

**AZƏRBAYCAN RESPUBLİKASI ELM VƏ TƏHSİL NAZİRLİYİ**

**AZƏRBAYCAN DÖVLƏT NEFT VƏ SƏNAYE UNİVERSİTETİ**

**MINISTRY OF SCIENCE AND EDUCATION**

**REPUBLIC OF AZERBAIJAN**

**AZERBAIJAN STATE UNIVERSITY OF OIL AND INDUSTRY**



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**SCIENTIFIC PROCEEDINGS**

**SCIENTIFIC RESEARCH INSTITUTE**

**“GEOTECHNOLOGICAL PROBLEMS OF OIL, GAS AND CHEMISTRY”**

**Sci. Proc. SRI GPOGC. Volume 24, Number 1, 2024**

**BAKU-2024**

# Modern aspects of studying the mechanisms of hydrogen embrittlement in metal-hydrogen systems

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**Abstract.** The use of pipelines made of steel resistant to hydrogen embrittlement (HE) is of great importance in the energy industry. Hydrogen penetration into the inner surface of a steel pipe during H<sub>2</sub> transportation reduces the ductility of the steel material and increases its brittleness. This leads to a noticeable increase in the brittleness of the metal, cracking and a decrease in the efficiency of gas transportation. Thus, as a result of the interaction of hydrogen dissolved in the metal with metal defects in certain places, the mechanical properties of the gas pipeline material change. The mechanisms of the processes occurring during hydrogen embrittlement in the metal-hydrogen system are diverse. These mechanisms include hydrogen capture in lattice defects on the metal surface, the formation of new phases in the microstructure of the metal, hydrogen penetration into the metal lattice, the formation of various deposits, etc. The article analyzes some of the results of studies of the process of hydrogen capture in metal lattice defects and the mechanisms of HE occurring on the surfaces of steel materials.

**Key words:** hydrogen embrittlement HE, high-strength steels, hydrogen capture in metal lattice defects, mechanisms of hydrogen embrittlement.

## 1. Introduction

The effect of hydrogen on the properties of metals and alloys has been studied for a long time. We have analyzed many works on this topic and some selected articles are given below as literary references [1-7]. The results of well-known studies show that the proposed models [3-9] and mechanisms of hydrogen embrittlement HE [10-13] do not allow us to answer the question of how to protect metals and alloys from the HE phenomenon?

Hydrogen embrittlement HE is associated with the destruction of the crystal lattice and deterioration of mechanical properties (e.g., ductility, tensile strength, fracture toughness) of some metals (M) due to the effect of atomic hydrogen (H) on the metal in the M–H system. High-strength steels, as well as aluminum, titanium, nickel and other alloys are susceptible to HE. Ruptures of steel gas-liquid pipelines usually occur in places with increased brittleness. This leads to economic losses and disasters. However, the effect of steel properties on HE and the growth rate of fracture microcracks has not been fully studied [1].

There are two types of hydrogen embrittlement. 1. Internal hydrogen embrittlement occurs when hydrogen enters the molten metal after it has solidified. 2. External hydrogen embrittlement occurs due

to hydrogen absorption by the solid metal as a result of various reactions (heat treatment, corrosion, galvanic coating, cathodic protection, when working in a hydrogen environment at elevated pressures).

The purpose of our paper is to analyze the modern mechanisms of hydrogen embrittlement and the causes of hydrogen penetration into the crystal lattice of metal structures during industrial processes, which leads to unwanted failures of structures. Knowing the causes and mechanisms of hydrogen embrittlement, necessary protective measures are taken to improve the durability and safety of metal components.

## **2. Methodology**

Burning natural gas, coal and oil is known to produce greenhouse gas emissions (CO<sub>2</sub>, NO<sub>2</sub>, CH<sub>4</sub> and O<sub>3</sub>). In addition, the use of natural resources depletes their reserves in nature. Therefore, it is necessary to use environmentally friendly and more efficient energy sources, such as hydrogen.

Hydrogen is one of the most abundant elements in the universe, which stands out among other elements for its high energy content and environmental friendliness. As an energy carrier, hydrogen is suitable for use in industries such as driving cars, heating homes, and many other areas.

A hydrogen economy can become a reality if a clean, sustainable and cost-effective method for producing, storing and transporting hydrogen can be found. Taking into account the above, various methods for producing, storing and transporting hydrogen have been discussed in the literature in recent years.

### *2.1. Main part. Aspects of Hydrogen Production, Storage and Transportation*

In particular, review [1] examines the common methods for hydrogen production (hydrocarbon steam, hydrocarbon pyrolysis, biomass, water splitting and hydrolysis), storage (compressed hydrogen, liquid hydrogen) and transport of hydrogen.

#### *2.1.1 Aspects of hydrogen production*

Hydrogen is known to be produced mainly by fossil fuel-based technologies. In particular, 80% of all hydrogen produced is produced by steam methane reforming. The energy efficiency of this process is 74-85%. However, steam methane reforming and other fossil fuel-based technologies are not green and sustainable processes. For renewable and clean hydrogen production, energy must come from a renewable source, such as wind or solar energy.

Using a renewable source (wind or solar energy), hydrogen is produced by electrolysis, biohydrogen, thermochemical cycles, photocatalysis and plasmolysis. Among the hydrogen production technologies, electrolysis provides the highest 4% of the total global energy demand. However, the production cost and energy efficiency of electrolysis are not economically high and are estimated at 10.3 \$/kg and 52%, respectively. Therefore, it is proposed to use plasmolysis along with electrolysis. with the advantage of low energy consumption, reduced equipment size and fundamental cost. The productivity of plasmolysis is 20 g/kWh with a projected cost of 0.09 €/kWh (or 6.36 \$/kg) and an efficiency of 79.2%, respectively. Thus, the advantage of plasmolysis over electrolysis is low energy consumption, reduced equipment size and cost [14].

Work is currently ongoing to address the problems of both renewable and non-renewable hydrogen production. In particular, a strategy is proposed for the simultaneous production and separation of hydrogen using microplasma and mass transfer using microbubbles.

A comparative analysis shows the following. When using  $H_2$ , its characteristics must be taken into account, in particular its high flammability limit in air (4–74%) compared to gasoline vapor (1.4–7.6%) and natural gas (5.3–15%). In addition, its high explosive limit in air ( $H_2 = 18.3–59\%$ ) must be taken into account compared to gasoline vapor (1.1–3.3%) and natural gas (5.7–14%). It is also necessary to control the low ignition energy of  $H_2$  (0.02 MJ) compared to gasoline vapor (0.20 MJ) and natural gas (0.29 MJ). The low boiling point of  $H_2$  ( $-253\text{ }^\circ\text{C}$ ) and its low density in the liquid state (70.8 g/l) (compared to gasoline vapor (37–205  $^\circ\text{C}$ ) with a density of 700 g/l and natural gas ( $-162\text{ }^\circ\text{C}$ ) with a density of 423 g/l) also require additional safety measures for hydrogen fuel [13].

Finding a suitable material for hydrogen transportation, along with hydrogen production and storage, is an important task. This is due to the need to improve the efficiency of hydrogen fuel implementation in various industrial applications. In addition, hydrogen transportation requires knowledge of its properties, in particular, hydrogen embrittlement mechanisms, in order to avoid accidents and incidents with hydrogen leakage. Since hydrogen can exist in several states depending on temperature and pressure, the mechanisms of interaction of hydrogen with metals will be different.

The small size of hydrogen atoms ensures tight retention of hydrogen on metal carriers with a high affinity for hydrogen. The density of hydrogen in metal hydrides is greater than that of liquid hydrogen [15]. Therefore, the use of metal hydrides is effective in hydrogen storage technologies. However, the volume and weight of the metal hydride create serious problems when transporting H. This limits the practical use of many metals.

Microstructural aspects of metals influence hydrogen solubility and general properties [16]. Material properties depend on the size of the M–H systems. This is especially noticeable when the size of the systems is small and non-bulk contributions predominate. Surfaces, nanocrystals, materials with high vacancy and dislocation content, thin films and multilayer materials interact with H differently. This is due to the fact that hydrogen solubility in M–H systems depends on the morphology and microstructure, as well as on the tension between the regions of the M lattice with different hydrogen concentrations.

### *2.1.2 Analysis of models and mechanisms of hydrogen embrittlement of metals*

Aging and destruction of a structure made of iron (steel) material can occur for various reasons. For example, in the work [17] a model of yield and strain aging of iron is proposed. It is proposed that segregation of carbon atoms forms an atmosphere around dislocations. The force required to release a dislocation from this atmosphere is estimated. Using the dependence of the yield strength on temperature, it is assumed that thermal fluctuations tear small dislocation loops from this atmosphere. These loops then expand and create fields that tear out other dislocations. The shape of the yield strength dependence on temperature is compared with experiment.

Hydrogen embrittlement occurs under the influence of a number of factors, such as tensile stresses and hydrogen dissolved in the metal. In particular, HE causes cracking of welded joints of parts, high-strength steels, titanium and aluminum alloys when hydrogen enters the part.

The reduction of hydrogen on the metal surface occurs via a cathodic reaction:  $H^+ + e^- \rightarrow H_{ads}$ . The resulting hydrogen atoms are adsorbed on the metal surface. Then some of them can combine with other hydrogen atoms, forming hydrogen molecules ( $H_2$ ). Another part of H diffuses from the metal surface into the volume and dissolves in the metal. H atoms, diffusing into the metal lattice, are concentrated in the lattice defects (voids), where they combine, forming  $H_2$ :  $2H_{ads} \rightarrow H_2$ . The specified mechanism is shown in Fig. 1.

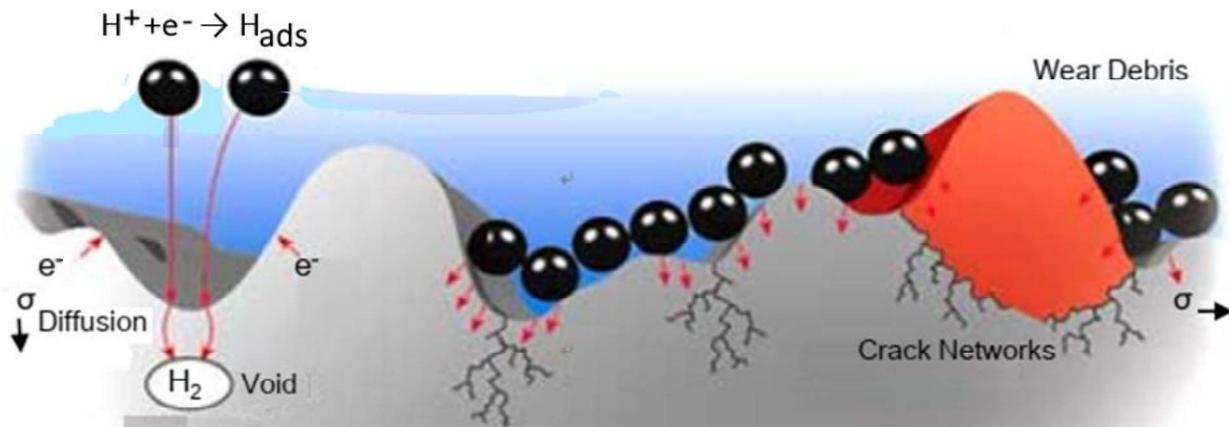
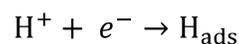
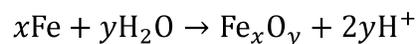


Figure 1. Scheme of interaction of hydrogen with the surface of a solid body with the formation of cracks [19].

The development of high-strength pipeline steels with low susceptibility to hydrogen embrittlement HE is of great importance in the energy industry. Available data on the hydrogen embrittlement of pipeline steels are often contradictory [18-20]. The HE phenomenon is associated with a complex interaction of hydrogen particles with the microstructure and chemical composition of steel (Figure 2).

The hydrogen trapping in the H-steel system is also influenced by the microstructural properties created during the steel processing. Hydrogen traps in steel are classified as structural defects, microstructural phases, inclusions and precipitates. In pipeline steels, hydrogen can be present both externally and in the volume of the alloy. When steel surfaces come into contact with moisture, reactions of hydrogen particle formation occur



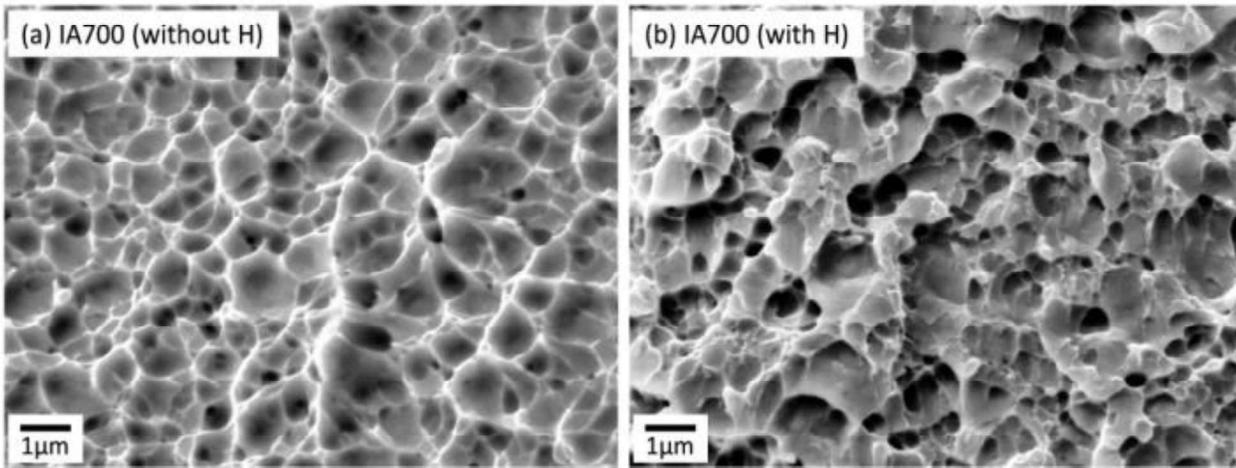


Figure 2. Typical SEM fractogram of a medium-manganese steel sample IA700 (a) without hydrogen pre-charging and (b) with hydrogen pre-charging at an annealing temperature of 700 °C [20].

Trapped hydrogen remains in trapping centers or in defects in the microstructure of the material for a longer time than if it were to move through the crystal lattice. Hydrogen atoms dissolve poorly in steel, but internal lattice defects increase the solubility of H and reduce the diffusion of H. Given the magnitude of the activation energy for hydrogen desorption, hydrogen capture by lattice defects can be either a reversible (diffusion) or irreversible (non-diffusion) process. Traps and structural defects that temporarily collect hydrogen atoms are reversible process traps. Defects that trap and retain hydrogen for a long time are irreversible process traps.

Thus, in reversible HE, hydrogen atoms migrate into the alloy structure. They accumulate in the places of potential cracking of the alloy and cause slow destruction of the alloy. In the irreversible HE processes; hydrogen atoms combine in structural defects of the alloy and form hydrogen molecules  $H_2$ . In such defects, the  $H_2$  pressure exceeds the critical strength of the material, and local  $H_2$  pressure is created there. As a result of this  $H_2$  pressure, cracks are formed in the alloy defects. Using various processing methods, it is possible to remove hydrogen from material defects in the reversible HE processes. The irreversible HE processes can persist in the material defects for a long time.

During HE of a metal or alloy, atomic hydrogen migrates to the sites of microcracks in the metal lattice and causes further expansion and development of macrocracks. The scheme of migration of hydrogen atoms H in the microstructure of the metal is presented as in Figure 3.

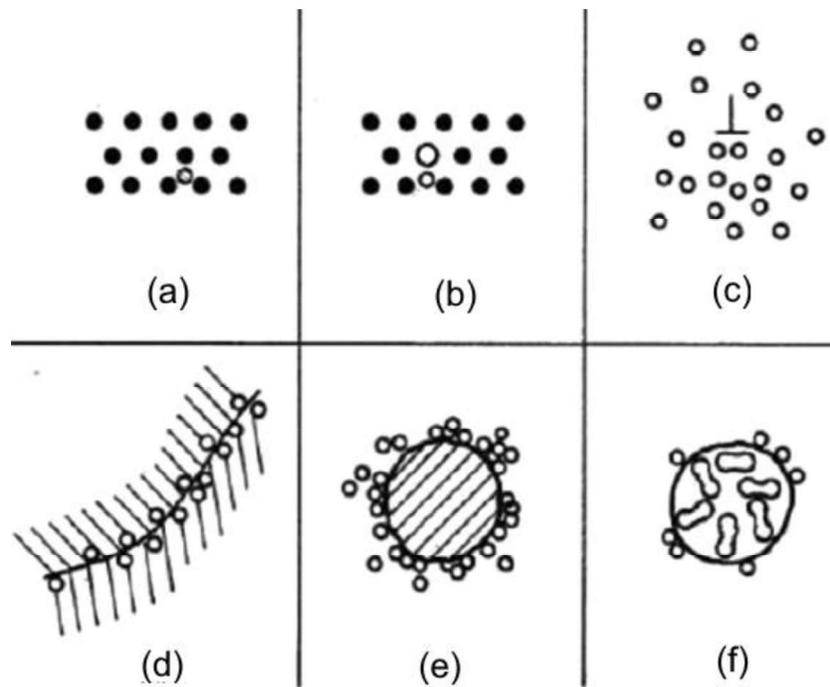


Figure 3. Different locations where hydrogen atoms H are present in the microstructure of a metal: (a) solid solution with H; (b) a pair of dissolved hydrogen atoms; (c) dislocation atmosphere of H; (d) accumulation of H at a grain boundary of the metal; (e) accumulation of H at a particle-matrix boundary; (f) a lattice defect (vacancy) containing recombined hydrogen.

Based on this and the fact that the effect of hydrogen on mechanical properties is not constant among steels, the mechanisms associated with hydrogen embrittlement in H traps differ from each other.

Individual metallurgical factors also influence the deterioration of mechanical properties under the influence of hydrogen. Thus, research in this area continues, in particular, a model for the analysis of hydrogen accumulation and critical conditions for the occurrence of cracks at inclusion-steel matrix boundaries has been proposed [21].

Based on the above and an analysis of the results of a number of different studies on hydrogen embrittlement of metals, it has been established that some issues have received little attention. Such issues include hydrogen retention on the surface, criteria for degradation of the crystalline structure of the metal, and mechanisms of hydrogen embrittlement. Investigation of these mechanisms will help prevent hydrogen embrittlement of metals, especially steel pipes, due to hydrogen capture on the surface.

The essence of H embrittlement is as follows [18]. Hydrogen can penetrate metals during their production and/or thermomechanical treatment in humid air through the decomposition of water. This produces hydrogen and metal oxide. Metal surfaces containing oxides can react with (i) humid air, (ii) water or (iii) organic substances. In these reactions, hydrogen is produced in atomic form. This accelerates the penetration of hydrogen into the metal compared to the action of gaseous hydrogen

molecules. The metal surface can be a membrane for water molecules or can be destroyed during corrosion. This also leads to the formation of hydrogen.

Even a few ppm of hydrogen can cause a loss of ductility in the metal in the M-H system. External and internal hydrogen causes accelerated growth of M cracks (e.g. steel). This phenomenon is harmful, in particular, for high-strength steels (yield strength above 600 N/mm<sup>2</sup>). The mechanical strength of steels can be reduced below the yield strength in the presence of H. There is a threshold stress below which HE does not occur in steels. This threshold stress is a function of the strength of the material and the environment. The higher the yield strength or tensile strength of the metal, the lower the threshold stresses. HE is associated with H absorption, and there is an incubation period for H charging and transport and/or defect formation in the metal. HE leads to deterioration of the mechanical strength and delayed failure of the metal.

Several mechanisms are known to explain HE phenomena, including the early hydrogen pressure (HP) models [22] and the hydrogen-induced phase transformation (HIPT) model [23]. The essence of the HP model is that hydrogen atoms preferentially segregate in defective areas of the metal (microvoids and inclusions) and degrade mechanical properties. The HIPT model shows that hydrogenation of austenitic steels leads to an increase in the concentration of vacancies in the host lattice. This leads to a loss of phase stability and a hydrogen-induced phase transformation of the alloy occurs. The mechanisms leading to H embrittlement are interrelated and depend on the external or internal conditions of the M-H systems [3-9, 24-31].

The main known HE models are listed below.

- I. HEDE – Hydrogen-Enhanced Decohesion (Troiano, 1960; Oriani, 1970; Oriani and Josephic, 1974).
- II. HYFO – Hydride Formation (Westlake, 1969; Gahr and Birnbaum, 1978).
- III. AIDE – Adsorption-Induced Dislocation Emission Mechanism (Lynch, 1979).
- IV. HELP – Hydrogen-Enhanced Localized Plasticity (Beacham, 1970; Birnbaum, Robertson and Sofronis, 1980; Birnbaum and Sofronis, 1994).
- V. HESIV – Hydrogen-Enhanced Strain-Induced Vacancy Mechanism (Nagumo and Takai 2004).

These mechanisms HE assumes that hydrogen penetration into metals (steel) in service occurs from two sources: hydrogen comes from the environment, i.e. (i) gaseous hydrogen and (ii) H is obtained due to internal reactions in the M-H system, in particular, corrosion. The schematic of the main mechanisms of HE is shown in Fig. 4.

The rate of interaction of a gas and a solid depends on the adsorption energy (physical adsorption and chemisorption). Such adsorption energy is associated with the bond energies of the M-H and H-H pairs. The energy of physical adsorption is low (<20 kJ mol<sup>-1</sup>). In this case, equilibrium in the M-H system is achieved at room temperature. At high temperatures, chemisorption occurs during hydrogen absorption. For the Fe-H bond, the adsorption energy is >280 kJ mol<sup>-1</sup>.

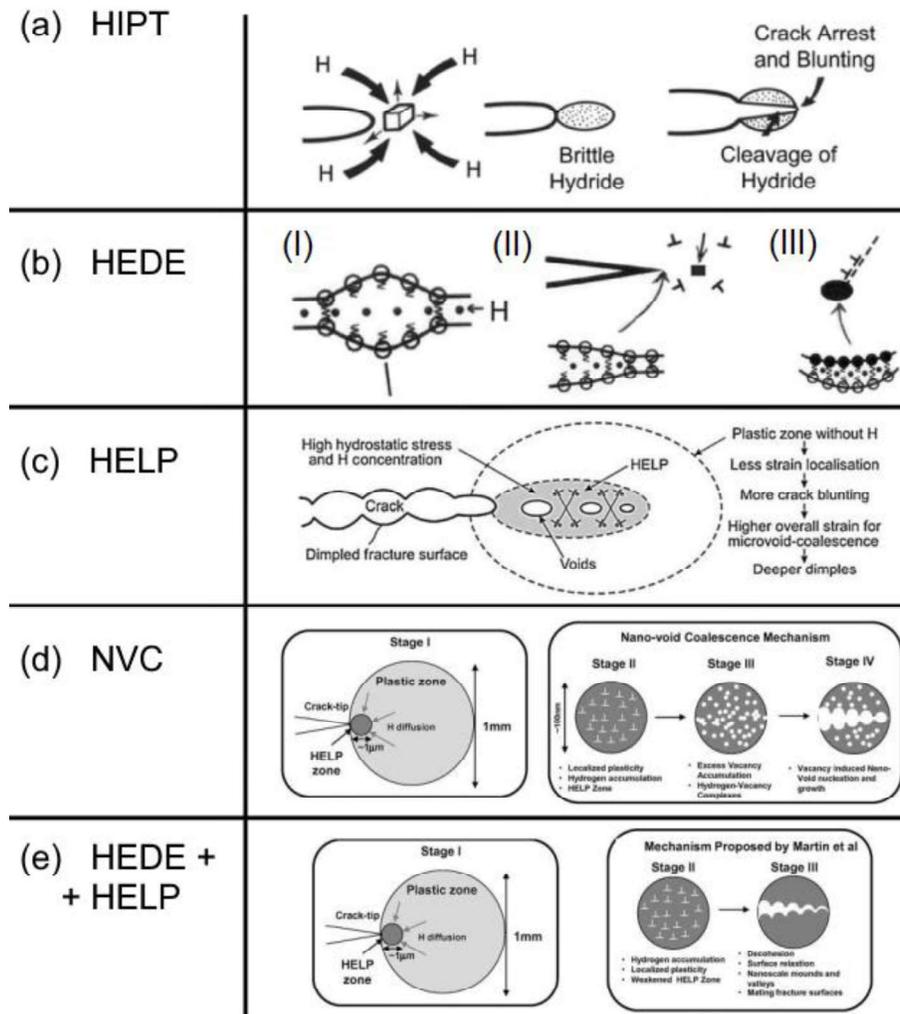


Figure 4. Schematic of the main mechanisms of HE: (a) HIPT: hydrogen-induced phase transformation theory; (b) HEDE: hydrogen-enhanced decohesion mechanism; (c) HELP: hydrogen-enhanced localized plasticity mechanism; (d) NVC: nanovoids coalescence mechanism; (e) HEDE +HELP]: combined effect of hydrogen-enhanced decohesion mechanism and hydrogen-enhanced localized plasticity mechanism [32].

Below is a schematic representation of the steps of typical corrosion reactions involving hydrogen in acidic solutions [33].

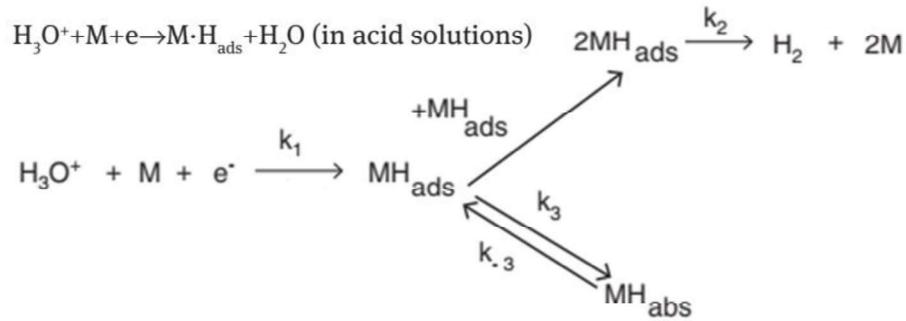


Figure 5. Adsorbed electrolytic hydrogen from the intermediate stage enters the metal (cathode) substrate. The reaction proceeds slowly in an acidic environment.

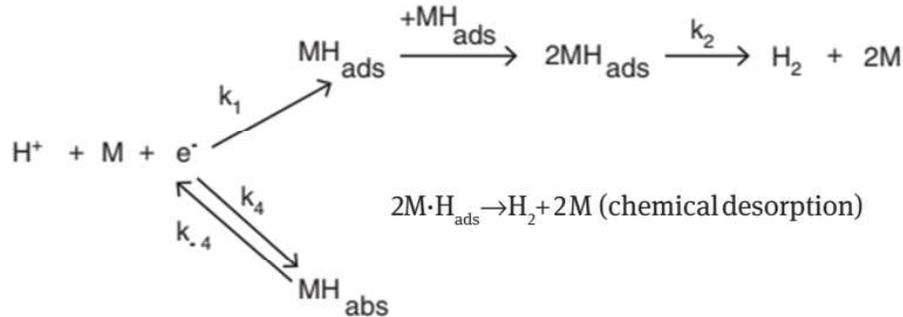


Figure 6. Hydrogen adsorption reactions, where the intermediate states of hydrogen adsorption on metal and hydrogen release are different.

To describe the composition-property-structure dependence in the M-H system, currently used, in particular, are ab initio quantum-chemical calculations [33-38]. For example, in the work [34] tight-binding approximations were used to describe the electronic structure and interatomic forces in iron containing hydrogen impurities. The canonical description of the *d*-band is evaluated in comparison with a non-orthogonal model including *s*- and *d*-bands. The calculated data of the models are compared with known properties, including the energy of hydrogen segregation into a vacancy and on the iron surface.

It is indicated that there is good agreement with the experiment. The figure 7 shows the energy bands for bcc-tetrahedral FeH calculated using the DFT LSDA and GGA potentials at the lattice constant of pure bcc-Fe. In the Figure 7, the colors of the curves are as follows: H *s* – red, Fe *d* – blue, Fe *s* – green. The Fermi energy is indicated by the horizontal line. The Fe 4*s* orbital (zone) extends above the Fe *d* zone.

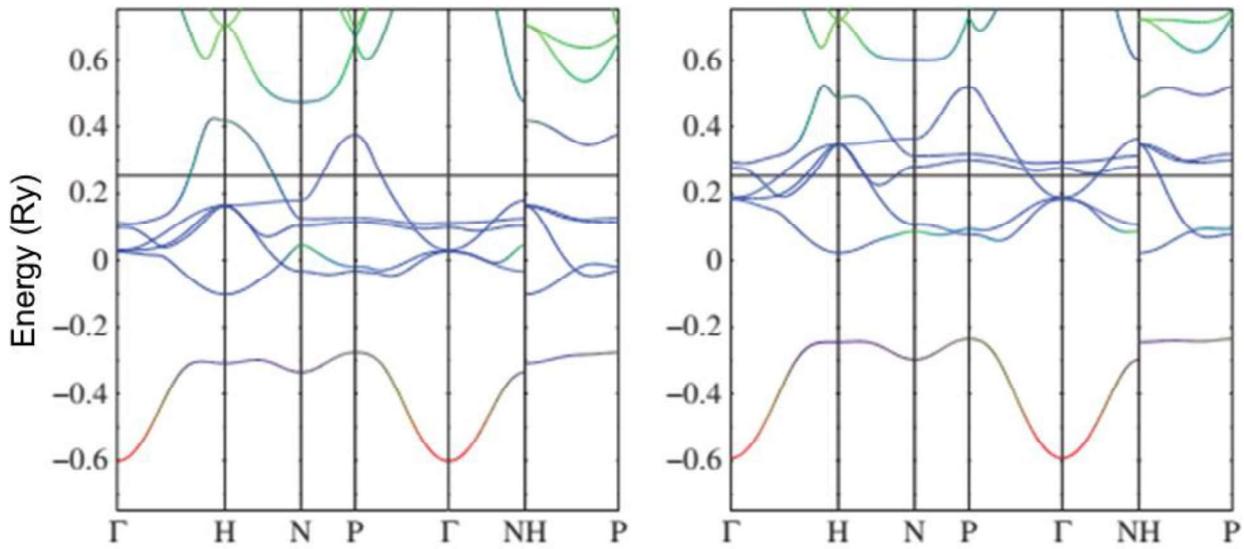


Figure 7. DFT calculated band structure of  $\alpha$ -iron containing hydrogen impurity. (a) local spin density approximation (LSDA), (b) generalized gradient approximation (GGA). The electron bands of different orbitals are colored differently: red -  $s$ -electrons, blue -  $d$ -electrons and green -  $p$ -electrons [34].

Thus, atomic hydrogen penetrates the metal lattice and can initiate HE in steels, i.e. create defects and cracks in structures. Therefore, research into various aspects of this phenomenon continues (see, for example, [39-40]). To prevent HE, various coatings are used [40]. In this case, the coating defects, plasticity and bond strength between the matrix and the coating are taken into account. Hydrogen penetration and the HE processes in the M–H system can be suppressed by compressing the crystal lattice of M, for example, austenite ( $\gamma$ -phase). When modifying the microstructure of austenite M, it is important to know the morphology and stabilization conditions of M. The elements C, N, Ni and Mn are used as additives for stabilization, for example, of austenite. Nanoscale precipitates in steel have an ambiguous effect on HE in dispersion-hardened steels. The density of nanoscale precipitates in steel is associated with the susceptibility of steel to HE.

The dependence of the metal defect energy  $\gamma$  on the chemical potential  $\mu_H$  of hydrogen can be described by the Gibbs adsorption isotherm (Equation (1))

$$\left. \frac{\partial \gamma}{\partial \mu_H} \right|_{V, T, p_i, n_M} = -\Gamma_H \quad (1)$$

According to equation (1), at very low chemical potentials ( $\mu_H \rightarrow -\infty$ ) the concentration of H and hence the excess  $\Gamma_H$  become small. Therefore, the slope of the curve  $\gamma$  versus  $\mu_H$  is zero. In the case of the formation of a new phase rich in hydrogen (curve 3), the logarithm of the concentration  $C_H$  is used for the abscissa (Fig. 8). The chemical potential, and therefore the excess H and  $\gamma$ , remain constant during phase separation, despite the increase in the H concentration.

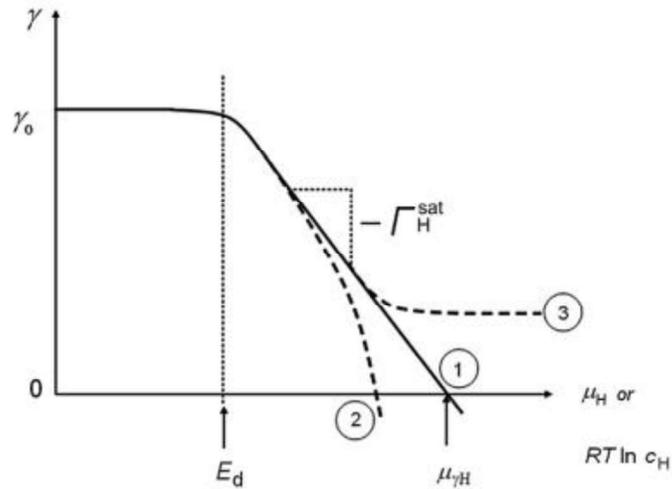


Figure 8. Dependence of the defect energy  $\gamma$  on the chemical potential  $\mu_H$  of hydrogen. 1 – saturation of the defect with hydrogen  $\Gamma_H^{\text{sat}}$  leads to a line with a constant slope, 2 – excess H does not saturate the defect, but continues to increase, 3 – the dependence characterizes the formation of a new phase rich in hydrogen.

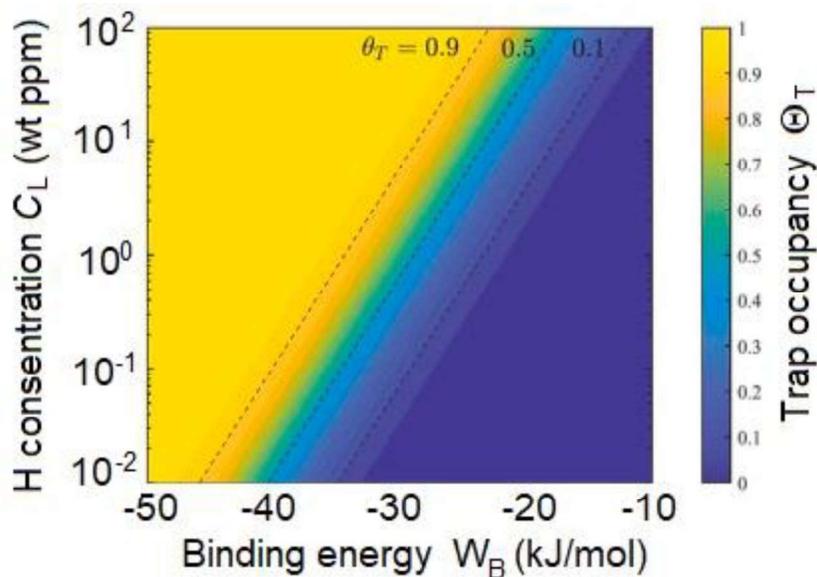


Figure 9. Dependence of the lattice hydrogen concentration  $C_L$  on the binding energy of the H-trap  $W_B$  taking into account the sensitivity of the H-trap population  $\theta_T$ .

It has been shown experimentally that the maximum achievable hydrogen concentration in the alloy lattice during cyclic analysis is sensitive to the relationship between the loading frequency and the effective diffusion coefficient [40]. This is observed both for samples pre-charged with hydrogen and for samples exposed to a constant source of hydrogen (Figure 9).

### 2.1.3 Accidents in pipelines and hydrogen transportation

In addition to the production and storage of hydrogen, one of the main issues that must be resolved for the use of hydrogen fuel in various industries is the search for suitable means of transporting it. In order to prevent hazards such as explosions and leaks in pipes during the transportation of hydrogen, it is important to know the properties of the M–H system. Pipelines have been used to transport hydrogen for over 50 years. Currently, for example, there are about 16000 km of pipelines supplying hydrogen to oil refineries and chemical plants.

Hydrogen pipelines exist, for example, in different parts of Europe. Their length depends on the volume of hydrogen to be transported and the distance of delivery. Liquid hydrogen pipes are preferable for small volumes and long distances. And compressed gaseous hydrogen pipes are suitable for transporting small volumes of hydrogen over short distances. Energy transportation via pipelines is an efficient method of delivery. However, energy transportation via pipelines has its drawbacks, in particular, those related to transportation failures and possible pipeline breakdowns.

Liquid pipelines have a higher failure rate than gas pipelines. Liquids in pipelines cause and/or accelerate corrosion, which reduces the performance of the material and increases the failure rate of pipelines. Material design, corrosion, and cracking of the material are the main problems for the inefficient operation of pipelines. Material corrosion is associated with both external and internal defects in the material of the pipelines. Cracks in pipes, along with corrosion, can also significantly affect the performance of pipelines. Figure 10 shows information related to accidents that may occur on pipelines.

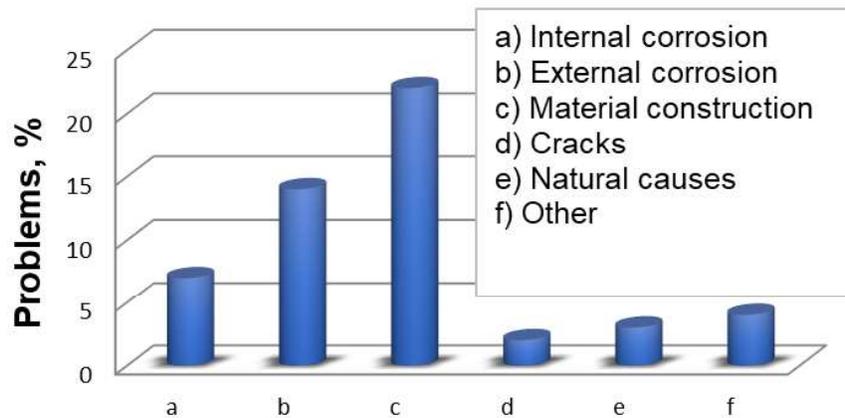


Figure 10. Statistical comparison of problems occurring in liquid pipelines [21].

The distribution of capital costs, annual operating costs and the contribution of costs to the cost of hydrogen supply, in particular, was assessed in [41] using the method of an integrated hydrogen supply system based on renewable energy sources (RES). In this case, a linear programming model is used. Different resources (wind turbine, photovoltaic panel and Stirling engine power systems) and hydrogen production technologies (water splitting using an alkaline electrolyzer and biomass gasifier) are used. This method allows to minimize the total annual costs.

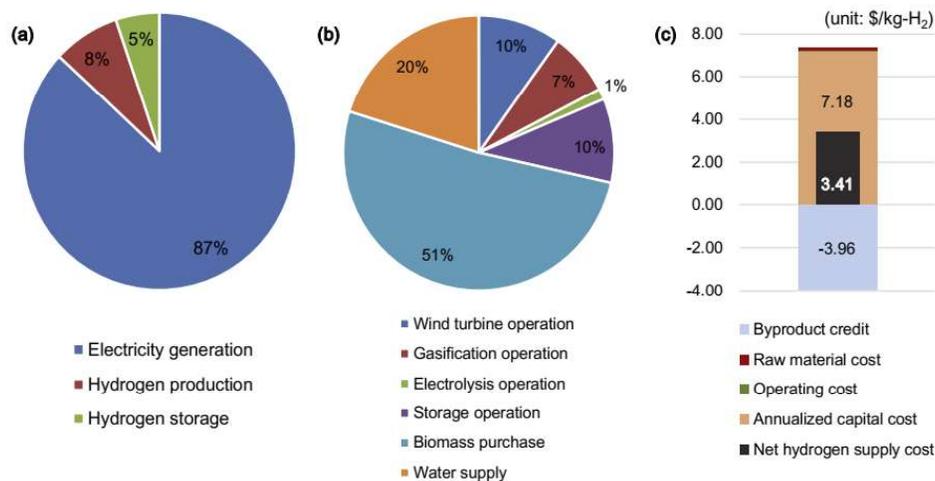


Figure 11. Distribution of capital costs (a), annual operating costs (b), and the contribution of costs to the cost of hydrogen supply of an integrated RES system (c) [41].

### 3. Conclusions

In modern descriptions of the HE mechanism in the M–H system (material, metal, alloy or steel), the explanations are moving from the microscale (HEDE) to the nanoscale (HELP, HESIV). However, there are some unresolved issues in the study of the HE mechanisms in the M–H system: (1) The quantitative relationship between the local hydrogen concentration and the nature of the interaction between the atoms of the M–H system during HE has not been experimentally or theoretically established for the HED mechanism. (2) The role of plastic deformation in the mechanism of HE formation in M–H is unclear, since the process of interaction of M–H atoms occurs on non-identical individual grains of the M crystal. To prove the HE mechanism, it is necessary to take into account the anisotropy of the crystal grains (e.g., Young's modulus, internal stresses) under the influence of external fields and stresses, leading to elastic and plastic deformations of the alloy. There is no answer to the question whether the plastic behavior of the alloy at the grain boundaries of M contributes to hydrogen embrittlement or whether the plastic behavior of the alloy itself is a result of HE? (3) Based on the microstructural evolution and surface fracture data of the alloys, NVC mechanisms and a synergistic HEDE+HELP model has been proposed, but have not yet been proven. (4) The existing HE mechanisms for M–H systems are only valid for certain materials in certain application areas. That is, a single HE mechanism for all materials has not yet been developed.

### Conflict of interest

The authors declare that they have no conflict of interest in relation to this research.

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