vicinity of fracture for instrumented Charpy testing that are oriented perpendicular to the axis of the damaged tube, Fig. 2a.; (4) Drawing of the diagram: variation of the KCV_{TOT} , and its components KCV_P and KCV_I for all Charpy specimens (Fig. 3 and Table 2), as a function of the specimen mean hardness (Figs. 2f and 3 and Table 2) - hydrogen concentration; (5) Defining all necessary critical values from the diagram (minimal allowable values for KCV_{TOT} , KCV_P and KCV_I), as well as three zones that relate to KCV_{TOT} , KCV_P and KCV_I values: zone 1 - "Safety", zone 2 - "Critical" and zone 3 - $KCV_P \leq KCV_I$ (Fig. 3) and (6) Using the obtained results and adopted criteria for evaluating the structural integrity of other boiler tubes (future early detection of HTHA), made of the same material in the same TPP boiler unit, that are exposed to HTHA during service and HE thereafter (Djukic et al., 2015).

According to the proposed structural integrity model, Fig. 3, two characteristic values of hydrogen concentration in the boiler tube metal may be classified. The first is the concentration $C_{H(0)}$, beginning from which hydrogen significantly affects impact strength of material KCV_{TOT} , and its component KCV_P , which are characterized by $KCV_{TOT(0)}$ and $KCV_{P(0)}$ values, respectively. A sudden drop in ductility and DBT leads to the KCV_P value that is approaching the KCV_I value ($KCV_P > KCV_I$) as a result of the further increase in hydrogen concentration. The second one is the critical concentration $C_{H(Critical)}$, which causes significant loss of both local KCV_{TOT} and KCV_P , whose low values are now $KCV_{TOT(Critical)}$ and $KCV_{P(Critical)1}$, Fig. 3. As an alternative criterion for reaching of $C_{H(Critical)}$, the phenomenon that a KCV_P has become less than KCV_I ($KCV_P \leq KCV_I$), characterized by $KCV_{P(Critical)2}$ value, due to a sharp drop in KCV_P with an additional negligible drop in the KCV_I as a consequence of increased activity and activation of the HEDE mechanism ($HEDE > HELP$), can also be adopted as even more valid.

In the diagram (Fig. 3) it is possible to define three zones (criteria) that determine the critical drop in material ductility (KCV_{TOT} , KCV_P and KCV_I) as a function of material hardness and hydrogen concentration: zone 1 - "Safety", zone 2 - "Critical" and zone 3 - $KCV_P \leq KCV_I$. In this way, through the implementation of standard macro-mechanical testing (Charpy testing: $KCV_{TOT, P \text{ and } I(Measured)}$ and hardness measurements) of specimens cut out from the boiler tube in the vicinity of hydrogen damage, or cut out from undamaged boiler tubes in critical areas saturated with hydrogen due to HTHA (Djukic et al., 2005), it is possible to assess the structural integrity of boiler tubes (Djukic et al., 2016) in accordance with the multiscale structural integrity model proposed in this paper, Fig. 3.

6. Conclusions

Testing of the samples, unevenly enriched with hydrogen during actual operation of boiler tubes, as well as the selected special multiscale experimental concept were designed to explore the mechanisms of hydrogen embrittlement in low carbon steel St.20 (equivalent to AISI 1020). The principal observations are:

- The structural integrity model for prediction of hydrogen embrittlement and damage in steels is based on the correlation of material macro-mechanical properties to scanning electron microscopy fractography analysis of Charpy specimens fracture surfaces in the presence of simultaneously active hydrogen embrittlement mechanisms: the hydrogen enhanced localized plasticity (HELP) and hydrogen-enhanced decohesion (HEDE).
- Simultaneous actions of both mechanisms (HELP+HEDE) were confirmed, depending on the local concentration of hydrogen in low carbon steel after the actual operation of boiler tubes and not through simulation or modeling.
- The proposed structural integrity model is practical for use as a predictive maintenance in thermal power plants (TPP) and other industrial components, since it is based on the use of standard macro-mechanical tests.
- During planned/forced outages for TPP maintenance and repairs, plant operators can carry out laboratory and in-situ tests of undamaged evaporator tubes in critical areas in accordance with the structural integrity model by using the proposed criteria for predicting and preventing future hydrogen-related failures of boiler tubes.
- The future development of a unified practical industrial model regarding the implementation of necessary measurements of the critical hydrogen concentration that relies on the use of non-destructive sensors, that have been developed for determination of hydrogen content in steels and further quantification of the synergy between the HELP (AIDE) and HEDE mechanisms of hydrogen embrittlement will be the subject of further researches.

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